



# Early Jurassic climate and atmospheric CO<sub>2</sub> concentration in the Sichuan paleobasin, Southwest China

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#### Abstract:

Unlike marine archives, terrestrial sediments show more complicated and dynamic environment and climate. This work presents new results of climate-sensitive sediment observation and carbon-oxygen isotope analyses of lacustrine and pedogenic carbonates for the Early Jurassic Ziliujing Formation from the grand Sichuan paleobasin (GSB), Southwest China. Lithofacies analysis indicates calcisols were widespread in riverine and flood plain facies. Climate-sensitive sediments and carbon-oxygen isotopes with palynofloral assemblages manifest that an overall (semi-) arid climate dominated the GSB; and that it became drier through time, accompanied by occasional evaporites in the Toarcian. This climate pattern is similar with the arid climate in Colorado Plateau, western America, but distinct from the relatively warm-humid climate in North China and northern Gondwanaland in Southern Hemisphere. The estimated Early Jurassic atmospheric  $CO_2$  concentration ( $pCO_2$ ) from carbon isotopes of pedogenic carbonates shows a range of 980-2610 ppmV (~ 3.5-10 times the pre-industrial value) with a mean 1660 ppmV. Three phases of  $pCO_2$  (the Sinemurian 1500-2000 ppmV, the Pliensbachian 1000-1500 ppmV, and the early Toarcian 1094-2610 ppmV) and two events of rapid falling  $pCO_2$  by ~1000-1300 ppmV are observed, illustrating the  $pCO_2$  perturbation in the Early Jurassic. The pattern and associated rapid falling events of  $pCO_2$  are compatible with the excursions of stable isotopes and seawater temperature from the coeval marine sediments, consistent with a positive feedback of climate to  $pCO_2$  through the Early Jurassic.





#### 1. Introduction

The Jurassic was a typical greenhouse period with global paleotemperatures possibly 5-10 °C higher than present based on 27 28 modelling results (e.g., Chandler et al., 1992; Rees et al., 1999; Sellwood and Valdes, 2008). The Early Jurassic epoch was 29 an interval of extreme environmental change, during which climate events were recorded by highly enhanced organic carbon 30 burial, multiple isotopic anomalies, clay mineral composition, oceanic anoxic regime, global sea-level change, vegetation 31 turnover, and mass extinction (e.g. Price, 1999; Hesselbo et al., 2000; Dera et al., 2009; Jenkyns, 2010; Korte and Hesselbo, 32 2011; Riding et al., 2013; Arabas et al., 2017) as well as pCO<sub>2</sub> perturbation (e.g., Beerling and Royer, 2002; McElwain et al., 33 2005; Berner, 2006; Retallack, 2001a, 2009; Steinthorsdottir and Vajda, 2015). Recently, examples of rapid transitions from cold, or even glacial, climates to super greenhouse events are documented in some intervals of the Jurassic (e.g., van de 34 35 Schootbrugge et al., 2005; Suan et al., 2010; Gómez et al., 2016; Arabas et al., 2017). The study of these deep-time climate events may serve as analogues for present-day and future environmental transitions (Hesselbo et al., 2013). 36 37 Though the climate events in the Early Jurassic epoch are largely based on the marine sedimentary and geochemical records, 38 data from the terrestrial realm provide important details of environmental change (e.g., Hesselbo et al., 2000; Suan et al., 2010; Jenkyns, 2010; Philippe et al., 2017). Terrestrial proxies, such as flora (e.g., Riding et al., 2013; Deng et al., 2017; 39 Philippe et al., 2017; Ros-Franch et al., 2019), vegetation (Pole, 2009), and geochemistry (e.g., Riding et al., 2013; Kenny, 40 41 2015; Tramoy et al., 2016) have begun to provide important information of the Mesozoic-Cenzoic climate and 42 environmental changes on continents. Particularly, a negative feedback in the global exogenic carbon cycle, from carbon isotopes of lacustrine organic matter, has been hypothesized to account for the Toarcian oceanic anoxic event (Xu et al., 43 44 2017), opening a new avenue to link marine and terrestrial climate in the Early Jurassic. However, few relatively continuous 45 terrestrial climate records and coupled environmental changes have been documented for the Early Jurassic. There are several large Triassic-Jurassic terrestrial basins in West China, providing a great opportunity to recover the coeval 46 terrestrial environment and climate. The Sichuan Basin has a relatively complete and continuous sedimentary sequence of 47 the Upper Triassic-Paleogene (e.g., SBGM, 1991, 1997; Wang et al., 2010). Correspondingly, the sedimentary archive could 48 49 play a key role in the global Early Jurassic correlation of the marine and terrestrial climate. In this work, we present new 50 results of field investigation, lithofacies and paleosol recognition, carbon-oxygen isotope analyses of both lacustrine and 51 pedogenic carbonates, and  $pCO_2$  estimates in the Early Juassic terrestrial Sichuan paleobasin, and we discuss the relationship 52 of terrestrial climatic change to that of the marine counterpart.

# 2. Geological setting and stratigraphy

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Southwest China, including the provinces of Yunnan, Sichuan, Chongqing, and Guizhou, had been the main part of the upper Yangtze Plate since the Proterozoic, possibly since the Neoarchean. With the amalgamation of the Cathaysia and





56 Yangtze plates, it became the western South China plate or cratonic basin in the Neoproterozoic (Sinian), and marine 57 Neoproterozoic through the Middle Triassic strata is well preserved. With the Indosinian orogeny, new foreland basins were 58 formed since the Late Triassic (e.g., He and Liao, 1985; Li et al., 2003), which record the Mesozoic and Cenozoic evolution 59 of tectonics, environment, and climate in Southwest China. The Mesozoic Sichuan paleobasin was confined by the Longmenshan thrust belt in the northwest, the Micangshan-Dabashan 60 arcuate thrust belt in the northeast (Fig. 1), and the northern hilly topography boundary of the Yunnan-Guizhou plateau in 61 62 the south and east. It was mainly developed during the Late Triassic-Jurassic and includes provincial areas of eastern Sichuan, entire Chongqing, northern Guizhou, western Hubei, and northwestern Hunan. This Triassic-Jurassic Sichuan 63 foreland basin was much larger than the present Sichuan Basin in the eastern Sichuan province. We estimate the size of 64 Sichuan paleobasin is roughly 480,000 km² by the lithofacies paleogeography (Fig. 1. Ma et al., 2009; Li and He, 2014), and 65 66 suggest naming this the grand Sichuan paleobasin (GSB). The Mesozoic terrestrial sediments accumulated up to ~9 km (Guo et al., 1996) in the GSB; and the Jurassic part can be as 67 much as 3-3.5 km thick (SBGM, 1991). Two types of Lower Jurassic deposits have been distinguished (Table 1): the 68 69 Baitianba Formation (Fm) in the north and the Ziliujing Fm (e.g., SBGM, 1991; Wang et al., 2010) in the south (over 90% of 70 the basin). 71 The Baitianba Fm was deposited unconformably on the Upper Triassic Xujiahe Fm and is overlain conformably by the 72 Middle Jurassic Xintiangou Fm / Qianfuyan Fm (Table 1). It is mainly composed of gravish shales and sandstones with coal 73 layers and massive conglomerates. Abundant plant fossils, sporopollens, conchostracans, bivalves, and gastropods indicate it 74 is the Early Jurassic (SBGM, 1991, 1997). Sporopollen assemblages of the Hettangian-Sinemurian age were found in the 75 lower part (Zhang and Meng, 1987) and the Pliensbachian-Toarcian assemblages were reported in the upper part (Wang et 76 77 The Ziliujing Fm is composed of variegated and reddish mudrocks (some shales) intercalated with sandstones, siltstones, and bioclastic limestones as well as marlstones, conformably or unconformably overlying the Xujiahe Fm or Luqiao Fm and 78 79 conformably underlying the Xintiangou Fm (SBGM, 1997. Table 1). It has been dated as the Early Jurassic by fossil 80 assemblages of dinosaurs, bivalves, ostracods, conchostracans, and plants, within which the dinosaur fauna can be well 81 correlated to the Lufeng Fauna in central Yunnan (e.g., Dong, 1984; SBGM, 1991, 1997; Peng, 2009). This formation is 82 subdivided as five parts in an ascending order: the Qijiang, Zhenzhuchong, Dongyuemiao, Ma'anshan, and Da'anzhai members (SBGM, 1997. Table 1). Of them, the former two are sometimes combined the Zhenzhuchong Fm (e.g., SBGM, 83 84 1991; Wang et al., 2010). The Da'anzhai Member is characterized by dark gray to black shales and bioclastic limestones with a southward increase of 85 reddish mudrocks (SBGM, 1991, 1997; Wang et al., 2010), which has been mainly regarded the sediment in a grand Sichuan 86 87 paleolake (e.g., Ma et al., 2009; Li and He, 2014). Ostracod assemblagse indicate it is the late Early Jurassic (e.g., Wei, 1982;





88 Wang et al., 2010). A Re-Os isochron age 180.3 ± 3.2 Ma combined with the organic carbon isotope excursion indicates that 89 the lower Da'anzhai Member corresponds to the Toarcian Oceanic Anoxic event (T-OAE. Xu et al., 2017), consistent with 90 the assigned Toarcian age. 91 The Ma'anshan Member is comprised of violet-red mudrocks with a few greyish, greenish thin-bedded fine sandstones and 92 siltstones, in which floral fossils are common (Li and Meng, 2003). The Dongyuemiao Member consists of greenish and reddish mudrocks and siltstones with greyish bioclastic limestone and marlstone, of which abundant bivalve and plant fossils 93 94 were reported from eastern Sichuan and Chongqing (Li and Meng, 2003; Meng et al., 2003; Wang et al., 2010). The Zhenzhuchong Member is dominated by violet red mudrocks/shales intercalated with thinned sandstones and / or siltstones 95 and numerous plant fossils of the Early Jurassic affinity (e.g., Duan and Chen, 1982; Ye et al., 1986). Taken together, fossil 96 associations suggest that the three members were deposited in the middle-late Early Jurassic. The age limitation of the 97 98 overlying Da'anzhai Member and the correlation to the Lufeng dinosaur fauna places these members in the Sinemurian -99 Pliensbachian, and the Zhengzhuchong and Dongyuemiao Fms are temporally suggested the Sinemurian age (Table 1). 100 The Qijiang Member is composed of quartz arenite interbedded/intercalated with dark shales. Coal seams can be often seen 101 in the middle of the Qijiang Member. This member mainly occurs in the central part of the GSB. It is likely the earliest

Jurassic, possibly Hettangian age, but plant fossils cannot precisely indicate the age (Wang et al., 2010).

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# 3. Materials and methods

Observation and description for sedimentary facies analysis were executed on six outcrop sections (Locations A1 to A4, A6 and A7, Fig. 1). Published description for other sections (Locations A5, A8, and A9, Fig. 1) is integrated into our observations. Details of microscopic examination of sedimentary rocks and sedimentary facies analysis which are the underpinning of climate analysis are attached as the supplementary data Note S1. Below are chiefly introduced materials and methods of climate-sensitive sediment observation, carbon-oxygen isotope analyses, and estimate of  $pCO_2$ .

# 3.1. Observation of climate-sensitive sediments

work.

Dolomites and gupsum are relatively easy to recognize in both field and under microscope. We distinguish dolomites from limetstones following Tucker (2003) and Flügel (2004). As Flügel (2004) stated, field distinctions of limestone and dolomite can also be made although detailed differentiation of carbonate rocks is best performed in the laboratory. The basic method that we use to examine dolomites is: limestone will fizz strongly and dolomite will show little or no reaction when add dilute 10 % hydrochloric acid on carbonate (Flügel, 2004); and limestone will stain pink to mauve but dolomite will be unstained

Climate-sensitive sediments are mainly the dolomites, gupsum, and paleosols, which are used to analyze the climate in this





118 (e.g., Tucker, 2003; Flügel, 2004) when Alizarin-red S in weak HCI is added on freash outcrop or coverslip-free thin section. 119 Gypsum is recognizable by properties of low Mohs hardness (2) and transparence to translucence. In field, we also recognize 120 gypsum by particular structures such as chichen-wire cage, gypsum pseudomorph, and cluster of (0.5-1 cm) pore. 121 There are multiple classifications of paleosols (e.g., Wright, 1992; Mack et al., 1993; Retallack, 2001b; Imbellone, 2011), mostly based on the US Soil Taxonomy. We recognized paleosols in the field based on color, structures, horizonation, root 122 traces, and textures, and followed the general classification paleosols by Mack et al. (1993) and Retallack (2001b). In this 123 124 paper, paleosols were described following the procedures of the Soil Survey Manual and classified according to Soil Survey 125 Staff (1998). Within the measured and observed sections, paleosol profiles were mainly identified from the two main locations/sections 126 A4 and A6 (Figs. S1 and S2, and Table S1). Horizonation, BK horizon thickness, boundary condition, structures, trace 127 128 fossils, rootlets, carbonate accumulations (calcretes), etc. were observed and described (Table S1). Paleosols interpreted in other cited sections (Fig. 1) rely on the diescription of lithology, streucture, and calcrete in the original references. 129 Based upon a modification of the Retallack (1998) categorization of paleosol maturity, the relative paleosol development 130 131 (maturity) was assigned. 132 3.2. Analyses of carbon-oxygen isotopes 133 Ten lacustrine carbonate samples were collected to analyse carbon-oxygen isotopes from the Da'anzhai Member of the 134 Ziliujing Fm at the Shaping section, Ya'an (Location A4. Fig. S1 and Table S3). Twenty-six pedogenic carbonate samples 135 were selected to measure carbon-oxygen isotopes from thirty-one paleosol horizons of the Ziliujing Fm at the same section (Fig. S1 and Table S4). Two or three microdrilling powder samples (columns 7 and 8 in Table S4) were taken from the same 136 137 individual calcrete for stable isotope analysis, and then a mean value for each calcrete sample was calculated (columns 9 and 10 in Table S4). 138 139 Before drilling, diagenetic fabrics of samples were studied under a microscope. Each sample was cut and prepared as thin sections for diagenetic diagnosis, and cathodoluminescence (CL) images (Fig. 2) were used to examine if the calcites were 140 evenly precipitated. Only the areas that were a uniform (often orange) luminescene (Fig. 2) were microsampled for isotope 141 analyses. Cracks, veins, and vug spaces in concretion samples were found to be filled by multidirectional growth of spar 142 143 crystals. These crack spar fills were avoided when microsampling as they were interpreted as recrystallization and replacement diagenetic phases. Microsampling of lacustrine carbonate samples focused on avoiding spar and sampling only 144 micrites. Powder samples were obtained by dentist drilling machine (aiguille diameter  $\phi$ =1-2 mm). 145 Isotopic analyses were conducted on  $0.3 \sim 0.5$  mg powder samples. Powder samples were dried in an oven at  $60^{\circ}$ C for 10146 hours before being moved to the instrument. Carbon dioxide for isotopic analysis was released using orthophosphoric acid at 147 148 70°C and analysed on-line in a DELTA-Plus xp (CF-IRMS) mass spectrometer at the State Key Laboratory for Mineral

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- 149 Deposits Research, Nanjing University. The precision of the measurements was regularly checked with a Chinese national
- carbonate standard (GBW04405) and the international standard (NBS19) and the standard deviation of  $\delta^{13}$ C was  $\pm 0.1\%$  over
- the period of analysis. Calibration to the international PeeDee Belemnite (PDB) scale was performed using NBS19 and
- 152 NBS18 standards.

## 3.3. Calculation of atmospheric CO<sub>2</sub> concentration

- 154 There are multiple methods to reconstruct the concentration of atmospheric carbon dioxide, i.e., pCO<sub>2</sub>, in deep time. It can be
- determined from the  $\delta^{13}$ C value of pedogenic carbonate using a paleobarometer model (Cerling, 1999), and the
- reconstruction of pCO<sub>2</sub> has been applied in the climate study of the Mesozoic time (e.g., Ekart et al., 1999; Nordt et al., 2003;
- 157 Myers et al., 2012; Li et al., 2014; Zhang et al., 2018).
- The Cerling (1999) equation was used to calculate the  $pCO_2$  using the carbon isotope of pedogenic carbonates as below:
- $C_a = S_{(z)}(\delta^{13}C_s 1.0044\delta^{13}C_r 4.4)/(\delta^{13}C_a \delta^{13}C_s)$
- where  $C_a$  is  $pCO_2$ ;  $\delta^{13}C_s$ ,  $\delta^{13}C_r$ ,  $\delta^{13}C_a$  are the isotopic compositions (‰) of soil  $CO_2$ , soil-respired  $CO_2$ , and atmospheric  $CO_2$ ,
- 161 respectively; and S(z) is the CO2 contributed by soil respiration (ppmV). Details of parameter usage and selection for the
- $pCO_2$  calculation are in the supplementary data Note S2.

# 163 **4. Results**

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- Based on the investigation of cross-sections (locations A1-A4, and A6-A7. Fig. 1), we have classified six sedimentary facies
- units in the Ziliujing Fm. They are alluvial fan, fluvial river, flood plain, lake, lake-delta, and swamp facies. Details of
- description and interpretation are in the supplementary data Note S1. Below are results of climate-sensitive sediment
- observation, stable isotope analyses, and  $pCO_2$  calculation.

# 4.1. Climate-sensitive sediments

- 169 Field observation combined with published calcrete materials shows that paleosols widely occur in the Lower Jurassic
- 170 Ziliujing Fm of the GSB (Figs. 1, 3, and 4). A total of 32 paleosols were observed and described at the Shaping section,
- Ya'an, and five paleosols were found at the Tanba section, Hechuan (Table S1).
- Most of paleosols are reddish (GSA Munsell Rock-Color 5R 2/2, 5R 3/4, 5R 4/2) and brownish (10R 3/4, 10R 5/4) (Fig. 3
- 173 and Table S1). Peds of paleosols are mainly angular and subangular, and a few are prismatic and platy. Slickensides are
- 174 common. Mottles (Fig. 3a), rootlets /rhizoliths (Fig. 3c), and burrows sometimes occur with strong leaching structures (Fig.
- 3a). Occasionally mudcracks are associated with the aforementioned structures (Fig. 3d).
- 176 All paleosols are calcic with more or less calcretes in Bk horizons. The thickness of Bk horizons is mainly 30-50 cm and
- 50-100 cm, and partly 100-170 cm (Table S1). Calcretes are generally ginger-like, ellipsoid, subglobular, and irregular in

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- shape (Fig. 3b and 3e) and nodules are 1-3 cm even up to 8-15 cm (paleosols J1z-10-01 and J1z-12-01) in size (Fig. 3e).
- 179 Calcrete is often less than 0.5-1% in an individual paleosol horizon, but a few can be up to 3-5% (paleosol J1z-3-01. Fig. 3b)
- 180 even 10% (paleosols J1z-5-02 and 18HC-10).
- 181 Based on the description of the paleosols described above, all are defined as relatively mature calcisols (Mack et al., 1993), a
- 182 kind of aridisol (Soil Survey Staff, 1998; Retallack, 2001b). The original lithofacies were chiefly argillaceous and silty
- 183 (split-fan) overbank, interchannel, and flood plain deposits (Figs. S1 and S2). Some formed landshare of the paleo-lakeshore.
- Dolomites were found at seven loactions in central and southern GSB (Figs. 1, 4, and Table S2), which are to some degree
- an indicative of arid/evapoatre climate. The dolomites chiefly occur in the Toracian Da'anzhai Member and a few in the
- 186 Sinemurian-Plienbachian Dongyuemiao and Ma'anshan members (Fig. 4). They are often massive whitish (Figs. 3f and S3e)
- and micritic (Figs. S4b and S4d), likely indicating a syndepositional origin.
- 188 Gypsum is only recorded in two loactions (Figs. 1, 4, and Table S2). One is located at Zigong (Location A5. SBG, 1980a).
- 189 The other lies at Hechuan (Location A6), which can be idientifed by chicken-wire cage structure and is associated with
- 190 micriditic dolomites (Fig. 3f).

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## 4.2. Carbon-oxygen isotope values

- $\delta^{13}$ C values of lacustrine carbonate samples range from -2.02% to -4.07% and  $\delta^{18}$ O values range from -9.91% to -12.28%
- 193 (Table S3 and Fig. 5). A distinct increasing trend of both carbon and oxygen isotope ratios can be detected from lower to
- upper horizons across a 40 m stratal interval of the lower Da'anzhai Member (Fig. 6).
- 195 Pedogenic carbonate samples have δ<sup>13</sup>C values from -3.52‰ to -8.10‰, which fall in the tyical stable isotope range for
- 196 pedogenic carbonates. Values of -6‰ to -8.0‰ characterize the sequence of the Zhenzhuchong Member and main
- 197 Ma'anshan Member, with an abrupt increase to -5.5% to -3.5% at the top of Ma'anshan Member (samples J1z-16-01 and
- 198 J1z-18-01. Fig. 6).  $\delta^{18}$ O values are mainly from -11.3% to -13.10% in the interval of the Zhenzhuchong Member and
- 199 Ma'anshan Member.  $\delta^{18}$ O follows  $\delta^{13}$ C with a sudden increase to -5.5% at the top of the Ma'anshan Member (Fig. 6). Large
- and frequent variations of both carbon and oxygen isotope ratios can be observed in the lower Da'anzhai Member (Fig. 6 and
- 201 Table S4).

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# 4.3. CO<sub>2</sub> concentrations

- 203 pCO<sub>2</sub> values of the Early Jurassic paleosols vary when different parameters are selected for calculation.
- 204 If  $S_{(z)}$ =2500 ppmV and  $\delta^{13}C_a$ =-6.5% (constant preindustrial atmosphere),  $pCO_2$  values range between ~1140 ppmV and
- 205 ~3460 ppmV with a mean of 1870 ppmV (column 15 in Table S4); and when  $S_{(2)}$ =2500 ppmV and  $\delta^{13}C_a$ =( $\delta^{13}C_r$ +18.67)/1.1,
- 206 pCO₂ values change between ~1230 ppmV and ~3260 ppmV with a mean of 2070 ppmV (column 16 in Table S4).





If  $S_{(z)}$ =2000 ppmV and  $\delta^{13}C_s$ =-8.98+ $\delta^{13}C_c$  are used,  $pCO_2$  values are ~940-2530 ppmV with the mean 1600 ppmV (column 207 17 in Table S4); and if  $S_{(2)}$ =2000 ppmV and  $\delta^{13}C_s = (\delta^{13}C_c + 1000) / ((11.98 - 0.12 * 25) / 1000 + 1) - 1000$  are adopted,  $pCO_2$ 208 209 values become ~980 ppmV to ~2610 ppmV with the mean 1660 ppmV (column 18 in Table S4). Details of the different 210 parameters and pCO<sub>2</sub> results can be seen in Table S4. Results further show that pCO<sub>2</sub> values at S<sub>(z)</sub>=2500 ppmV are larger than at S<sub>(z)</sub>=2000 ppmV, and the discrepancy of the 211 highest pCO2 is ~ 1000 (3640-2610) ppmV, but that of the lowest value is ~300 (1230-930) ppmV and that of the mean 212 213 value is  $\sim 370 (2070-1600)$  ppmV. In addition, when  $S_{(2)}$  is the same, the pCO<sub>2</sub> values are close even if other parameters are different (comp. between columns 15 and 16, 17 and 18 in Table S4, and Fig. 6). 214 However, the trend of  $pCO_2$  over the epoch is quite similar using different values of  $S_{(z)}$  and other parameters (Fig. 6). We 215 chose S(z)=2000 ppmV (column 18 in Table S4) to illustrate the nature of the Early Jurassic  $pCO_2$  estimated from calcisols 216 217 in the GSB. pCO<sub>2</sub> values mostly range between 980 ppmV and 2610 ppmV, and the mean 1660 ppmV is ~6 times the 275 ppmV. Most of 218 the pCO<sub>2</sub> values are 1000-2000 ppmV with the mean 1580 ppmV in the Zhenzhuchong and Ma'anshan members, ~3.5-7.5 219 220 times the pre-industrial pCO<sub>2</sub> value. 221 5. Discussion Results show that the depositional environment and paleoclimate in the Early Jurassic were distinctly different from those in 222 223 the Late Triassic in Southwest China. As a whole, the climate became dry and  $pCO_2$  varied in three phases through the Early 224 Jurassic. 225 Sedimentary facies analysis indicates two lithofacies cycles were developed and calcisols were largely spread in the Lower Jurassic Ziliujing Fm in the GSB, Southwest China. The first cycle is the riverine and flood plain lithofacies of the Qijiang 226 227 Membe and Zhenzhuchong Member succeeded by the lacustrine facies of the Dongyuemiao Member, and the second is the 228 flood plain and river facies with swamp lithofacies of the Ma'anshan Member followed by the lacustrine facies of the 229 Da'anzhai Member. We interpret the two packages to reflect two major lake stages (for details refer to supplementary data 230 Note S1). 231 With the change of depositional environments, paleoclimate and pCO<sub>2</sub> changed, as reflected by climate-sensitive facies and 232 stable isotope analyses. 233 5.1. Paleoclimate variation During the Late Triassic, Southwest China was warm-hot and humid in a tropical and subtropical zone, as demonstrted by 234 palynoflora, coals, and perennial riverine and lacustrine lithofacies in the Xujiahe Fm (e.g., Huang, 1995; Xu et al., 2015; Li 235 et al., 2016; Yang et al., 2019), and a distinct transfer of climate took palce in the Early Jurassic manifested by 236



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climate-senstive sediments and stable isotopes of the Ziliujing Fm in GSB.

#### 1) The Hettangian Age

By the Hettangian time (the Qijiang Member), a warm-humid climate followed the Late Triassic in the GSB. The limited sedimentary records are mainly mature quartz sandstones and siltstones with coals and siderite concretions (Fig. 7), indicating a stable tectonic setting and warm-humid climate in the eastern and southern GSB. In the northern margin, the climate was similar, because multiple coal layers occur in the lower Baitianba Fm and the hosted alluvial fan system (Figs. 7 and S6) is characterized by moderate-good roundness and sorting of gravels with sandy fillings (Fig. S3a. e.g., Liu et al., 2016; Qian et al., 2016; and this work). In the Newark basin of eastern America, climate-sensitive sediments such as nodules of carbonate and gypsum (pseudomorph) as well as mudcrack in mudflat facies indicate an arid climate in the fifth cycle of the Hettangian (>199 Ma. Kent et al., 2017) Passaic Fm (Smoot and Olsen, 1994). More widespread, the eolian Navajo Sandstone, dated as Hettangian-Sinemurian (200-195 Ma. Parrish et al., 2019), indicate an arid climate in Colorado Plateau. Obvieously, the arid climate in western America was different from that in the GSB at the time.

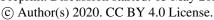
## 2) The Sinemurian Age

The early Sinemurian Zhenzhuchong Member is of riverine and flood plain facies with lacustrine facies, in which the lithology is dominated by violet-red mudrocks with few thin greyish, greenish fine sandstones and siltstones. The reddish color of rocks may indicate a change of climate even if there is little difference in the appearance of reddish color sediment in the western and central basin. That is, the reddish rocks developed through the whole member in the western margin (Location A4. Fig. 6), but it started in the middle member in the central basin (Location A6. Fig. S2). With the red color mudrocks, a kind of climate-sensitive pedogenesis is recognized from the flood plain facies. Multiple calcisol horizons were observed at the Shaping section, Ya'an (Location A4. Figs. 1, 4, and 7), within which a strong leaching calcisol horizon can be found (Fig. S3c). Calcisols were also interpreted with the description of abundant calcretes at sections of Dafang (Location A8. Zhang et al., 2016), Tianzhu (Location A9. Li and Chen, 2010), and Weiyuan (Location A10. SBG, 1980a), respectively. Calcisols indicate that a (semi-) arid climate at least began to replace the previous humid climate in western and southern margins of the basin (Figs.1, 4, and 7 and Table S2). This climate change, indicated from paleosols, is consistent with the climatic signal from floral fossils (e.g., Huang, 2001; Wang et al., 2010), suggesting a decrease in humidity and an increase in temperature across the interval, compared to that in the Hettangian Qijiang Member and Late Triassic Xujiahe Fm. However, the climate was not distinct in humidity and temperatue in the northern GSB without proxies of sediments and flora even though alluvial fan and lacustrine delta facies are common in the middle of the Baitianba Fm (Fig. S6. e.g., Qian et al., 2016). No climate-sensitive sediments are documented in the late Sinemurian Dongyuemiao Member from previous studies, in which it is characterized by lacustrine limestones. However, similar to the Zhenzhuchong Member, reddish mudrocks and





269 reported at Dafang (Location A8. Zhang et al., 2016), Tianzhu (Location A9. Li and Chen, 2010), and Yunyang (Location 270 A15. Meng et al., 2005) in the central and southern GSB (Figs 4 and 7). The probable calcisols indicate the (semi-) arid 271 climate may have interrupted the long-term warm and (semi-) humid climate interpreted based on flora in the Early Jurassic 272 (e.g., Meng et al., 1997; Li and Meng, 2003). This interpretation of (semi-) arid punctuation is also supported by the floral changes (Meng et al., 1997; Li and Meng, 2003) and geochemistry of mudrocks (Guo et al., 2017). 273 274 Few records of coeval terrestrial climate are known from other continents or regions in the literature. A report occurs in 275 eastern England, where the co-occurrence of the acmes of thermophilic pollens Classopollis classoides and Liasidium variabile indicates the warm-humid climate in the late Sinemurian (Riding et al., 2013). The Whitmore Point Member of the 276 Moenave Fm deposited in dryland lakes (Tanner and Lucas, 2008) and the upper part of eolian Navajo Sandstone (Blakey et 277 al., 1988) could represent the coevally similar climate in Colorado Plateau although relatively cool (~9 to 18 °C) continental 278 279 climate was inferred from oxygen and hydrogen isotope composition of chert precipitated in interdune, freshwater lakes in the Navajo Sandstone (Kenny, 2015). 280 3) The Pliensbachian Age 281 282 The Ma'anshan Member is likely the Pliensbachian, though age information is lacking. In comparison to the previous member, the Ma'anshan Member diaplays a prominent change in the distribution extent of red color sediment and 283 284 pedogenesis. The reddish sediments extend through the entire member (comp. Figs. 6 and S2) and can be observed across the 285 GSB. Calcisols are documented in both the western and central GSB (Figs. 6, 7, S1, and S2). Ten calcisol horizons were recognized at the Shaping section, Ya'an (Figs. 6 and S1), and strong leaching structure and mudcrack are seen in Bed H8 of 286 the Tanba section, Hechuan (Fig. 3a and 3d). Other more abundant calcretes within terrestrial red mudrocks were widely 287 288 described at the Gaoxian section of Dafang (Location A8. Zhang et al., 2016), the Hulukou section of Weiyuan (Location A10. SBG, 1980a), the Geyaoguan section of Gulin (Location A13. SBG, 1976), the Taiyuan section of Fengdu (Location 289 290 A16. SBG, 1975), and the Yaxi section of Zunyi (Location A17. Yang, 2015). We interpret these calcretes were formed by pedogenesis. The widespread distribution of redbeds and calcisols (Figs. 4 and 7) implies a (semi-) arid climate had been 291 292 intensified in the GSB during the Pliensbachian age. 293 Plant and sporopollen fossils also indicate a change to drier climate in the Pliensbachian. With comparison to the 294 Zhenzhuchong and Dongyuemiao Members, much fewer plant fossils were reported in this member (e.g., Meng and Chen, 295 1997; Wang et al, 2010), likely implying a rapid climatic change. The Pliensbachian-Toarcian sporopollen assemblages are dominated by classical sporomorph genera (Dictyophyllidites- Cyathidites- Classopollis), in which the dry-type gymnosperm 296 297 spore Classopollis is more prevalent than in the Hettangian-Sinemurian (Zhang and Meng, 1987), also indicating the 298 intensification of arid climate. 299 Similar dry temperate / subtropical climate was verified by the upland coniferous forest in Qaidam Basin, Northwest China 300 (Wang et al., 2005). However, at the same time, it was the probably coolest / most humid climate in South Kazakhstan,





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303 (e.g., Bromley, 1992) indicate similar arid climate in Colorado Plateau, western America. 304 4) The Toarcian Age In spite the fact that the Da'anzhai Member was deposited in the largest lacustrine transgression period (details see Appedix 305 1), climate-sensitive facies and stable isotoic geochemistry indicate that aridification could be the most intensive in the late 306 307 Early Jurassic in the GSB. Redbeds with abundant calcretes are well developed in the Da'anzhai Member (Figs. 4 and 7). Four calcisols horizons in the 308 Shaping section, Ya'an (Figs. 6 and S1) and the leaching structure (Bed H13) in the Tanba section, Hechuan (Fig. 3c), were 309 observed. Calcretes at sections of Dafang (Location A8. Zhang et al., 2016), Nanxi (Location A11. SBG 1980a), Gongxian 310 311 (Location A12. Liang et al., 2006), and Yunyang (Location A15. Meng et al., 2005), also record the occurrence of calcisols. 312 The widespread occurance of calcisols reveals that subaerial exposure of sediments often interrupted the lake environment, illustrating dynamic lake level fluctuations and aridification. 313 314 In addition to redbeds and calcisols, gypsum and micritic dolomites (SBG, 1980a; Mo and Yu, 1987; Peng, 2009; and this work) were reported in the western and southern GSB (Figs. 1, 4, and 7). It is plausible that gypsum and dolomites indicate 315 arid climate type. Thoguh dolomites have been in dispute for the significance of climate due to great deal of diagenetic 316 317 dolomites in deep time, a high abundance of dolomite was interpreted to form during greenhouse periods, characterized by warm climates, probably reflecting favourable conditions for evaporite deposition and dolomitization via hypersaline reflux 318 (Warren, 2000). Dolomites are aslo thought the results of interplay of climate and sea-level / base-level change (e.g., 319 320 Newport et al., 2017) or are interacted with climatic regimes (Vandeginste et al., 2012). So, the widespread micritic dolomites in the Da'anzhai Fm, associated with gypsum (Fig. 3f), can serve the determination of climate and suggest an arid 321 322 climate. Gypsum occasionally occurs at Maliuping of Hechuan (Fig. 3f) and Wujiaba of Zigong (SBG, 1980a), implying a 323 short-term evaporitic climate in the central GSB. 324 Carbon and oxygen isotopes of lacustrine carbonates further support the aird climate in the Toarcian age in the GSB. In general, -9.0% to -3.0% of  $\delta^{13}$ C and  $\delta^{18}$ O values represent a range of normal river-lake and groundwater carbonates 325 (Alonso-Zarza, 2003). Therefore, the mainly positive δ<sup>13</sup>C values 0 to 2 ‰ (Fig. 5) from Hechuan (Wang et al., 2006) 326 indicate the lakes were brackish or even saline, and the relatively heavy negative  $\delta^{13}$ C values -1% to -3.5 % (Fig. 5) from 327 Zigong (Wang et al. 2006) and Ya'an (this work) denote low depletions of 13C during calcite/aragonite precipitation and 328 329 mean that the lakes were possibly brackish. In other hand, lightly negative  $\delta^{18}O$  values -5% to -12 % (Fig. 5) dominate the lacustrine carbonates, suggesting that closed lacustrine, palustrine and pond systems formed in a regional arid-semiarid 330 331 climate with significant evaporation relative to precipitation. The covariance of  $\delta^{13}$ C and  $\delta^{18}$ O is also a criterion to distinguish closed or open lakes (e.g., Talbot, 1990; Li and Ku, 1997). 332

central Asia (Tramoy et al., 2016). These discrepancies might corroborate the unstable and heterogeneous climate in the

mid-latitude area of North Hemisphere in the Pliensbachian. In other hand, interdune playa mudstones of the Kayenta Fm





That is, high  $\delta^{18}$ O and low  $\delta^{13}$ C values will be produced in relatively low temperature lake water when the covariation is 333 negative; high values of both  $\delta^{18}$ O and  $\delta^{13}$ C will be produced in high-temperature meteoric water and indicate increased 334 evaporation when the covariation is positive. Pronounced positive covariances (R2=0.44-0.96) between carbon and oxygen 335 336 isotopes (Fig. 5) indicate a typical arid-semiarid pattern of lakes in the central and western GSB. The Da'anzhai Member has the same palynofloral assemblage with the Ma'anshan Member, in which the dry-type 337 gymnosperm spore Classopollis is much more than in previous strata (e.g., Zhang and Meng, 1987; Wang et al., 2010), 338 339 supporting the aridification indicated by climate-sensitive sediments and stable isotope ratios of lacustrine carbonates 340 aforementioned. Coastal Cheirolepidiacean (gymnosperm) forests indicate (temperate to subtropical) warm-humid climate punctuated by 341 locally dry and/or arid events in the Toarcian in Qaidam Basin, Northwest China (Wang et al., 2005). In Inner Mongolia of 342 343 North China, the thermophilous plants such as the dipteridaceous fern Hausmannia, bennettitales Ptilophyllum, display similar warm and humid climate interrupted by hot and even arid conditions in a short intervals of the Toarcian (Deng et al., 344 345 2017). The warm-wet climate was also indicated by assemblages of sporomorph and vegetation in the late Early Jurassic in 346 Jurong of Jiangsu, Lower Yangtze area (Huang et al., 2000). In South Kazakhstan, central Asia, paleoflora and  $\delta^2$ H values suggest slightly less humid and warmer conditions starting from the early Toarcian (Tramoy et al., 2016). 347 348 In summary, climate-sensitive sediments, carbon-oxygen isotope values and covariance, and palynoflora, together indicate 349 that an overall (semi-) arid climate dominated the GSB during the Early Jurassic, possibly accompanied by occasional evaporitic climate. Relatively abundant calcisols suggest that the GSB was in a subtropical arid zone based on the 350 paleoclimatic zonation model of paleosols (Mack and James, 1994) during the middle-late Early Jurassic. Through the Early 351 352 Jurassic, this (semi-) arid climate in GSB is thoroughly comparable with the simultaneous arid climate recorded in dryland lacustrine and eolian facies in Colorado Plateau, western America (e.g., Blakey et al., 1988; Bromley, 1992; Tanner and 353 354 Lucas, 2008; Parrish et al., 2017), but distinct from the relatively warm-humid climate indicated by sedimentological and floral characteristics in North China (e.g., Wang et al., 2005, Deng et al., 2017) and in the northern margin of Gondwanaland, 355 356 relatively high latitudes of Southern Hemisphere (Jansson et al., 2008; Pole, 2009).

#### 5.2. pCO<sub>2</sub> perturbations and events

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Pedogenic carbonates found in various continental settings precipitate in direct contact with soil atmosphere and bed rock and hold a meaningful signature of past climate (Alonso-Zarza and Tanner, 2006). Ancient  $pCO_2$  has been estimated by carbon isotope of pedogenic carbonates using the empirical (Cerling, 1991) and optimized (Ekart et al., 1999) formula. This paleosol method has roughly been applying in the Phanerozoic  $pCO_2$  estimate (e.g., Cerling, 1991; Ekart et al., 1999; Retallack, 2001a) with >10 Myr interval of age resolution. There are few high age resolution  $pCO_2$  reconstructions for the Early Jurassic. The focus on  $pCO_2$  estimates has on the event horizons, such as the transition of the Triassic to Jurassic (e.g.,



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ray flux, biota, volcanic eruption, and so on.



Tanner et al., 2001; Schaller et al., 2011). Herein we present the  $pCO_2$  estimate in ~1.0 Myr age resolution of the Early 365 Jurassic (Figs. 6 and 8c). 5.2.1. pCO<sub>2</sub> perturbation 366 367 Results of model estimates show that the pCO<sub>2</sub> values range 980-2610 ppmV with a mean 1660 ppmV in the Early Jurassic except for the Hettangian and can be divided into three intervals (Figs. 6 and 8c): phase I, stable 1500-2000 (mean ~1700) 368 ppmV in the Zhenzhuchong and Dongyuemiao Members (Sinemurian age); phase II, main 1000-1500 (mean ~ 1300) ppmV 369 370 in the Ma'anshan Members (Pliensbachian age); and phase III, great fluctuation 1094-2610 (mean ~1980) ppmV in the lower 371 Da'anzhai Member (early Toarcian age). 372 The evolution and level of pCO2 estimated by carbon isotope ratios of the pedogenic carbonates from the GSB are roughly comparable with the global composite based on the plant stomata method (data of the composite curve see Table S6), but 373 374 difficult to compare to the global composite pCO<sub>2</sub> based on paleosols (Fig. 8c. Suchecki et al., 1988; Cerling, 1991; Ekart et al., 1999), which may be attributed to the shortage of global data and large age uncertainties (Table S5 and S6). 375 376 On the other hand, the swing of the  $pCO_2$  has a similar pattern to coeval seawater temperature through the Early Jurassic 377 although there are some discrepancies in pace (comp. Fig. 8b and 8c). That is, the relatively high pCO<sub>2</sub> 1500-2000 ppmV 378 approximately corresponds to the relatively high seawater mean temperature -2°C to +2°C in the Sinemurian age (Fig. 8b), 379 low pCO<sub>2</sub> 1000-1500 ppmV to low seawater mean temperature -5°C to -2°C in the Pliensbachian age (Fig. 8b), and quick 380 rising pCO<sub>2</sub> of 1200 ppmV to  $\sim$ 2500 ppmV to the rapidly increased seawater temperature of -4°C to +4°C in the late 381 Pliensbachian-early Toarcian (Fig. 8b). The pCO2 record and the carbon isotope of the marine carbonates are also somewhat 382 comparable (comp. Fig. 8a and 8c). It has been disputed whether climate change was resulted from pCO<sub>2</sub> perturbation in the Phanerozoic (e.g., Veizer et al., 383 2000; Crowley and Berner, 2001; Royer, 2006). For instance, the pCO<sub>2</sub> has a strong control over global temperatures for 384 385 much of the Phanerozoic (e.g., Crowley and Berner, 2001; Royer, 2006; Price et al., 2013; Mills et al., 2019), but a decoupling of CO<sub>2</sub> and temperature has also been suggested (e.g., Veizer et al., 2000; Dera et al., 2011; Schaller et al., 2011; 386 Kashiwagi, 2016). The pattern of the Early Jurassic pCO<sub>2</sub> reconstructed from the carbon isotope of pedogenic carbonates in 387 GSB, Southwest China, supports the coupling relationship of CO<sub>2</sub>-temperature at a ~1.0 Myr resolution scale. Even so, 388 389 models of the coupling and decoupling of CO<sub>2</sub>-temperature have to consider: 1), age order of CO<sub>2</sub>-temperature relevance, i.e. they should be related in the same age (long term or short term) hierarchy; 2) precise age constrain of individual CO2 and 390 391 temperature data; 3) methods of CO<sub>2</sub> and temperature estimates, depending on precondition, presumptions, parameters, 392 uncertainty, sample diagenesis, etc.; 4) controls or influences of key factors such ice sheet, tectonic, paleogeography, cosmic





#### 5.2.2. Rapid pCO<sub>2</sub> falling events

The recovered Early Jurassic  $pCO_2$  curve reveals two rapid falling events (Fig. 6 and 8c). The first event ( $1E_{CO2}$ ) shows a 395 396 quick drop from ~2370 ppmV (sample J1z-08-01 at depth 84.7 m) to 1350 ppmV (sample J1z-10-02 at depth 94.4 m) near 397 the boundary of the Dongyuemiao and Ma'anshan Members (Fig. 6), or to 1075 ppmV (sample J1z-11-02 at depth 111.7 m), 398 which took place in the early Pliensbachian ( $\sim$ 190.4-189.9/189.1 Ma. Fig. 8c). The extent of the rapid falling  $pCO_2$  is 399 ~1000-1300 ppmV in 9.7-17.0 m. In other words, ~1000 ppmV drop could be accomplished within ~0.5-1.0 Myr based on 400 the estimate of the rate of sediment deposition (Table S4). While the corresponding early Pliensbachian climatic and isotopic-shifting events cannot be observed in the smoothed 401 curves of the Early Jurassic seawater temperature and carbon cycle (Dera et al. 2011), the rapid falling event 1E<sub>CO2</sub> is well 402 403 correlated to the nearly coeval excursion events of carbon-oxygen isotopes recorded in western Tethys (Fig. 8). The 1E<sub>CO2</sub> 404 compares well to: 1) the rapid carbon isotope negative excursion of (oysters, belemnites, and brachiopods) shells from the 405 Cleveland Basin, UK (Korte and Hesselbo, 2011) and northwest Algeria (Baghli et al., 2020), 2) that of organic matter and marine carbonates from southern Pairs Basin (Bougeault et al., 2017; Peti, et al., 2017), and 3) rapid oxygen isotope negative 406 407 excursion (seawater warming) of belemnites from northern Spain (van de Schootbrugge et al., 2005). The rapid change of the stable isotope record had been called the Sinemurian-Pliensbachian boundary event (SPBE) and dated in the ammonite of 408 409 the upper Raricostatum - lower Jamesoni zones (Bougeault et al., 2017). The second event 2E<sub>CO2</sub> displays a large drop of 2574 ppmV (sample J1z-18-01 at depth 252.7 m) to 1094 ppmV (sample 410 J1z-19-01 at depth 272.3 m), ~1500 ppmV decrease within 19.6 m (estimated age interval ~0.8 Myr. Table S4 and Fig. 8c). 411 412 Following the second drop, pCO2 rises rapidly by ~1300 ppmV of 1094 ppmV to 2386 ppmV (sample J1Z-20-01 at depth 413 294.3 m) although only a few samples support the this cycle of pCO<sub>2</sub> falling-rising. Strata in western Sichuan (Xu et al., 2017), may correlate to the time interval of the T-OAE, during which pCO<sub>2</sub> doubled 414 over background values, from ~1000 ppmV to ~2000 ppmV (e.g., Beerling and Royer, 2002; McElwain et al., 2005; Berner, 415 416 2006). Given the chronostratigraphical correlation is challenging, the pCO<sub>2</sub> falling-rising cycle might correspond to the quick shifting cycle of stable isotopes during the T-OAE (Fig. 8b and 8c). In detail, the rapid falling-rising of  $pCO_2$  is 417 consistent with: 1) the quick negative-positive carbon isotope excursion of marine carbonates from Italy (Jenkyns and 418 419 Clayton, 1986; Sabatino et al., 2009), England and Wales (Jenkyns and Clayton, 1997), north Spain (van de Schootbrugge et 420 al., 2005), the Lusitanian Basin of Portugal (Hesselbo et al., 2007), Paris Basin (Hermoso et al., 2009), and Morocco (Bodin 421 et al., 2016); 2) that of invertebrate calcareous shells from the Cleveland Basin of UK (Korte and Hesselbo, 2011) and 422 northwest Algeria (Baghli et al., 2020); 3) that of marine organic matter from Morocco (Bodin et al., 2016), Yorkshire of 423 England (Cohen et al., 2004; Kemp et al, 2005), Cardigan Bay Basin of UK (Xu et al., 2018), northern Germany (van de 424 Schootbrugge et al., 2013), Alberta and British Columbia of Canada (Them II et al., 2017), northern Tibet of China (Fu et al.,





425 2016), and Japan (Izumi et al., 2018); 4) that of terrestrial organic matter from Sichuan Basin, China (Xu et al., 2017); and 5) 426 quick oxygen isotope negative-positive shifting (seawater warming) of brachiopods (Suan et al., 2008) and fossil wood 427 (Hesselbo et al., 2007) from the Lusitanian Basin, Portugal. 428 Multiple hypotheses have been proposed to interpret the 5°-6 °C decrease of sea surface temperatures in the late Pliensbachian (Bailey et al., 2003; van de Schootbrugge et al., 2005; Suan et al., 2010) and warming ~8 °C in the early 429 Toarcian (Bailey et al., 2003; Suan et al., 2010), such as the sea level falling and rising (Hallam, 1978; Hesselbo and Jenkyns, 430 431 1998), methane release (e.g., Hesselbo et al., 2000; Kemp et al, 2005; Hermoso et al., 2009; Them II et al., 2017), and the Karoo-Ferrar eruptions (e.g., Hesselbo et al., 2000; Beerling and Brentnall, 2007; Bodin et al., 2016). Perhaps, these 432 hypotheses somewhat explain the rapid change of sea surface temperatures, but it remains unclear how link the hypotheses to 433 434 drastic falling of  $pCO_2$  in a high age resolution. 435 To sum up, the perturbation and rapid falling events of the Early Jurassic pCO<sub>2</sub> values estimated from the carbon isotope of pedogenic carbonates in the GSB, are compatible with the response of stable isotopes and seawater temperature from coeval 436 437 marine sediments. Whatever caused the rapid variations of sea surface temperatures, stable isotopes, and pCO2, their 438 concordance implies that it is a positive feedback of the sea surface temperature to pCO2 through the Early Jurassic; accordingly, positive linkage could have taken place between the Early Jurassic climate and pCO<sub>2</sub>. 439

# 6. Conclusions

- 441 Based on analyses of climate-sensitive sediments and stable isotopes of the GSB, leading to a reconstruction of paleoclimate
- and  $pCO_2$ , we conclude:
- 443 1) Climate–sensitive sediments and carbon-oxygen isotope values and covariances with palynofloral reference indicate that
- an overall warm-hot and (semi-) arid climate dominated the GSB during the Early Jurassic, possibly accompanied by
- 445 occasional evaporitic climate in the Toarcian. This (semi-) arid climate in GSB is comparable with that in Colorado Plateau,
- 446 western America, but distinct from the relatively warm-humid terrestrial climate recognized in other places of Chinese
- 447 mainland (e.g., Qaidam, Inner Mongolia, and Lower Yangtze) and the northern Gondwanaland, relatively high latitudes of
- 448 Southern Hemisphere,.
- 449 2) The Early Jurassic pCO<sub>2</sub> values estimated from the carbon isotope of pedogenic carbonates in GSB show that a range
- between 980 ppmV and 2610 ppmV is ~3.5-10 times the pre-industrial value 275 ppmV and the mean 1720 ppmV is ~6
- 451 times the pre-industrial value.
- 452 3) Three phases of pCO<sub>2</sub> values were distinguished: 1500-2000 (mean ~1700) ppmV in the Sinemurian age, 1000-1500
- 453 (mean ~ 1300) ppmV in the Pliensbachian age, and 1094-2610 (mean ~1980) ppmV in the early Toarcian. The phases
- manifest the perturbation of pCO2 in the Early Jurassic.





- 455 4) Two events of rapidly falling pCO<sub>2</sub> were also recognized: ~1000-1300 ppmV drop at the Sinemurian-Pliensbachian
- boundary and quick falling (-rising) by  $\sim$ 1500 ppmV in the early Toarcian. The rapid falling events of  $pCO_2$  are compatible
- 457 with the response of stable isotopes and seawater temperature from the coeval marine sediments, implying a positive
- 458 feedback of climate to pCO<sub>2</sub> during the Early Jurassic.

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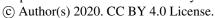


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## 781 Figures

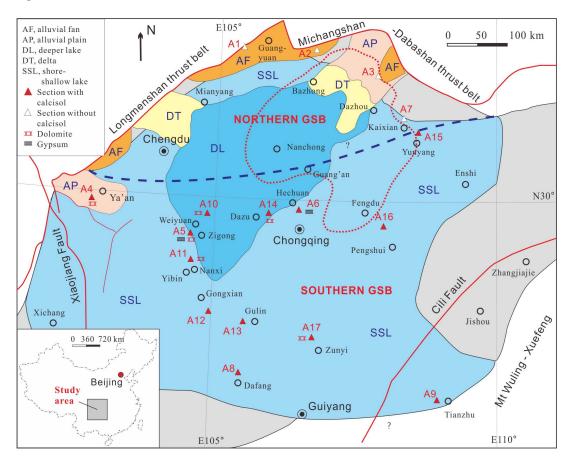


Figure 1 Lithofacies paleogeographic sketch of the grand Sichuan paleobasin (GSB) in the early Early Jurassic (Zhenzhuchong and Dongyuemiao members) showing locations of the observed and analysed sections and climate-sensitve sediments. Lithofacies paleogeographic map was composed and modified from Ma et al. (2009) and Li and He (2014). Blue area is the extent of paleolake, estemted as ~380,000 km²; blue + gray region is the basin shape, eitmated ~480,000 km². Dot red line confines the deeper lake area in the late Early Jurassic (Ma'anshan and Da'anzhai members). Bold dashed line is the northern edge of calcisol occurrence, which may separate the climate of the GSB as the northern and southern types. Triangles with numbers are locations of observed and analysed sections: A1, Xiasi section, Jian'ge; A2, Puji section, Wangcang; A3, Shiguansi section, Wanyuan; A4, Shaping section, Ya'an (bed and thickness from Wen and Zhao, 2010); A6, Tanba and Maliping section, Hechuan (bed and thickness from Wang et al., 2010); A7, Wenquan section, Kaixian (thickness from Wang et al., 2010). Location and source data s of sections A5 and A8-A17 (climate-sensitive sediments) refer to supplementary data Table S2.



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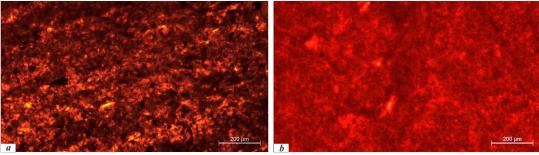


Figure 2 Microscopic cathodoluminescence photos of representative calcrete samples from the Ziliujing Fm at the Shaping section, Ya'an. a, Sample J<sub>1</sub>z-12-01, Bed B12, Ma'anshan Mem; b, Sample J<sub>1</sub>z-22-01, Bed B22, Da'anzhai Mem. Pedogenic calcites are light orange and muds are not luminescent. Pedogenic calcites of both samples are evenly luminescent light orange.

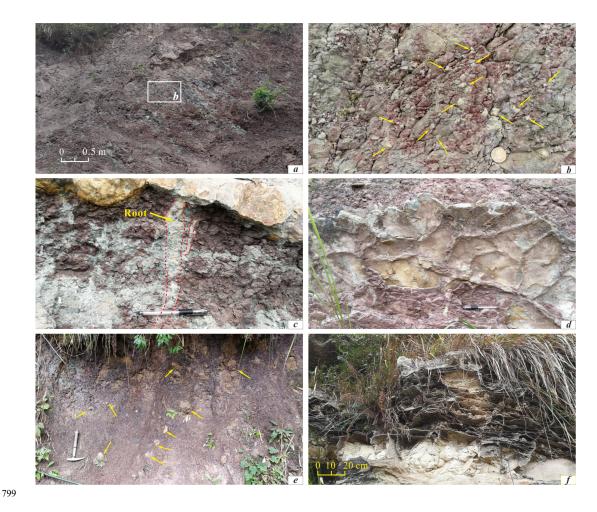


Figure 3 Field photographs of climate-sensitive sediments from the Lower Jurassic Ziliujing Fm in GSB. a, Reddish purple calcisol with strong leaching structure. Lower Bed H8 of the upper Ma'anshan Mem at Tanba village, Hechuan. b, Reddish purple calcisol showing the density and size of calcretes. The horizon and location same as a. Arrows point to calcretes. Coin 2.0 cm in diameter. c, Reddish purple calcisol with strong leaching structure and rhizoliths. Bed H13 of the top Ma'anshan Mem at





Maliuping, Hechuna. Pen 15 cm long. d, Mudcracks. Lower Bed H8 of the upper Ma'anshan Mem at Maliuping, Hechuan. Pen 15 cm long. e, Brownish red calcisol with big calcretes (calcareous concretions). Arrows point to big calcretes. Calcisol horizon  $J_1z$ -10-01, Bed B10 of Ma'anshan Mem at Shaping village, Ya'an. Hammer 34 cm long. f, Chicken-wire structure. Bed H12 of the Da'anzhai Mem at Maliuping village, Hechuan.

Series 174	Stage	Fm	Mem	A4	A10	A14	A6	A16	A15	A5	A11	A12	A13	A17	A8	A9
(Ma)				West					North → South							
176— 178— 180—	Toa	?	Da'anzhai	<b>T</b>	==	ш	<u></u>		•		<b>T</b>	•			•	
Lower Jurassi	Pli	Ziliujing	Ma'anshan	•	<b>A</b>		•	<b>A</b>		Ā			<b>A</b>	<b>A</b>	<b>A</b>	
194-	Sin	1	Dongyue- n/ao						•					<b>□</b>	<b>A</b>	
196 — 198 —	199.3	2 0	hengzhu- hong	<b>A</b>	•										<b>A</b>	<b>A</b>
200 —	201 3 Het	1 1	Qijiang	Ш	-	Qijiar	ng	Ш	?	?		Qi	jiang			Ш

IIII Hiatus ▲ Calcisol ■ Dolomitic sediment ■ Gypsum?

Figure 4 Diagram showing the temporal and spatial variation of climate-sensitive sediments in GSB. Section loactions and data sources refer to Table S2.

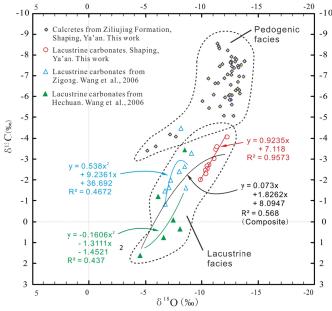


Figure 5 Cross-plot and covariance of carbon and oxygen isotopic values of the Lower Jurassic pedogenic and lacustrine carbonates from GSB. Note, the pronounced covariance ( $R^2$ =0.957) between  $\delta^{13}C$  and  $\delta^{18}O$  from Shaping section, Ya'an, indicating a compositional arid-evaporate and closed pattern lake; the moderate covariance ( $R^2$ =0.47 and 0.44) between  $\delta^{13}C$  and  $\delta^{18}O$  from Zigong and Hechuan, indicating a (semi-) arid and semi-closed pattern lake.





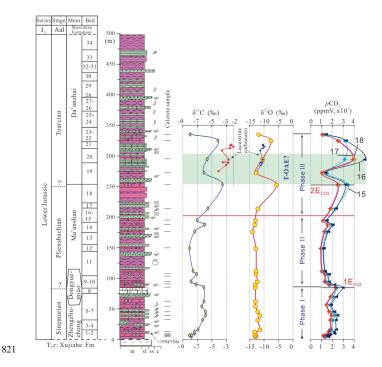


Figure 6 Diagram of the Lower Jurassic strata and litholigical log at the Shaping section, Ya'an with carbon-oxygen isotope values of pedogenic and lacustrine carbonates and  $pCO_2$  cruve. Three phases and two events can be observed for both stable isotope values of pedogenic carbonates and  $pCO_2$  estimate. T-OAE, Toarcian oceanic anoxic event.  $1E_{CO2}$  and  $2E_{CO2}$ , rapid falling event of  $pCO_2$ . Numbers 15 to 18 are the curves of  $pCO_2$  in different parameters, and details refer to supplementary Table S4.





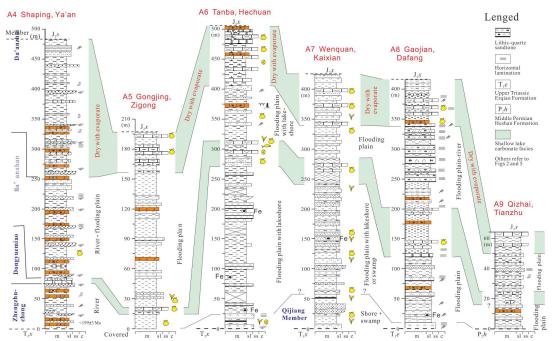


Figure 7 Stratigraphic correlation and depositional environment interpretation of the Lower Jurassic in the GSB. Data of sections refer to figure 1 and Table S2. Note, two lacustrine transgressive cycles are marked by correlative pale green areas.





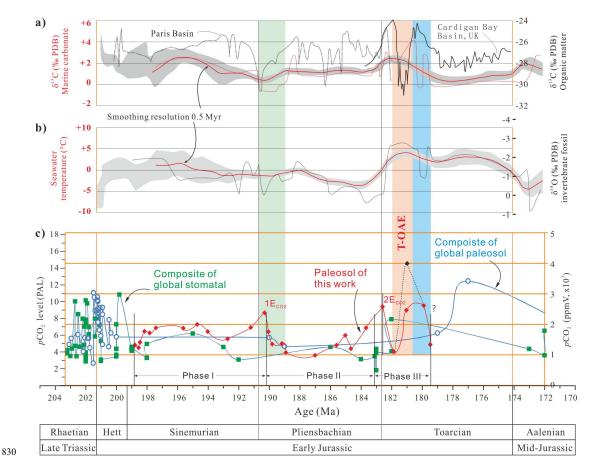


Figure 8 Comparison among the Early Jurassic  $pCO_2$ ,  $\delta^{13}C$  of marine carbonates and organic matters,  $\delta^{18}O$  of invertebrate fossils, and seawater temperature. a),  $\delta^{13}C$  (red dot line) of marine carbonates composed from Jenkyns and Clayton (1986, 1997), Hesselbo et al. (2000), Dera et al. (2011), Arabas et al., 2017;  $\delta^{13}C$  (black dot and solid lines) of organic matters are from Paris Basin, France (Peti et al., 2017) and Cardigan Bay Basin, UK (Xu et al., 2018). b),  $\delta^{18}O$  and seawater temperature (black dot line) of marine invertebrate fossils compiled from McArthur et al. (2000), Rosales et al. (2001, 2004), Jenkyns et al. (2002), Bailey et al. (2003), van de Schootbrugge et al. (2005), Gómez et al. (2008), Metodiev and Koleva-Rekalova (2008), Suan et al. (2008), Korte et al. (2009), Dera et al. (2011), Gómez et al. (2016). Smoothed  $\delta^{18}O$  and seawater temperature (red curves) in a) and b) are after Dera et al. (2011). c),  $pCO_2$  values, the composite  $pCO_2$  by paleosol and stomatal index collected from the published literatures refer to supplementary Table S5 and S6. Note: 1)  $pCO_2 = 4027$  ppmV (black solid diamond, sample J1z-20-01) if the  $\delta^{13}Cr = -29.0$  % at 181 Ma from Xu et al. (2018) in case of other constant parameters; 2) the early published  $pCO_2$  values from both carbon isotope of pedogenic carbonates and stomatal index of fossil plants (data refer to Table S5 and S6) were awfully rough dated with the average age of a lithostratigraphic formation or group, with which the uncertainty can be upto 10 Myr, leading to the difficulty of precise and accurate  $pCO_2$  correlation in pace, frequency, and event in deep time.





#### 848 Table

#### Table 1 Stratigraphic framework of the Lower Jurassic Ziliujing Fm in Sichuan and adjacent area (GSB), Southwest China

Epoch	Age	Formation	W Sichuan (Ya'an)	E Sichuan and Chongqing	S Sichuan and N Guizhou	N Sichuan	
Middle Jurassic	Aalenian	Xintiangou Fm	Xintiangou Fm	Xintiangou Fm	Xintiangou Fm	Qianfuyan / Xintiangou Fm	
	Toarcian		Da'anzhai Mem (Bed 20-34) Da'anzhai Mem Da'anzhai Mem		Da'anzhai Mem		
	Pliensbachian		Ma'anshan Mem (Bed 9-18)	Ma'anshan Mem	Ma'anshan Mem		
Early Jurassic	Sinemurian	Ziliujing Fm	Dongyuemiao Mem (Bed 8)	Dongyuemiao Mem	Dongyuemiao Mem	Baitianba Fm	
			Zhengzhuchong Mem (Bed 1-7)	Zhengzhuchong Mem	Zhengzhuchong Mem		
	Hettangian		Hiatus	Qijiang Mem	Qijiang Mem	??	
Late Triassic	Rhaetian	Xujiahe Fm	Xujiahe Fm	Xujiahe Fm	Xujiahe Fm	Xujiahe Fm	

Notes: Straigraphic classification and correlation were composed from Dong (1984); SBGM (1997), Wang et al. (2010), Wen and Zhao (2010), Xu et al (2017). Re-Os isotope age of the lower Da'anzhai Member is 180.3 ± 3.2 Ma in western Sichuan (Xu et al., 2017). Fm, Formation; Mem, Member.

### Supplementary data

# Captions of supplemenatary figures

Figure S1 Lithological log of the Lower Jurassic Ziliujing Fm with depositional environment interpretations and sample positions at the Shaping section, Ya'an of Sichuan. Bed number and thickness are partly referred to Wen and Zhao (2010)

Figure S2 Lithological log of the Lower Jurassic Ziliujing Fm at the Tanba-Maliuping section, Hechuan of Chongqing with depositional environment interpretations and sample positions. Bed number and thickness are partly referred to Wang et al (2010).

 Figure S3 Field photographs of the Lower Jurassic Ziliujing Fm lithofacies in GSB. a, Well roundness and sorting gravels in the alluvial fan conglomerate. Basal and lower Baitianba Fm. Puji, Wangcang. Hammer 30 cm long. b, Large trough cross-bedding with scours in the point bar and channel sandstones. Upper Baitianba Fm; Puji, Wangcang. c, Calcisol developed within strong leaching overbank mudrocks on channelized sandstones. Middle of Bed B2, the Zhenzhuchong Mem; Shaping section, Ya'an. d, Purple red mudrocks intercalated with thin siltstones in flood plain facies. Bed H7 of the Ma'anshan Mem; Tanba section, Hechuan. e, Whitish medium-thick micritic dolomites in lacustrine facies. Bed H12 of the Da'anzhai Mem; Maliuping section, Hechuan. Hammer 34 cm long. f, Greeinsh gray lacustrine muddy dolomites and dolomitic mudrocks associated with brownish / reddish purple mudrocks. Bed B21 of the Da'anzhai Mem; Shaping section, Ya'an.

Figure S4 Microscopic photos showing lithological microfacies of the Lower Jurassic Ziliujing Fm. *a*, Fine lithic (quartz) sandstone. Lithic-dominant fragments are mudrock. Sample J<sub>1</sub>z-02-01b, Zhenzhuchong Mem; Shaping section, Ya'an. Plain-polarised light. *b*, Laminated muddy dolomite and dolomitic mudrocks. Sample J<sub>1</sub>z-21S2B, Da'anzhai Mem; Shaping section, Ya'an. Plain-polarised light. *c*, Fine quartz arenite. Sample 18HC-02b3, Bed H2, Qijiang Mem, Tanba section, Hechuan. Cross-polarised light. *d*, Micritic dolomite. Sample 18HC-06b, Bed H12, Da'anzhai Mem. Maliuping section, Hechuan. Plain-polarised light. *e*, Coquina. Shell wall of bivalves were micritized. Mud and recrystalline calcites filled inter-shells and

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875 intra-shells. Sample 18HC-04b, Base of Bed H12, Da'anzhai Mem. Maliuping section, Hechuan. Cross-polarised light. f, Relict of coquina. Shell wall of bivalves were parly micritized. Strongly recrystalline calcites replaced the fills and shells. Sample 18HC-05b, 876 877 Bed H12, Da'anzhai Mem. Maliuping section, Hechuan. Cross-polarised light. 878 Figure S5 Field photographs of the Lower Jurassic Ziliujing Fm lithofacies in GSB. a, Lithofacies and stratigraphic sequence. 879 880 Beds B8 to B10 of the lower Ma'anshan Mem and Dongyuemiao Mem at Shaping village, Ya'an. b, Karstified gravels within the 881 limestone. The horizon and location is same as a. Pen 15 cm long, c, Layered dolomites with Karstified cave gravels. Bed H12 of 882 the Da'anzhai Mem at Maliuping village, Hechuan. d, Karstified cave gravels. The horizon and location is same as c. Hammer 34 883 cm long. 884 885 Figure S6 Stratigraphic correlation of the Lower Jurassic Baitianba Fm in northern GSB. Locations and sources refer to Figure 886 1. Plant fossils and stratal thickness in the Shiguansi section, Wanyuan are cited from SBG (1980b). 887 Captions of supplemenatary tables 888 Table S1 Early Jurassic paleosols in Ya'an of Sichuan and Hechuan of Chongging, Southwest China 889 890 891 Table S2 Carbon-oxygen isotopes of lacustrine carbonates from the Lower Jurassic Ziliujing Fm (Da'anzhai Mem) in the GSB 892 Table S3 pCO2 estimate by carbon isotope of pedogenic carbonates from the Lower Jurassic Ziliujing Fm at the Shaping section, 893 894 Ya'an of Sichuan 895 896 Table S4 Occurrence list of the Early Jurassic climate-sensitive sediments in the GSB 897 898 Table S5 Global pCO2 data of the Latest Triassic - Early Jurassic estimated by carbon isotope of pedogenic carbonates 899 900 Table S6 Global pCO2 data of the Latest Triassic - Early Jurassic by stomatal method 901 Captions of supplemenatary notes 902 903 Note S1, Description and interpretation of sedimentary facies and its evolution 904 905 Note S2, Notes of parameter usage and selection for the pCO<sub>2</sub> calculation 906