1 **Climate-driven paleolimnological change controls lacustrine mudstone**
2 **depositional process and organic matter accumulation: constraints from**
3 **lithofacies and geochemical studies in the Zhanhua Depression, eastern China**

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19 **ABSTRACT**

20 The influence of paleoclimate on the depositional process of lacustrine
21 mudstone and organic matter accumulation is important to both paleoclimate
22 reconstruction and hydrocarbon exploration. Here we study the lower third
23 Member (Es3L) of the Eocene Shahejie Formation in the Zhanhua Depression,
24 Bohai Bay Basin, eastern China in order to understand how paleoclimate
25 influenced depositional processes and organic matter accumulation of lacustrine
26 organic-rich mudstone. By combining detailed core descriptions and microscopic
27 observations, and high-resolution mineralogical and geochemical analyses, we
28 identified two major lithofacies, including massive calcareous mudstone and
29 laminated calcareous mudstone, from the Luo 69 drilling core. The sedimentologic
30 observations and changes of geochemical proxies, including detritus index,
31 $\text{Ln}(\text{Al}_2\text{O}_3/\text{Na}_2\text{O})$, B/Ga , and $\text{V}/(\text{V}+\text{Ni})$, suggest that the massive calcareous
32 mudstone was deposited in a small, shallow, salt lake that was dysoxic-anoxic, and
33 the paleoclimate in the lake catchment was cool and arid, and the laminated
34 calcareous mudstone was deposited in a large and deep stratified lake, which has
35 anoxic, highly saline bottom and oxic, less saline surface water, and the lake
36 catchment was more humid and warm. The dominant lithofacies changed from
37 laminated calcareous mudstone to massive calcareous mudstone in the studied core,
38 suggesting that the lake became shallower and smaller when the paleoclimate

39 became cooler and drier through time. Such a climate trend may be a response to
40 global cooling during the middle Eocene. The average total organic content (TOC)
41 in both lacustrine highstand and lowstand are comparable even though the lake
42 water chemistry and amount of terrigenous input are different. We infer that the
43 accumulation of organic matter within the lacustrine highstand was controlled by
44 the combination of primary productivity, carbonate production, and preservation in
45 anoxic bottom water, while accumulation within lacustrine lowstand was only
46 controlled by primary productivity.

47 **Keywords:** mudstone; lithofacies; paleoclimate; Shahejie Formation;
48 geochemistry; paleolimnology

49

50 **1. Introduction**

51 Organic-rich mudstones deposited in lacustrine basins can record continental
52 paleoclimate and paleoenvironment (e.g., Anderson and Dean, 1988; Bonk et al.,
53 2014; Aswasereelert et al., 2013; Chamberlain et al., 2013; Smith et al., 2015;
54 Wang et al., 2015b). Because lacustrine basins are relatively restricted, climate
55 changes in lake catchments may influence the balances of evaporation and
56 freshwater by driving perturbations in lake hydrological condition (Carroll and
57 Bohacs, 1999; Jiang et al., 2007; Kent-Corson et al., 2010; Chamberlain et al.,
58 2013; Vázquez-Urbez et al., 2013), and cause changes in lake water chemistry and

59 productivity, and terrigenous input, which in turn influences depositional processes
60 in lake basins (Aswasereelert et al., 2013; Doebbert et al., 2010; Graf et al., 2015;
61 Norsted et al., 2015; Smith et al., 2015). Despite the importance of lacustrine
62 mudstone, the extraction of useful paleoclimate and paleoenvironment information
63 from lacustrine mudstone is often hampered by tectonic influence on lake basin
64 development and depositional processes (e.g., Carroll and Bohacs, 1999; Pietras et
65 al., 2003; Hao et al., 2011), and lack of appropriate techniques for paleoclimate
66 and paleoenvironment reconstructions (Smith and Carroll, 2015; Smith et al.,
67 2015).

68 Organic-rich lacustrine mudstones are also important source rocks and
69 unconventional reservoirs (e.g., Talbot, 1988; Anadón et al., 1989; Genik, 1993;
70 Bruhn, 1999; Carroll and Bohacs, 2001; Rhodes et al., 2002; Bohacs et al., 2003;
71 Wang et al., 2011; Wang et al., 2015a; Ma et al., 2015). Factors controlling organic
72 matter accumulation include primary productivity in surface water (Pedersen and
73 Calvert, 1990; Caplan and Bustin, 1999), preservation controlled by water redox
74 condition and organic matter degradation (Demaison and Moore, 1980; Mort et al.,
75 2007), influx of clastic material and rate of carbonate precipitation, which may
76 dilute organic matter (e.g., Murphy et al., 2000; Sageman et al., 2003; Yan et al.,
77 2015), or the combination of these factors (e.g., Ma et al., 2016). Identifying the
78 primary factors influencing organic matter accumulation in a lake basin may guide

79 future hydrocarbon exploration in unconventional reservoirs.

80 Herein we present detailed sedimentologic description and high-resolution
81 geochemical analysis of lacustrine mudstone of the lower third Member (Es3L) of
82 the Eocene Shahejie Formation in the Zhanhua Depression, eastern China (Fig. 1).
83 The Member was deposited when the basin experienced extension and continuous
84 tectonic subsidence (Wang et al., 2005; Liu et al., 2015). Based on depositional
85 processes interpreted from sedimentologic observations, lake water chemistry and
86 terrigenous input reconstructed from geochemical proxies, we inferred the
87 paleoclimate in the lake drainage and its influence on depositional processes of the
88 lacustrine mudstone and lake water chemistry. Then we explored the relationship
89 between organic matter accumulation and paleoclimate in the lake drainage.

90 **2. Geological Setting**

91 As one of the most important petroleum basins in China, the Bohai Bay Basin
92 is a Cenozoic rift basin located in the eastern coast of China with an area of
93 approximately 200,000 km² (Fig. 1A) (Ye et al., 1985; Hsiao et al., 2004). This
94 region was in a back-arc basin during the Mesozoic and evolved into an
95 intracratonic rift basin during the Cenozoic (Hao et al., 2010). The basin was at a
96 synrift stage during the Paleogene with a series of grabens and half grabens
97 developed along major northwest- and northeast-trending fault sets (Fig. 2; Allen
98 et al., 1997; Huang and Pearson, 1999). These grabens and half grabens coalesced

99 to form the large Bohai Bay Basin, and the basin entered the postrift stage at the
100 end of the Oligocene (Hao et al., 2009; 2011; Tian et al., 2014).

101 The Bohai Bay Basin consists of seven sub-basins, namely the Linqing,
102 Jizhong, Huanghua, Jiyang, Bozhong, Liaodong Bay, and Liaohe sub-basins (Fig.
103 1A; Gong, 1997). The Zhanhua Depression is located in the northern part of the
104 Jiyang sub-basin (Fig. 1A). With an area of 3610 km², the depression has a
105 cumulative proved oil in place of 1.84×10^8 tons and a predicted oil in place of
106 1.68×10^8 tons (Song, 2011; Wang et al., 2015a), thus is one of the most
107 petroliferous depressions of the Bohai Bay Basin (Shi et al., 2005; Hao et al., 2010;
108 Wang et al., 2015a). The depression is bounded by the Chengjiazhuang uplift to the
109 south, Yihezhuang uplift to the north and northwest, Chengdong uplift to the north,
110 and the Gudao uplift to the east (Fig. 1B). Within the Zhanhua Depression,
111 northeast- to east-trending strike-slip, normal faults produced several low-relief
112 uplifts and half-grabens with gentle slopes in the south and steep slopes in the
113 north (Jiu et al., 2013). The subsidence of the basin was further complicated by
114 northwest- or near north-striking normal faults, which formed secondary local
115 uplifts and sags (Fig. 1 C).

116 The thick Cenozoic basin fills in the Zhanhua Depression are divided into the
117 synrift and post rift depositional sequences (e.g., Gong, 1997; Wang et al., 2005;
118 Hao et al., 2009; 2011). The synrift sequence unconformably overlies the

119 Mesozoic strata, and includes the Paleogene Kongdian, Shahejie, and Dongying
120 formations that were deposited primarily in lacustrine environments in localized
121 grabens and half grabens (Fig. 2). The postrift sequence includes the Neogene
122 Guantao, Minhuazhen, and Pinyuan formations that were deposited primarily in
123 fluvial environments when the entire basin experienced thermal subsidence (Fig.
124 2).

125 The early Eocene-early Oligocene Shahejie Formation is widely distributed in
126 grabens and half-grabens in the Zhanhua Depression, and is mostly absent on the
127 structural uplifts that separate these grabens (Shi et al., 2005; Wang et al., 2005).
128 The formation consists of interlayered sandstone, siltstone, oil shale, mudstone,
129 and evaporites that were deposited in fluvial-lacustrine-deltaic depositional
130 environments (Fig. 2). This formation is divided into the Es1, Es2, Es3 and Es4
131 members (Fig. 2). The Es1, Es2, and Es4 members are further divided into upper
132 and lower members, while the Es3 Member is divided into upper, middle and
133 lower members (Jiu et al., 2013; Wang et al., 2015a).

134 Our study target is the lower Es3 (Es3L) Member in the Zhanhua Depression,
135 which has significant petroleum source potential because the discovered
136 commercial shale oil flow wells are mainly concentrated in this member (Jiu et al.,
137 2013; Wang et al, 2015b). During the deposition of the Es3 Member, basin
138 subsidence rate generally exceeded sediment accumulation rate and a lacustrine

139 basin formed and became wider and deeper through time (Liu et al., 2015).
140 Streams originated from the bounding uplifts carried clastic sediments resulting in
141 turbidity flows in the lake basin (Shi et al., 2005; Wang et al., 2015a). The Es3L
142 Member is dominated by organic-rich oil shale, dark mudstone, and calcareous
143 mudstone that were deposited in lacustrine environment (Fig. 2) (Hu et al., 2001;
144 Zhang et al., 2012), and contains abundant aquatic organic matter (Li et al., 2006;
145 Wang et al., 2015a). The Es3L Member is conformably overlain by the middle Es3
146 (Es3M) Member, which consists of predominantly dark-gray lacustrine mudstone
147 intercalated with siltstone and sandstone formed as turbidites (Shi et al., 2005).
148 The upper Es3 (Es3U) Member is dominated by gray lacustrine mudstone,
149 interbedded with sandstone formed in fluvial-deltaic environments (Shi et al.,
150 2005).

151 **3. Materials and methods**

152 Our study focused on the Luo 69 Well, which is located in nearly the center of
153 the Bohai Bay Basin, where the influence of secondary faulting is negligible and
154 deposition of the organic-rich mudstone sequence of the Es3L Member is
155 complete. The core is 185.55 m long and has a very good recovery rate (94.2%).
156 A total of 44 standard thin sections, 373 mineralogy analysis, 207 total organic
157 content (TOC wt.%) analysis, 373 major and trace elements analysis, and 211
158 Rock-Eval pyrolysis were used for this study. Samples used for analyses were

159 freshly cut after removing weathered surface of the core.

160 Our sedimentological descriptions include the identification of fresh rock
161 color, grain size, sedimentary structures, degree of bioturbation, presence and
162 type of bioclasts, type and distribution of lenses, mineralogy of cements, presence
163 and relative abundance of fractures following the guidelines in Lazar et al. (2015).
164 The names of the mudstone lithofacies are modified by sedimentary structures
165 and composition (Lazar et al., 2015). Basic lithofacies descriptions are
166 summarized in Figure 3.

167 Mineral compositions were analyzed using X-ray diffraction (XRD). The
168 samples were ground to less than 40 μm using an agate mortar to thoroughly
169 disperse minerals. No chemical pre-treatment was employed and the powdered
170 samples were oven-dried at 40°C for two days prior to analysis. Powdered
171 samples were scanned using a D8 DISCOVER X-ray diffractometer with Cu-K α
172 radiation, automatic divergent and antiscatter slits, and a secondary graphite
173 monochromator with a scintillation counter. The generator settings were 40 kV
174 and 25 mA. Samples were scanned from 3° to 70° with a step size of 0.02°.
175 Computer analysis of diffractograms provides identification of minerals and
176 semi-quantitative results of the weight percentage of each mineral type.

177 Total organic carbon contents were analyzed using a LECO CS-600
178 carbon/sulfur analyzer with a precision of 0.5% at the Petroleum Geology

179 Research Center of the Geological Sciences Institute of the Shengli Oil Field,
180 SINOPEC.

181 The concentrations of elements Al, Na, B, Ga, V and Ni were determined
182 using an inductively coupled plasma-atomic emission spectrometer (ICP-AES) at
183 the Petroleum Geology Research Center of the Geological Sciences Institute of the
184 Shengli Oil Field, SINOPEC. The powdered samples were digested in Teflon
185 bombs using 10 ml mixture of HClO₄ (60%), HNO₃ (65%), and ultrapure H₂O
186 with a ratio of 6.5: 2.5: 1, and 10 ml HF (40%) before analysis. The detailed
187 digestion procedures and analytical precision and accuracy are described in
188 Nijenhuis et al. (1999). The analytical errors of our data are less than 5 %.

189 Rock-Eval Pyrolysis was conducted using Rock-Eval 6 analyzer under
190 standard operation conditions following Behar et al. (2001). The parameters
191 measured included the free hydrocarbons (S₁), the hydrocarbons cracked from
192 kerogen (S₂), and the temperature of maximum yield of pyrolysate (T_{max}).
193 Hydrogen index (HI) was calculated following Tissot and Welte (1984). The
194 uncertainty of the reported values is better than 5% based on replicate analyses of
195 Chinese National Standard GB/T 18602-2001.

196 **4. Geochemical Proxies**

197 Because quartz grains in the Luo 69 Well are principally detrital based on our
198 sedimentological description and interpretation, we applied a detritus index

199 (Table 1), calculated by summing quartz, clay minerals, and feldspar contents
200 (e.g., Keller and Pardo, 2004; Westermann et al., 2010; Stein et al., 2011; Bomou
201 et al., 2013; Montero-Serrano et al., 2015) to document changes in detrital influx.
202 High detritus index values indicate high input of terrigenous material from
203 continental sources, and/or decreased carbonate productivity and dissolution
204 (Bomou et al., 2013; Montero-Serrano et al., 2015).

205 Although the chemical index of alteration (CIA) is widely used to evaluate
206 the degree of chemical weathering and climate changes at lake catchment (e.g.
207 Minyuk et al., 2007; McHenry, 2009; Ma et al., 2015; Jiang et al., 2016), samples
208 which contain a lot carbonate, such as in this study, may suggest misleading
209 conclusions about degree of weathering if CIA is used (von Eynatten et al., 2003;
210 Montero-Serrano et al., 2015). Therefore, we used $\text{Ln}(\text{Al}_2\text{O}_3/\text{Na}_2\text{O})$ (Table 1) as a
211 weathering index similar to CIA to infer changes in the degree of chemical
212 weathering during deposition of the Es3L Member (von Eynatten et al., 2003;
213 Montero-Serrano et al., 2015; Jiang et al., 2016). Statistic comparison of
214 $\text{Ln}(\text{Al}_2\text{O}_3/\text{Na}_2\text{O})$ and CIA showed that $\text{Ln}(\text{Al}_2\text{O}_3/\text{Na}_2\text{O})$ can precisely reflect
215 weathering trend and has the additional advantage of avoiding uncertainties
216 concerning the correction for CaO associated with phosphate and carbonate
217 phases (von Eynatten et al., 2003). Relatively high $\text{Ln}(\text{Al}_2\text{O}_3/\text{Na}_2\text{O})$ values reflect
218 strong rainfall, enhanced chemical weathering and continental runoff under warm

219 and humid climate. Relatively low values of this parameter indicate the near
220 absence of chemical weathering, thereby reflecting cool and arid climate
221 (Montero-Serrano et al., 2015).

222 The ratio of B/Ga in sediments is commonly used as a proxy for paleosalinity
223 of lakes (e.g., Chen and Chen, 1997; Fu et al., 2015a; Wang et al., 2015b). The
224 B/Ga ratio of fresh water deposit is typically lower than 3, while a B/Ga ratio
225 between 3 and 5 indicates brackish water, and a ratio greater than 5 indicates
226 salty water. V/(V+Ni) ratio is reputed as a paleoredox index (e.g., Algeo and
227 Maynard, 2004; Tribovillard et al., 2006; Algeo and Ingall, 2007; Akinlua et al.,
228 2010; Yang et al., 2010; Fu et al., 2015b; Montero-Serrano et al., 2015). V/(V+Ni)
229 ratios lower than 0.46, 0.46-0.60, 0.54-0.82, and greater than 0.82 indicate oxic,
230 dysoxic, anoxic, and euxinic conditions in the water column, respectively (Hatch
231 and Leventhal, 1992).

232 **5. Results**

233 **5.1 Bulk mineralogical composition, TOC and geochemical characteristics**

234 The mudstone in the Es3L Member of the Zhanhua Depression contains
235 mostly clay minerals, quartz, calcite, and minor amounts of feldspar, dolomite, and
236 pyrite (Fig. 4). Siderite was present only in some samples. Due to generally low
237 amount (<5 wt.%), it is not discussed here. Calcite content (0-88 wt.%) decreases
238 generally toward the top of the core and inversely related to clay minerals content

239 (1-45 wt.%) (Fig. 4). Quartz content (3-45 wt.%) positively correlates with clay
240 minerals content (Figs. 4, 5). Dolomite content ranges from 0 to 26 wt.%, with an
241 average of 5 wt.%. Feldspar content is less than 9 wt.%, and pyrite content ranges
242 from 0 to 16 wt.%. Detrital index, and clay minerals and quartz contents are well
243 correlated (Fig. 5), and all steadily increase upward (Fig. 4).

244 The mudstone contains 1.0-9.3 wt.% TOC with an average of 3.0 wt.% and
245 TOC displays parabolic relations to both quartz and clay minerals contents (Fig. 6).
246 TOC is positively correlated with quartz and clay minerals contents when quartz
247 content is lower than 23 wt.% and clay minerals content is lower than 27 wt.%,
248 respectively. Otherwise, TOC is negatively correlated with quartz and clay
249 minerals contents. TOC is negatively correlated with calcite content (Fig. 4).

250 S_1 values range from 0.1 to 32.6 mg HC/g rock, with an average of 3.2 mg
251 HC/g rock. S_2 is a proxy for generative source potential of source rocks (Alias et
252 al., 2012; Shalaby et al., 2012; Zhao et al., 2014). S_2 values are in the range of
253 1.2-78.6 mg HC/g rock, with an average of 15.3 mg HC/g rock. HI values range
254 from 82 to 844 mg HC/g TOC with an average of 439 mg HC/g TOC (Fig. 7).

255 $\ln (Al_2O_3/Na_2O)$ of samples in the Es3L Member ranges from 2.29 to 3.64,
256 with an average of 2.99 (Fig. 7). The B/Ga ratios range from 3.23 to 12.66, and the
257 $V/(V+Ni)$ ratios range from 0.1 to 0.99, with an average of 0.67 (Fig. 7).

258 **5.2 Lithofacies**

259 Two lithofacies are present throughout the Es3L Member in the Luo 69 Well,
260 including massive calcareous mudstone and laminated calcareous mudstone (Fig.
261 3).

262 **5.2.1 Massive calcareous mudstone**

263 Light- to dark-brown massive calcareous mudstone occurs as beds of 0.03-7.4
264 m thick mainly in the upper part, and occasionally in the lower part of the Es3L
265 Member (Fig. 3). Bioturbation is absent from this lithofacies (Fig. 8A-D). The
266 grain size is dominantly medium and coarse mud. This lithofacies is mostly blocky
267 and homogeneous (Fig. 8A), and contains minor amount of uncrushed silt-sized
268 ostracod fragments (Fig. 8B, C) and pyrite (Fig. 8D), but abundant scattered,
269 silt-sized, angular to subrounded, monocrystalline quartz grains (Fig. 8B).
270 Carbonate is present as shell fragments and as cryptocrystalline cement in the
271 mudstone (Fig. 8B-D). This lithofacies has relatively high average clay mineral
272 and quartz contents and low average calcite content, which are 24 wt.%, 19 wt.%,
273 and 46 wt.%, respectively (Table 1). TOC content varies between 2.0 wt.% and 7.7
274 wt.%, with an average of 3.4 wt.%. This lithofacies also has relatively high detritus
275 index, and low $\text{Ln}(\text{Al}_2\text{O}_3/\text{Na}_2\text{O})$, B/Ga, and $\text{V}/(\text{V}+\text{Ni})$ average values, which are
276 45 wt.%, 2.81, 5.47, and 0.59, respectively (Table 1).

277 **5.2.2 Laminated calcareous mudstone**

278 Light-brown to dark laminated calcareous mudstone (Fig. 9 A-I) is the

279 dominant lithofacies in the Es3L Member (Fig. 3). The grain size varies from fine
280 to medium mud. The lamination is generally continuous, planar and parallel in
281 hand specimen, but continuous, wavy laminae and ripple cross-laminae are also
282 documented by microscopic observations (Fig. 9 A-D). The laminae are very thin
283 and generally range from less than 1 mm to a few millimeters thick in hand
284 specimen and 0.03-0.10 mm thick based on thin section observation (Fig. 9 A-D).
285 In the middle part of the Es3L Member, this lithofacies has some intervals
286 containing abundant carbonate laminae (Fig. 3). The carbonate laminae are
287 generally less than 0.5 cm thick, parallel and continuous or lenticular (Fig. 9 E).
288 Abundant bedding-parallel fractures are observed in the intervals, and most of
289 them are filled by sparry calcite (Fig. 9 E). The carbonate laminae and lenses also
290 contain fibrous calcite crystals perpendicular to bedding planes (Fig. 9 F). These
291 sparry calcite lenses have clear outlines, taper laterally, and mostly show irregular
292 edges (Fig. 9 F). Thin section observation found that oil migrated along the
293 fractures in the calcite-rich intervals (Fig. 9 G).

294 This lithofacies is predominantly composed of alternating clay-rich laminae
295 and cryptocrystalline calcite-rich laminae (Fig. 9 B-C). Organic matter present as
296 band or is star-like in the clay-rich laminae (Fig. 9 B). Horizontally transected
297 fishbone of a few millimeters long commonly presents in this lithofacies (Fig. 9 D).
298 The lithofacies also contains abundant, sometimes oriented ostracod fragments

299 (Fig. 9 H) and shows no sign of bioturbation. Most samples in the lithofacies
300 contain carbonaceous lenses with diameters up to a few millimeters and the lenses
301 are embedded in the planar laminae (Fig. 9 A, E). Some vegetation can be
302 observed in the hand specimen (Fig. 9B). Quartz grains are of silt size, and present
303 mostly as rounded monocrystalline grains and occasionally as subrounded grains
304 in the clay-rich laminae (Fig. 9 I). Carbonate presents as bioclasts,
305 cryptocrystalline cement, and microcrystalline grains. Quartz and clay minerals
306 account for 15 wt.% and 15 wt.% of the total mineral (Table 1), which are lower
307 than those of the massive calcareous mudstone. Calcite and pyrite contents are
308 higher than those of the massive calcareous mudstone. This lithofacies contains
309 minor amount of dolomite with an average of 5 wt.%, and dolomite presents as
310 microcrystalline grains in cryptocrystalline calcite-rich laminae. TOC content
311 varies between 1.0 wt.% and 7.5 wt.%, with an average of 3.3 wt.%. The
312 lithofacies has relatively high $\text{Ln}(\text{Al}_2\text{O}_3/\text{Na}_2\text{O})$, $\text{V}/(\text{V}+\text{Ni})$ and B/Ga values, which
313 are in average 3.08, 0.72, and 7.58, respectively (Table 1). The detritus index
314 values are lower than those of the massive calcareous mudstone.

315 **6 Discussion**

316 **6.1 Lithofacies and their sedimentary processes**

317 Our interpretations of depositional processes aimed at reconstructing the
318 environmental conditions and processes under which the Es3L Member was

319 deposited, based on the petrological, sedimentological, and geochemical
320 observations described above.

321 **6.1.1 Massive calcareous mudstone**

322 Homogeneous fabric, uncrushed ostracod fragments and lack of bioturbation
323 (Fig. 8 A-D) reflect original deposition under relatively quiet and poorly
324 oxygenated lacustrine setting (Bruhn, 1999). Dyni and Hawkins (1981) and Boyer
325 (1982) also suggested that homogenous muds deposit in relatively shallow parts of
326 lake above the chemocline. The lack of sedimentary structures and evidence for
327 traction currents (e.g. ripple cross-lamination) probably indicate negligible
328 influence of continental runoff during the deposition (Schieber and Southard, 2009;
329 Schieber and Yawar, 2009). Silt-sized angular to subrounded monocrystalline
330 quartz is generally from terrigenous sources (Zuffa et al., 1980; Underwood et al.,
331 2005; Liang et al., 2012). Given the lack of evidence for traction currents and
332 fluvial depositional process, the scattered angular to subrounded quartz grains (Fig.
333 8 B) were most likely of aeolian origin (Egenhoff and Fishman, 2013). The high
334 detrital index in this lithofacies (Figs. 4, 7) reflects low carbonate production in the
335 lake basin, rather than high terrigenous input. This is because the Es3L Member in
336 the Luo 69 Well has predominantly type I oil prone kerogen (Wang et al., 2015a),
337 and terrestrial organic matter should be expected if there was high terrigenous
338 input. The cryptocrystalline calcite was formed by photosynthesis in the

339 epilimnion during algal blooms (Thompson et al., 1997; Bonk et al., 2014).

340 The low B/Ga and V/(V+Ni) ratios (Table 1) suggest that this lithofacies was
341 deposited in saltwater column under highly dysoxic to anoxic conditions that were
342 hostile to benthic life (Hatch and Leventhal, 1992; Fu et al., 2015a; Wang et al.,
343 2015b). This inference is supported by the absence of bioturbation. The presence
344 of abundant detrital quartz grains of aeolian origin typically suggests that the lake
345 catchment was under arid paleoclimate (e.g. May et al., 1999; Stanley and De
346 Deckker, 2002). This interpretation is further supported by our geochemistry data.
347 Low $\text{Ln}(\text{Al}_2\text{O}_3/\text{Na}_2\text{O})$ value indicates the near absence of chemical weathering in
348 cool and arid climate (e.g., Bomou et al., 2013; Montero-Serrano et al., 2015).
349 Under cool and arid climate, decreased precipitation in lake catchment could
350 reduce the transport capacity of rivers and freshwater input, causing relatively
351 shallow lake level. Therefore, we consider that the massive calcareous mudstone
352 was deposited in a relatively small and shallow salt lake that was dysoxic-anoxic,
353 and the paleoclimate in the lake catchment was cool and arid.

354

355 **6.1.2 Laminated calcareous mudstone**

356 Mudstones with bedding-parallel veins of fibrous sparry calcite (Fig. 9 E-G)
357 were found in thermally mature and organic-rich stratigraphic sequences in many
358 sites around globe (e.g., Rodrigues et al., 2009; Cobbold et al., 2013; Zanella et al.,

2014; Zhang et al., 2016). It has been suggested that organic acids, a by-product of oil generation, promote dissolution of micritic carbonate in mudstone, and precipitation of diagenetic sparry calcite in spaces created by oil generation and migration (Zhang et al., 2016). Oil migration through fractures was observed in this study (Fig. 9 G), suggesting that the sparry calcite may be formed by deep diagenesis during hydrocarbon generation. The bedding-parallel fractures (Fig. 9 E) may be formed by an upward seepage force of fluid overpressure (Zhang et al., 2016). Because the carbonate laminae-rich intervals were influenced by deep diagenesis, their chemical compositions were excluded from this lithofacies when discussing water chemistry and terrigenous input.

Continuous, planar lamination (Fig. 9 A-C) suggests low-energy deposition condition (Abouelresh and Slatt, 2012). Continuous, wavy lamination and ripple cross-lamination (Fig. 9 B-D) imply sediment reworking by wave activity (Lazar et al., 2015). Ripple cross-lamination and orientation of ostracod shells (Fig. 9 D, H) also indicate traction currents (O'Brian, 1996; Schieber et al. 2007; Macquaker and Bohacs 2007; Macquaker et al., 2010). Alternations of clay-rich laminae and cryptocrystalline calcite-rich laminae (Fig. 9 B, C) typically reflect sedimentologic responses to seasonal forcing on water salinity, organic productivity, and runoff in lake catchment (e.g., Anderson and Dean, 1988; Lindqvist and Lee, 2009). Because calcite precipitates rapidly by photosynthesis in the epilimnion during

379 algal blooms in warm seasons (Thompson et al., 1997; Freytet and Verrecchia,
380 2002; Bonk et al., 2014), the carbonate-rich laminae were most likely deposited
381 during warm seasons when organic productivity was high. The clay-rich laminae
382 with abundant silt-sized, rounded, monocrystalline quartz grains (Fig. 9 C) may be
383 formed during relatively cold seasons, and the quartz grains and clay minerals
384 were transported by continental runoff (Freytet and Verrecchia, 2002). The
385 preferential presence of organic matter in the clay-rich laminae (Fig. 9 B) probably
386 results from adsorption of organic matter by clay minerals after algae deterioration
387 during cold seasons (cf. Li et al., 2014).

388 Although dolomite is often of diagenetic origin, authigenic dolomite formed
389 in anoxic and hypersaline environments with high organic matter content has been
390 previously documented (Cole and Picard, 1978; Vasconcelos and McKenzie, 1997;
391 Warthmann et al., 2000). Microcrystalline dolomite in this lithofacies was most
392 likely formed by bacterial sulfate reduction soon after deposition of organic matter
393 and calcium carbonate minerals (e.g. Wright and Wacey, 2005; Braissant et al.,
394 2007; Deng et al., 2010). Therefore, the presence of dolomite suggests that the lake
395 was stratified, and the bottom water was anoxic. The interpretation of saline
396 bottom water with anoxic condition is further supported by the relatively high
397 B/Ga and V(V+Ni) values (Table 1) (cf. Hatch and Leventhal, 1992). The presence
398 of fishbone implies that the surface water was oxic, and suitable for organic matter

399 production (e.g. Boyer, 1982; Fischer and Roberts, 1991; Bruner et al., 2015) (Fig.
400 9 D). Therefore, we suggest that the laminated calcareous mudstone was deposited
401 in stratified water column, which has anoxic, highly saline bottom water that
402 favors preservation of organic matter and oxic, less saline surface water that favors
403 bioproductivity (cf. Talbot, 1988; Huc et al., 1990; Carroll and Bohacs, 2001;
404 Jagniecki and Lowenstein, 2015). This interpretation is consistent with a previous
405 study that suggested well-laminated calcareous mudstone in the Shulu Sag, which
406 is a small Eocene half-graben lake basin to the west of the Zhanhua Depression,
407 deposited in thermally stratified lake with anoxic bottom water (Jiang et al., 2007).

408 The relatively high $\text{Ln}(\text{Al}_2\text{O}_3/\text{Na}_2\text{O})$ values (Table 1) suggest that this
409 lithofacies was deposited during times of enhanced chemical weathering (e.g.
410 Bomou et al., 2013; Montero-Serrano et al., 2015). Elevated precipitation in humid
411 climate promotes chemical weathering and provides fresh water rich in nutrients to
412 lake, resulting in a progressive rise in lake level (Bohacs et al., 2003; Jiang et al.,
413 2007; Smith et al., 2014). Therefore, the overall climate during the deposition of
414 this lithofacies was generally more humid compared to the climate during the
415 deposition of the massive calcareous mudstone. This lithofacies has relatively low
416 detritus index suggesting low clastic sediments input to the lake, which could
417 result from dense vegetation acting as traps and baffles in the drainage area (e.g.
418 Einsele and Hinderer, 1998; Bohacs et al., 2003; Jiang et al., 2007; Smith et al.,

2014). Increased inflow of fresh surface water causes ectogenic meromixis and promotes lake stratification (Boyer, 1982; Carroll and Bohacs, 2001), which in turn may have helped preservation of organic matter, leading to high TOC (Table 1). Rich nutrients also promote algal blooms and calcite production (Bohacs et al., 2003; Jiang et al., 2007). Given the inferred high continental runoff and humid climate, we suggest laminated calcareous mudstone was deposited in a deep and large lake.

6.2 Climate control on depositional processes

During the deposition of the Es3L mudstone, the dominant lithofacies changed from the laminated calcareous mudstone to the massive calcareous mudstone (Fig. 10), suggesting a change in depositional environment from a relatively large and deep lake (highstand) associated with warm and humid climate to a small and shallow lake (lowstand) associated with cool and arid climate. Previous chronostratigraphy studies of the Es3L Member in the Zhanhua Depression and Jiyang sub-basin, based on micropaleotologic and palynologic data, and paleomagnetostratigraphy (Wang and Lin, 2012; Wang et al., 2015a; Feng et al., 2016), suggested that the member was deposited during 42-45 Ma. This is a period when global climate experienced gradual cooling based on deep-sea oxygen isotope record (Zachos et al., 2001, 2008). Therefore, we infer that the change to more arid and cooler climate recorded in the Es3L Member of the Zhanhua

439 Depression followed global climate change during the middle Eocene.

440 Based on the interpreted depositional processes, lake water chemistry and
441 redox conditions, and inferred paleoclimate in lake catchment, we develop a
442 depositional model to explain how paleoclimate influenced lacustrine mudstone
443 depositional process by changing lake water chemistry and redox conditions (Fig.
444 10). During the lacustrine highstand, warm and humid climate may have
445 influenced lacustrine depositional processes in two ways. Firstly, high annual
446 precipitation, or diminished evaporation, or both lead to broad expansion of the
447 lake by increasing discharge of rivers and input of freshwater, which possibly
448 caused traction currents and deposition of ripples and oriented ostracod shells near
449 the lake center (Anderson and Dean, 1988; Carroll et al., 2010). Secondly, humid
450 and warm climate promoted vegetative stabilization in lake drainage area, which
451 acted as traps and baffles to clastic sediments, thus reduced terrigenous input but
452 enhanced solute transport (e.g., Einsele and Hinderer, 1998; Bohacs et al., 2003;
453 Smith et al., 2014). Elevated precipitation may have caused lake water
454 stratification by promoting ectogenic meromixis and hampering mixing between
455 saline bottom water and fresh surface water (Jiang et al., 2007; Smith et al., 2008),
456 which explains the coexistence of relatively high $V/(V+Ni)$ and B/Ga values and
457 abundant fishbone fossils. This interpretation is similar with a previous study that
458 suggested laminated calcareous mudstone in the Eocene Green River Formation in

459 Wyoming, U.S.A. was deposited during lake highstands (Rhodes et al., 2002).

460 During the lacustrine lowstand, cool and arid climate may have lowered lake
461 level by reducing fresh water input and the capacity of river transporting detritus,
462 and enhancing evaporation. Only aeolian transport brought large grains into the
463 lake center. Because early and middle Eocene climate was generally warm (Zachos
464 et al., 2001, 2008), aridity may be the major factor that influenced the lake water
465 chemistry. Strong evaporation and limited fresh water input in arid climate may
466 have caused salty water column, supported by moderate $V/(V+Ni)$ and B/Ga
467 values (Table 1).

468 **6.3 Mechanisms of TOC enrichment in the Es3L Member**

469 Organic matter accumulation in a lake basin is determined by primary
470 productivity in surface water, preservation of organic matter, and sedimentation
471 rate (Demaision and Moore, 1980; Pedersen and Calvert, 1990; Caplan and Bustin,
472 1999; Mort et al., 2007). Although the water redox conditions, primary
473 productivity, influx of clastic material, and carbonate production are different
474 during the inferred lacustrine highstand and lowstand, TOC contents in the two
475 mudstone lithofacies are comparable and high (Table 1). Here we develop a model
476 to explain the mechanism of organic matter enrichment in the Es3L Member in the
477 Zhanhua Depression.

478 Within the lacustrine highstand, TOC is strongly correlated with HI ($r = 0.70$),

479 but moderately correlated with detritus index ($r = 0.57$) and carbonate content ($r =$
480 -0.57) (Fig. 11 A, C, E). Detritus index and carbonate content reflect rates of
481 terrigenous input from lake catchment and carbonate production in the lake basin.
482 The correlation relationships suggest that during the lacustrine highstand, TOC was
483 modestly promoted by terrigenous input, but diluted by carbonate production.
484 Under warm and humid climate, enhanced precipitation may have promoted
485 chemical weathering in lake catchment, and carried fresh water inflow and
486 dissolved inorganic carbon, nitrate and other nutrients into the lake to cause high
487 primary productivity (Jiang et al., 2007; Smith et al., 2008; Chetel et al., 2011).
488 Elevated ectogenic meromixis further may have led to more stable stratification of
489 lake water, which, in turn, could have been extremely favorable for the production
490 and preservation of organic matter (e.g., Bohacs et al., 2000). HI values reflect
491 variations in primary productivity and degree of water column anoxia (Demaison
492 and Moore, 1980; Curiale and Stout, 1993). Because the $V/(V+Ni)$ ratio for
493 mudstone deposited in the highstand was relatively high and stable, the variation of
494 HI values must reflect the variations of lake primary productivity (Fig. 7). The
495 relatively good correlation between HI and TOC (Fig. 11A) supports that primary
496 productivity in surface water and preservation in anoxic bottom water influenced
497 organic matter accumulation. In summary, organic matter accumulation in the
498 lacustrine highstand was influenced by surface water primary productivity,

499 preservation in anoxic bottom water, and carbonate production rate (Fig. 12A).

500 Within the lacustrine lowstand, TOC is well correlated with HI ($r = 0.66$), but
501 not correlated with detritus index ($r = 0.14$) and carbonate content ($r = -0.02$) (Fig.
502 11 B, D, F). The lack of correlation between TOC and detritus index and carbonate
503 content suggests that sedimentation rate was not important in organic matter
504 sequestration in the lacustrine lowstand. Because the $V/(V+Ni)$ ratio for the
505 mudstone deposited in the lowstand was relatively low (Table 1), reflecting less
506 anoxic bottom water condition than during the lacustrine highstand, the moderately
507 high HI values during this period must be caused by high lake primary productivity.
508 The good correlation between HI and TOC (Fig. 11 B) further indicates that
509 organic matter accumulation was primarily controlled by lake productivity during
510 the lacustrine lowstand (Fig. 12 B). Although it seems unlikely that productivity
511 was high in salty lake developed under arid and cool climate, it has been suggested
512 previously that salty water column favors concentrated nutrient and alkaline
513 conditions with abundant CO_3^{2-} ions (Fuhrmann et al., 2004; Hao et al., 2011),
514 which may have led to moderately high primary productivity during the lacustrine
515 lowstand.

516 Although it is generally thought that increased silicate content is usually
517 accompanied by high nutrient supply, which promotes primary productivity (Littke
518 et al., 1991; Rippen et al., 2013; Sachse et al., 2012), in our study, TOC decreases

519 when the contents of clay minerals and quartz are higher than ~25 wt.% (Fig. 6).
520 Therefore, increased silicate content does not always favor organic matter
521 accumulation, and excessive clastic dilution could dilute organic carbon burial flux
522 (Sageman et al., 2003). Our study showing that quartz=23 wt.% and clay=27 wt.%
523 (Fig. 6) are probably the threshold of clastic dilution which reduces organic matter
524 concentration in the Es3L Member of the Zhanhua Depression, which is important
525 for unconventional reservoir exploration in the Bohai Bay Basin in eastern China.

526

527 **7. Conclusions**

528 The Es3L Member of the Eocene Shahejie Formation in the Zhanhua
529 Depression of the Bohai Bay Basin, eastern China was studied to understand
530 paleoclimatic controls on lacustrine depositional processes, lake water chemistry,
531 and organic matter accumulation. We identified two major lithofacies that were
532 deposited in different lake hydrologic conditions and paleoclimate. The massive
533 calcareous mudstone was deposited in a relatively small and shallow lake with
534 salty water column and dysoxic-anoxic condition, and the lake catchment had cool
535 and arid climate. The laminated calcareous mudstone was deposited in a relatively
536 deep and large stratified lake with anoxic bottom water and oxic surface water, and
537 the lake catchment had warm and humid climate.

538 We proposed a paleolimnologic model to explain the evolution of the

539 depositional environment of the Es3L Member. The lake changed from lacustrine
540 highstand to lowstand through time, associated with systematic variations in
541 precipitation intensity, lake hydrologic condition, lake level, and amount of
542 terrigenous input, which, in turn, were strongly influenced by paleoclimate. The
543 change of dominant lithofacies from laminated calcareous mudstone to massive
544 calcareous mudstone suggests that the paleoclimate became more arid and cooler
545 during the middle Eocene, possibly in response to global climate change.

546 The interpreted depositional processes and geochemical proxies are used to
547 understand the reasons of organic matter accumulation. The accumulation of
548 organic matter during the lacustrine highstand of the Es3L Member was controlled
549 by the combination of primary productivity, preservation, terrigenous input, and
550 carbonate production, while the accumulation of organic matter during the
551 lacustrine lowstand was only controlled by primary productivity. Although
552 increased silicate content in carbonate-dominated depositional system is usually
553 accompanied by increased nutrient supply and promotes organic matter production,
554 excessive silicate content could dilute organic matter, such as in some depositional
555 intervals of the Es3L Member.

556

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991 **Figure captions**

992 **Figure 1.** (A) The Bohai Bay Basin and its tectonic units. Sub-basin classification
 993 is from Allen et al. (1997). The location of the Zhuanhua Depression in the
 994 northern part of the Jiyang sub-basin is highlighted with a box. (B) Map showing
 995 the structural features within and surrounding the Zhanhua Depression, and the
 996 location of the Luo 69 Well. (C) S-N cross-section of the Zhanhua Depression. See
 997 B for location of the cross-section. Modified from Jiu et al. (2013). Q: Quaternary,
 998 Nm: Minghuazhen Formation (Pliocene), Ng: Guantao Formation (Miocene), Ed:

999 Dongying Formation (Oligocene), Es1: the first Member of the Shahejie Formation
1000 (Oligocene), Es2: the second Member of the Shahejie Formation
1001 (Eocene-Oligocene), Es3: the third Member of the Shahejie Formation
1002 (Eocene-Oligocene), Es4U: the upper fourth Member of the Shahejie Formation
1003 (Eocene), Es4L: the lower fourth Member of the Shahejie Formation (Eocene), Ek:
1004 Kongdian Formation (Eocene).

1005

1006 **Figure 2.** Generalized Cenozoic stratigraphy of the Zhanhua Depression. Modified
1007 from Wang and Lin (2012), Jiu et al. (2013), Liu et al. (2015), Wang et al. (2015a)
1008 and Feng et al. (2016).

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1010 **Figure 3.** Schematic summary of core description of the Luo 69 Well in the
1011 Zhanhua Depression, eastern China, illustrating vertical variability in lithofacies,
1012 texture, bedding, sedimentary structures, type and distribution of lenses.

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1014 **Figure 4.** Lithofacies log, mineral content, and TOC of the Es3L Member in the
1015 Luo 69 Well.

1016

1017 **Figure 5.** Linear correlations detritus index vs. clay and quartz contents, and linear
1018 correlation between quartz and clay contents of the Es3L Member in the Luo 69

1019 Well.

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1021 **Figure 6.** Correlations TOC vs. quartz and clay contents of the Es3L Member in
1022 the Luo 69 Well.

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1024 **Figure 7.** Plots showing general trends of detritus index, $\text{Ln}(\text{Al}_2\text{O}_3/\text{Na}_2\text{O})$, B/Ga,
1025 $\text{V}/(\text{V}+\text{Ni})$, HI and TOC.

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1027 **Figure 8.** Characteristic sedimentologic observations of the massive calcareous
1028 mudstone. (A) Core photograph at 2981.63–2982.23 m showing calcareous
1029 mudstone with massive bedding. (B) Thin-section photograph at 2949.91 m
1030 showing angular to subrounded, silt-sized quartz (yellow arrows) and uncrushed
1031 ostracod fragments (white arrows). (C) Thin-section photograph at 2992.84 m
1032 showing that ostracod bodies are replaced by sparry calcite (white arrows). (D)
1033 Thin-section photograph at 2989.23 m showing that carbonate presents as
1034 cryptocrystalline cement.

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1036 **Figure 9.** Characteristic sedimentologic observations of the laminated calcareous
1037 mudstone. (A) Core photograph at 3105.13–3105.33 m showing laminated
1038 calcareous mudstone with continuous, planar lamination and carbonaceous lenses

1039 with a few millimeters in diameter. (B) Thin-section photograph at 3119.65 m
1040 showing continuous, parallel to wavy-parallel, alternating clay-rich laminae (white
1041 arrow) and calcite-rich laminae (yellow arrow). Note: organic matter was enriched
1042 in the clay-rich laminae (red arrow). (C) Thin-section photograph at 3095.41 m
1043 showing rounded silt-sized quartz grains preferentially accumulated in the
1044 clay-rich laminae (white arrow). (D) Thin-section photograph at 3112.15 m
1045 showing ripple cross-lamination and fishbone (arrows). (E) Core photograph at
1046 3059.26–3059.50 m showing sparry calcite in continuous or lenticular laminae
1047 (white color). Note: carbonaceous lens (white arrows) and fractures (yellow arrows)
1048 present. (F) Thin-section photograph at 3061.60 m showing abundant
1049 bedding-parallel sparry calcite lens. (G) Thin-section photograph at 3059.85 m
1050 showing oil migrated along a fracture (arrow) in a carbonate laminae-rich interval
1051 where sparry calcite laminae is present. (H) Thin-section photograph at 2978.63 m
1052 showing oriented ostracod fragments. (I) Core photograph at 3072.62 m showing
1053 plant fossils (white arrows).

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1055 **Figure 10.** Paleolimnologic models showing lake hydrologic conditions and
1056 depositional processes in lacustrine highstand (A) and lacustrine lowstand (B).

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1058 **Figure 11.** Linear correlations between TOC and HI values in lacustrine highstand

1059 (A) and lowstand (B); linear correlations between TOC and detritus index in
1060 lacustrine highstand (C) and lowstand (D); and linear correlations between TOC and
1061 carbonate content in lacustrine highstand (E) and lowstand (F).

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1063 **Figure 12.** Schematic illustration showing factors controlling accumulation of
1064 organic matter in the lacustrine highstand (A) and lowstand (B).

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Lithofacies		Massive calcareous mudstone	Laminated calcareous mudstone
Quantity of samples		97	276
Quartz (wt.%)	Min.–Max.	14-45	3-35
	Average	19	15
Feldspar (wt.%)	Min.–Max.	0-9	0-4
	Average	2	1
Calcite (wt.%)	Min.–Max.	0-63	0-88
	Average	46	61
Dolomite (wt.%)	Min.–Max.	2-14	0-26
	Average	5	5
Clay mineral (wt.%)	Min.–Max.	12-40	1-45
	Average	24	15
Pyrite (wt.%)	Min.–Max.	0-9	1-16
	Average	3	4
Quantity of samples		54	153
TOC (wt.%)	Min.–Max.	2.0-7.7	1.0-7.5
	Average	3.4	3.3
Quantity of samples		97	276
Detritus index (wt.%)	Min.–Max.	28-80	4-79
	Average	45	31
Ln(Al ₂ O ₃ /Na ₂ O)	Min.–Max.	2.29-3.37	2.43-3.64
	Average	2.81	3.08
V/(V+Ni)	Min.–Max.	0.10-0.81	0.47-0.99
	Average	0.59	0.72
Quantity of samples		10	28
B/Ga	Min.–Max.	3.23-7.96	4.64-12.66
	Average	5.47	7.58

1079 Table 1. Mineral composition (wt.%), total organic carbon (TOC wt.%) and
1080 geochemical indexes of the two lithofacies in the Es3L mudstone succession.

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Cool and arid climate

Warm and humid climate

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