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

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Abstract: Climatic oscillations had been developed through the (Early) Jurassic from marine sedimentary archives, but 10 remain unclear from terrestrial records. This work presents investigation of climate-sensitive sediments and carbon and 11 12 oxygen isotope analyses of lacustrine and pedogenic carbonates for the Early Jurassic Ziliujing Formation from the grand 13 Sichuan paleobasin (GSB), Southwest China. Sedimentary and stable isotope proxies manifest that an overall secular (semi-) 14 arid climate dominated the GSB during the Early Jurassic except for the Hettangian. This climate pattern is similar to the 15 arid climate in the Colorado Plateau region, western North America, but distinct from the relatively warm-humid climate in North China and high latitude in Southern Hemisphere. The estimated atmospheric  $CO_2$  concentration ( $pCO_2$ ) from carbon 16 isotopes of pedogenic carbonates shows a range of 980-2610 ppmV (~ 3.5-10 times the pre-industrial value) with a mean of 17 1660 ppmV. Three phases of pCO<sub>2</sub> (the Sinemurian 1500-2000 ppmV, the Pliensbachian 1000-1500 ppmV, and the early 18 19 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 perturbation of  $pCO_2$  is compatible with seawater temperature and carbon cycle from 20 the coeval marine sediments, suggesting a positive feedback of climate to  $pCO_2$  through the Early Jurassic. 21

## 23 1. Introduction

Global paleotemperatures were possibly 5-10°C higher than present druing the Jurassic period based on climate modelling 24 results (e.g., Rees et al., 1999; Sellwood and Valdes, 2008). However, seawater temperature fluctuated by-5 °C to +5 °C, or 25 26 even much higher magnitude (e.g., Suan et al., 2008; Littler et al., 2010), based on esitmates from the oxygen isotopes of the 27 belemnite and bivalve fossils (Dera et al., 2011, and references therein). In the Sinemurian-Pliensbachian age, the mean sea 28 surface temperatures of the North Atlantic were in excess of 28°C (TEX<sub>86</sub>), comparable with similar palaeolatitudes during 29 the Cretaceous and Early Cenozoic (Robinson et al., 2017); whereas in the late Pliensbachian age, the northern West Tethys Ocean (e.g., Paris basin, northern Spain basin) was ~12.7°C (e.g., Gómez et al., 2008; Gómez and Goy, 2011; Arabas et al., 30 31 2017), leading to a polar icesheet hypothesis (e.g., Sellwood and Valdes, 2008; Suan et al., 2010; Dera et al., 2011; Gómez et al., 2015). At ~183 Ma of the early Toarcian oceanic anoxia event (T-OAE), the surface seawater temperature was high to 32 33 ~35°C (e.g., Bailey et al., 2003; Korte et al., 2015), and a high temperature (plateau) even continued in the whole Toarcian (Dera et al., 2011). Examples of seawater temperature transitions between cold and hot show the climate oscillation through 34 35 the Early Jurassic.

36 Data from the terrestrial realm also provide important details of environmental and climatic change (e.g., Hesselbo et al., 2000; Suan et al., 2010; Jenkyns, 2010; Philippe et al., 2017), from which the oscillated climate could be observed and 37 38 revealed too. Terrestrial proxies, such as flora (e.g., Riding et al., 2013; Deng et al., 2017; Philippe et al., 2017), vegetation 39 (Pole, 2009), and geochemistry (e.g., Riding et al., 2013; Kenny, 2015; Tramoy et al., 2016) as well as the pCO<sub>2</sub> record (e.g., Retallack, 2001a; Beerling and Royer, 2002; McElwain et al., 2005; Berner, 2006; Steinthorsdottir and Vajda, 2015) provide 40 41 an emerging record of the Early Jurassic terrestrial climate and environment changes. Correspondingly, the proxy application 42 of terrestrial sedimentary archives could play a key role in the global Early Jurassic correlation of the marine and terrestrial 43 climate.

Proxies for  $pCO_2$  are the important linkage between the marine and terrestrial climatic condition. Studies of the terrestrial  $pCO_2$  record have focused on the Triassic-Jurassic boundary (e.g., Tanner et al., 2001; Cleveland et al., 2008; Schaller et al., 2011; Steinthorsdottir and Vajda, 2015) and the Toarcian oceanic anoxic event (McElwain et al., 2005), where  $pCO_2$ estimates range 1000 ppm to 4000 ppmV (e.g., Tanner et al., 2001; Cleveland et al., 2008; Schaller et al., 2011). Few relatively continuous  $pCO_2$  records and coupled terrestrial climate changes have been documented for the Early Jurassic.

There are several large Triassic-Jurassic terrestrial basins in West China, in which the Sichuan Basin has a relatively complete and continuous continental sedimentary sequence of the Upper Triassic-Paleogene (e.g., SBGM, 1991, 1997; Wang et al., 2010). During the Early Jurassic, the Sichuan Basin was in a Boreotropical climate zone based on climate-sensitve sediments (Fig. 1a. Boucot et al., 2013), or a warm temperate climate is suggested based on clay mineralogy and phytogeography (e.g., Dera et al., 2009). In this work, we present a field investigation, including lithofacies and paleosol 54 interpretation, and carbon and oxygen isotope analyses of both lacustrine and pedogenic carbonates in Sichuan Basin. New

results allow us to reconstruct the paleoclimate and relatively consecutiove  $pCO_2$  record through the Early Juassic, for which

56 we compare to stable isotopes of marine sediments and estimated sea water temperature.

#### 57 **2. Geological setting and stratigraphy**

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 Yangtze plates, it became the western South China plate or cratonic basin since the Neoproterozoic (Sinian), and continued to the late Middle Triassic. By the Indosinian orogeny, new foreland basins were formed since the Late Triassic (e.g., He and Liao, 1985; Li et al., 2003), recording the Mesozoic and Cenozoic evolution 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 arcuate thrust belt in the northeast (Fig. 1b), and the northern hilly topography boundary of the Yunnan-Guizhou plateau in 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 foreland basin was much larger than the present Sichuan Basin in the eastern Sichuan province. We estimate the size of Sichuan paleobasin is roughly 480,000 km<sup>2</sup> based on lithofacies paleogeography (Fig. 1b. Ma et al., 2009; Li and He, 2014), and 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 much as 3-3.5 km thick (SBGM, 1991). Two types of Lower Jurassic deposits have been distinguished (Table 1): the Baitianba Formation (Fm) in the north (~10%) and the Ziliujing Fm (e.g., SBGM, 1991; Wang et al., 2010) in the south (over 90% of the basin).

The Baitianba Fm was deposited unconformably on the Upper Triassic Xujiahe Fm and is overlain conformably by the Middle Jurassic Xintiangou Fm / Qianfuyan Fm (Table 1). It is mainly composed of grayish shales and sandstones with coal layers and massive conglomerates. Abundant plant fossils, sporopollens, conchostracans, bivalves, and gastropods indicate it is of the Early Jurassic (SBGM, 1991, 1997). Sporopollen assemblages of the Hettangian-Sinemurian age were found in the lower part (Zhang and Meng, 1987) and the Pliensbachian-Toarcian assemblages were reported in the upper part (Wang et al., 2010).

The Ziliujing Fm is composed of variegated and reddish mudrocks (some shales) intercalated with sandstones, siltstones, and bioclastic limestones as well as dolomitic marlstones / limy dolomites, conformably or unconformably overlying the Xujiahe Fm or Luqiao Fm and conformably underlying the Xintiangou Fm (SBGM, 1997. Table 1). It has been dated as the Early 84 Jurassic by fossil assemblages of bivalves, ostracods, conchostracans, and plants. Dinosaur fauna can be well correlated to

the Lufeng Fauna in central Yunnan (e.g., Dong, 1984; SBGM, 1991, 1997; Peng, 2009). This formation is subdivided into
five parts in an ascending order: the Qijiang, Zhenzhuchong, Dongyuemiao, Ma'anshan, and Da'anzhai members (SBGM,
1997. Table 1). Of these, the former two are sometimes combined as the Zhenzhuchong Fm (e.g., SBGM, 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 reddish mudrocks (SBGM, 1991, 1997; Wang et al., 2010), and is regarded the sediment in a grand Sichuan paleolake (e.g., Ma et al., 2009; Li and He, 2014). Ostracod assembleges indicate it is the late Early Jurassic (e.g., Wei, 1982; Wang et al., 2010). A Re–Os isochron age of  $180.3 \pm 3.2$  Ma associated with an organic carbon isotope excursion indicates that the lower Da'anzhai Member corresponds to the T-OAE (Xu et al., 2017).

94 The Ma'anshan Member is comprised of violet-red mudrocks with a few greyish, greenish thin-bedded fine sandstones and siltstones, in which floral fossils are common (Li and Meng, 2003). The Dongyuemiao Member consists of greenish and 95 96 reddish mudrocks and siltstones with greyish bioclastic limestone and marlstone, of which abundant bivalve and plant fossils 97 were reported from eastern Sichuan and Chongqing (Li and Meng, 2003; Meng et al., 2003; Wang et al., 2010). The 98 Zhenzhuchong Member is dominated by violet red mudrocks/shales intercalated with thin-bedded sandstones and / or 99 siltstones and numerous plant fossils of the Early Jurassic affinity (e.g., Duan and Chen, 1982; Ye et al., 1986). Taken 100 together, fossil associations suggest that the three members were deposited in the middle-late Early Jurassic. The age 101 limitation of the overlying Da'anzhai Member and the correlation to the Lufeng dinosaur fauna places these members in the 102 Sinemurian – Pliensbachian, and the Zhenzhuchong and Dongyuemiao Fms are suggested to be the Sinemurian (Table 1).

The Qijiang Member is composed of quartz arenite interbedded/intercalated with dark shales. Coal seams are often seen 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).

## 106 **3. Materials and methods**

We have measured sections and made detatiled observations and descriptions of sedimentary characteristics for lithofacies analysis at six outcrop sections (Locations A1 to A4, A6 and A7, Fig. 1). Published descriptions for other sections (Locations A5, A8, and A9, Fig. 1) are integrated into our observations. Details of microscopic examination of sedimentary rocks and analysis of sedimentary facies underpinning the climate analysis are attached as the supplementary data Note S1. Below we state climate-sensitive sediment observation, carbon and oxygen isotope analyses, and estimate of  $pCO_2$ .

## 112 **3.1. Observation of climate-sensitive sediments**

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

114 (Table S1).

Dolomites and gypsum are relatively easy to recognize in both field and under microscope. We distinguish dolomites from limetstones following Tucker (2011) 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. In field, we recognize gypsum by particular structures such as chicken-wire cage, gypsum pseudomorph, and cluster of (0.5-1 cm) pore.

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 traces, and textures, and followed the general classification paleosols by Mack et al. (1993) and Retallack (2001b). In this paper, paleosols are described following the procedures of the Soil Survey Manual and classified according to Soil Survey Staff (1998).

Within the measured and observed sections, paleosol profiles were mainly identified from the two main locations/sections A4 and A6 (Figs. S1 and S2, and Table S2). Horizonation, BK horizon thickness, boundaries, structures, trace fossils, rootlets, carbonate accumulations (calcretes), etc. were recorded (Table S2). Paleosols interpreted in other cited sections (Fig. 1) rely on the description of lithology, structure, and calcrete in the original references. Based upon a modification of the Retallack (1998) categorization of paleosol maturity, the relative paleosol development (maturity) was assigned.

# 129 **3.2.** Analyses of carbon andoxygen isotopes

Ten lacustrine carbonate samples were analysed for carbon and oxygen isotopes from the Da'anzhai Member at the Shaping section, Ya'an (Location A4. Fig. S1 and Table S3). 26 pedogenic carbonate samples were analyzed for carbon and oxygen isotopes from 32 paleosols 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 individual calcrete for stable isotope analysis, and then a mean value for each calcrete sample was calculated (columns 9 and 10 in Table S4).

At the field scale, calcretes are ginger-like and sporadically spaced within the soil horizon. We observed no linear and planar 135 calcretes that would indicate precipitation at or below the water table. Before drilling, thin-sections were petrographically 136 137 studied using polorized light microscopy and cathodoluminescence imaging. Micritic calcite is predominant in both 138 lacustrine and pedogenic carbonate samples, with no evidence for carbonate detritus in calcretes (Fig. 2a and 2b). The 139 micritic calcites used for stable isotope analyses are chiefly null- to non-luminescent, with <10% light orange and brownish 140 luminescence, indicating genesis primarily in the vadose zone. While luminescent calcretes indicate a high possibility of 141 hydrological influence (e.g., Mintz, et al., 2016), we sampled to avoid this. Based on petrography and CL imaging together 142 with the field observations, the dense micritic zones sampled for the stable isotope composition should give pristine  $\delta^{13}$ C 143 values that can be used to estimate  $pCO_2$ .

144 Microsampling of lacustrine and pedogenic carbonates focused on only micrites, avoiding diagenetic spar from cracks, veins,

145 and vug spaces. Powder samples were obtained using a dental drill (aiguille diameter  $\phi$ =1-2 mm).

Isotopic analyses were conducted on  $0.3 \sim 0.5$  mg powder samples. Powder samples were dried in an oven at 60°C for 10 hours before being moved to the instrument. Carbon dioxide for isotopic analysis was released using orthophosphoric acid at 70°C and analysed on-line in a DELTA-Plus xp (CF-IRMS) mass spectrometer at the State Key Laboratory for Mineral 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 ±0.1‰ over the period of analysis. Calibration to the international PeeDee Belemnite (PDB) scale was performed using NBS19 and NBS18 standards.

- 153 **3.3. Calculation of atmospheric CO<sub>2</sub> concentration**
- The Cerling (1991, 1999) equation was used to calculate the  $pCO_2$  using the carbon isotope of pedogenic carbonates as below:
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$$C_{a} = S_{(z)}(\delta^{13}C_{s} - 1.0044\delta^{13}C_{r} - 4.4)/(\delta^{13}C_{a} - \delta^{13}C_{s})$$

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<sub>2</sub>, soil-respired CO<sub>2</sub>, and atmospheric CO<sub>2</sub>, respectively; and  $S_{(z)}$  is the CO<sub>2</sub> contributed by soil respiration (ppmV).

 $\delta^{13}C_s$  is often calibrated by fractionation factor -8.98‰ with the formula -8.98‰+ $\delta^{13}C_c$  (Ekart et al., 1999), with which  $\delta^{13}C_c$ is the measured result of pedogenic calcrete. Alternatively,  $\delta^{13}C_s$  can be replaced by  $\delta^{13}C_{sc}$ , which is calibrated by carbon isotope ratio of pedogenic carbonate at 25°C based on latitude–temperature correlations (Besse and Courtillot, 1988; Ekart et al., 1999) following the equation  $\delta^{13}C_{sc} = (\delta^{13}C_c+1000)/((11.98-0.12*T)/1000+1)$  -1000 (Romanek et al., 1992). We used both  $\delta^{13}C_s$  and  $\delta^{13}C_{sc}$  to calculate the *p*CO<sub>2</sub> (Table S4).

- $\delta^{13}C_r$  represents carbon isotope ratio of average bulk C3 vascular tissue (Arens et al., 2000), reflecting atmospheric  $\delta^{13}C$ (Jahren et al., 2008). The  $\delta^{13}C_{om}$  of organic matter within paleosols based on the range of modern C3 ecosystem fractionations (Buchmann, et al., 1998; Ekart et al., 1999), is commonly used for  $\delta^{13}C_r$ . However, the  $\delta^{13}C_r$  could be compromised in fossil soils due to oxidation and metabolism of organic matter after burial (Nadelhofer and Fry, 1988). In this paper, we use the  $\delta^{13}C_{om}$  from the Paris Basin (Bougeault et al., 2017; Peti et al., 2017) for the Sinemurian-Pliensbachian  $\delta^{13}C_r$  and from Cardigan Bay, UK (Xu et al., 2018) for the Toarcian.
- $δ^{13}C_a$ , the carbon isotopic composition of the atmosphere, was about -8‰ in the 1980s, being depleted relative to the pre-industrial atmosphere which was around -6.5‰ (Friedli et al., 1986). The average value of -6.5‰ has been chosen as the  $\delta^{13}C_a$  for acquiring  $\delta^{13}C_r$  and  $S_{(z)}$  (e.g., Ekart et al., 1999; Robinson et al., 2002), and the  $\delta^{13}C_a$  was generally calibrated as  $\delta^{13}C_{ac}$  from  $\delta^{13}C_r$  using the formula ( $\delta^{13}C_r$ +18.67)/1.1 (Arens et al., 2000). Herein we used both calibrations to calculate the
- 174  $\delta^{13}C_a$  (Table S4).

 $S_{(z)}$  is the largest source of uncertainty in pCO<sub>2</sub> estimates (Breecker, 2013) and the uncertainty arises primarily from their 175 176 sensitivity to soil-respired CO<sub>2</sub> (S<sub>(z)</sub> (Montañez, 2013). It is a function of depth and effectively constant below 50 cm (e.g., 177 Cerling, 1991). S<sub>(z)</sub>=2500 ppmV is suggested for the sub-humid temperate and tropical climates (Breecker et al., 2010), 178 2500-5000 ppmV for higher moisture and productivity soil (Montañez, 2013), 2000 ppmV for semi-arid areas (Breecker et 179 al., 2009), 1500-2000 ppmV for aridisols and alfisols (calcisol-argillisol) and 2000±1000 for paleo-vertisol (Montañez, 180 2013), and 1000 ppmV in desert areas (Breecker et al., 2010) or  $400 \pm 200$  ppmV for immature soil (Montañez, 2013). In this context, we chose the  $S_{(2)}=2000 \text{ ppmV}$  for calculating  $pCO_2$  at  $25^{\circ}C$  as the calcisols are reddish-brownish aridisols, and 181 182 we also compared the results with that by  $S_{(z)}=2500$  ppmV (Table S4). Additionally, we took samples at the middle and lower Bk horizon (often  $> \sim 20-30$  cm to the BK top). That means the depth of calcrete samples in the examined palaeosols 183 184 was generally deeper than 50 cm below the paleosol surface, meeting the requirement for a constant value of  $S_{(2)}$ .

#### 185 **4. Results**

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.

## 190 **4.1. Climate-sensitive sediments**

Field observation combined with published calcrete materials shows that paleosols widely occur in the Lower Jurassic 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 S2).

- 194 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
- and Table S2). Peds of paleosols are mainly angular and subangular, and a few are prismatic and platy. Slickensides are
- 196 common. Mottles (Fig. 3a), rootlets /rhizoliths (Fig. 3c), and burrows sometimes occur with strong leaching structures (Fig.
- 197 3a). Occasionally mudcracks are associated with the aforementioned structures (Fig. 3d).
- All paleosols are calcic with more or less calcretes in Bk horizons. The thickness of Bk horizons mainly changes from 30 cm
- and 100 cm, and partly up to 170 cm (Table S2). Calcretes are generally ginger-like, ellipsoid, subglobular, and irregular in
- 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).
- 201 Calcrete is often less than 0.5-1% in an individual paleosol, but a few can be up to 3-5% (paleosol J1z-3-01. Fig. 3b) even 10%
- 202 (paleosols J1z-5-02 and 18HC-10).
- All above paleosols are defined as relatively mature calcisols (Mack et al., 1993), a kind of aridisol (Soil Survey Staff, 1998;
- 204 Retallack, 2001b). The original lithofacies were chiefly argillaceous and silty (split-fan) overbank, interchannel, and flood

- 205 plain deposits (Figs. S1 and S2). Some formed landward the paleo-lakeshore.
- 206 Dolomites were found at seven loactions in central and southern GSB (Figs. 1, 4, and Table S1). The dolomites chiefly occur
- 207 in the Toracian Da'anzhai Member and a few in the 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 an authigenic origin.
- 209 Gypsum is recorded in two loactions (Figs. 1, 4, and Table S1). One is located at Zigong (Location A5. SBG, 1980a). The
- 210 other lies at Hechuan (Location A6), which can be idientifed by chicken-wire cage structure and is associated with micriditic
- 211 dolomites (Fig. 3f).

## 4.2. Carbon and 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‰ (Table S3 and Fig. 5). An increasing trend of both carbon and oxygen isotope ratios is observed from lower to upper horizons across a 45 m stratal interval of the lower Da'anzhai Member (Fig. 6).
- Pedogenic carbonate samples have  $\delta^{13}$ C values from -3.52‰ to -8.10‰, which fall in the typical stable isotope range for pedogenic carbonates. Values of -6‰ to -8.0‰ characterize the sequence of the Zhenzhuchong Member and main 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 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 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 Table S4).

# 223 4.3. CO<sub>2</sub> concentrations

- $pCO_2$  values based on paleobarometer modelling of paleosol calcite (Cerling, 1999) vary depending on the parameters used for the calculation.
- 226 If  $S_{(z)}=2500$  ppmV and  $\delta^{13}C_a=-6.5\%$  (constant pre-industrial atmosphere), pCO<sub>2</sub> values range between ~1140 ppmV and
- $\sim 3460 \text{ ppmV}$  with a mean of 1870 ppmV (column 15 in Table S4); and when  $S_{(z)}=2500 \text{ ppmV}$  and  $\delta^{13}C_a=(\delta^{13}C_r+18.67)/1.1$ ,
- $pCO_2$  values change between ~1230 ppmV and ~3260 ppmV with a mean of 2070 ppmV (column 16 in Table S4).
- 229 When  $S_{(z)}=2000$  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
- 230 (column 17 in Table S4); and if  $S_{(z)}=2000$  ppmV and  $\delta^{13}C_s = (\delta^{13}C_c+1000) / ((11.98-0.12*25) / 1000+1) -1000$  are adopted,
- $pCO_2$  values become ~980 ppmV to ~2610 ppmV with the mean 1660 ppmV (column 18 in Table S4). Details of the
- different parameters and  $pCO_2$  results can be seen in Table S4.
- 233 Results further show that  $pCO_2$  values at  $S_{(z)}=2500$  ppmV are larger than at  $S_{(z)}=2000$  ppmV. The highest difference is ~
- 1000 (3640-2610) ppmV, while the lowest is  $\sim 300 (1230-930) \text{ ppmV}$  and the mean is  $\sim 370 (2070-1600) \text{ ppmV}$ . In addition,

when  $S_{(z)}$  is the same, the pCO<sub>2</sub> values are close even if other parameters are different (comp. between columns 15 and 16,

236 17 and 18 in Table S4, and Fig. 6).

- Whatever parameters used, the trend of  $pCO_2$  over the epoch is quite similar (Fig. 6). We chose  $S_{(z)}=2000$  ppmV (column 18 in Table S4) to illustrate the nature of the Early Jurassic  $pCO_2$  in the GSB.
- $pCO_2$  values mostly range between 980 ppmV and 2610 ppmV, and the mean 1660 ppmV is ~6 times the pre-indutrial 275 ppmV. Most of the  $pCO_2$  values are 1000-2000 ppmV with the mean 1580 ppmV in the Zhenzhuchong and Ma'anshan members, ~3.5-7.5 times the pre-industrial  $pCO_2$  value.
- It is noted that the errors of  $pCO_2$  range from 384 ppmV to 1017 ppmV with a mean 647 ppmV (Table S5), leading to a large 242 243 uncertainty of the mean ~39%. The largest source of the uncertainty is the standard error (766 ppmV) of modern soil carbonate 244 (Breecker and Retallack, 2014). The  $pCO_2$  uncertainty decreases by ~ 20% if half (383 ppmv) of the standard error of soil carbonate is selected, and decreases to ~12% if 1/4 (~191 ppmV) standard error is used. The second largest source of error in 245 the  $pCO_2$  is the  $S_{(2)}$  estimate. The uncertainty of  $pCO_2$  becomes much smaller when the  $S_{(2)}$  is larger, e.g., it will fall from ~39% 246 to ~17% if  $S_{(z)}$ =5000 ppmV instead of 2000 ppmV. Other parameters such as temperature,  $\delta^{13}C_r$ ,  $\delta^{13}C_a$ ,  $\delta^{13}C_s$ , contribute very 247 little to the calculated pCO<sub>2</sub> uncertainty. The uncertainty of pCO<sub>2</sub> is same when either  $\delta^{13}C_a$  is determined by the transfer from 248 249  $\delta^{13}C_c$  of marine planktonic fossil carbonates (Table S8) or organic matters (Table S5).

## 250 **5. Discussion**

251 The Jurassic marine record shows climatic and environmental oscillations (e.g., van de Schootbrugge et al., 2005; Dera et al., 252 2011; Gómez et al., 2015; Arabas et al., 2017), including sea water temperature fluctuation and carbon cycle reorganization 253 recorded in both carbonate and organic matter. The climate changes and events recorded in the the marine realm have been 254 mainly attributed to Karoo-Ferrar volcanism (e.g., Hesselbo et al., 2000; Caruthers et a., 2013), sea-level change (e.g., Hesselbo and Jenkyns, 1998; Hallam and Wignall, 1999), orbital forcing (e.g., Kemp et al., 2005; Huang and Hesselbo, 2014, 255 Storm et al., 2020), and / or the opening of the Hispanic corridor (e.g., van de Schootbrugge et al., 2005; Arias, 2009). 256 257 Eruption of the Karoo-Ferrar and Central Atlantic mgama is thought to have released large amounts of CO<sub>2</sub> into the atmosphere in a short amount of time, resulting in rising temperatures of both marine and continental realms. The nearly 258 259 continuous record of Jurassic strata in the GSB provides an excellent test of this hypothesis in the terrestrial realm. We 260 compare the climate and  $pCO_2$  record from the GSB in relationship to the marine temperature records.

## 261 **5.1. Paleoclimate variation**

During the Late Triassic, Southwest China was warm-hot and humid and occupied a tropical and / or subtropical zone, as demonstrted by palynoflora, coals, and perennial riverine and lacustrine lithofacies in the Xujiahe Fm (e.g., Huang, 1995; Li et al., 2016). However, the climate became dry through the Early Jurassic manifested by climate-sensitive sediments and stable isotopes albeit there are two lithofacies packages reflecting two major lake stages (for details refer to supplementary
data Note S1) in the GSB.

# 267 5.1.1 The Hettangian Age

In the Hettangian, the climate was warm-humid like the Late Triassic in the GSB. The Qijiang Member is comprised of 268 269 mainly mature quartz arenites and siltstones with coals (Fig. 7) as well as siderite concretions, indicating a stable tectonic 270 setting and warm-humid climate in the eastern and southern GSB. Climate was similar across the whole region, because 271 multiple coal layers occur in the lower Baitianba Fm. The alluvial fan system of the lower Baitianba Fm. (Figs. 7 and S6) is 272 characterized by moderate-good roundness and sorting of gravels with sandy matrix (Fig. S3a. e.g., Liu et al., 2016; Qian et al., 2016; and this work). In the Newark basin of eastern North America, climate-sensitive sediments such as nodules of 273 274 carbonate and gypsum (pseudomorph) as well as mudcrack in mudflat facies indicate an arid climate in the fifth cycle of the 275 Hettangian (>199 Ma) Passaic Fm (Kent et al., 2017). More widespread, the eolian Navajo Sandstone, dated as 276 Hettangian-Sinemurian (200-195 Ma. Parrish et al., 2019), indicates arid in Colorado Plateau (Fig. 1a. Boucot et al., 2013).

# 277 5.1.2 The Sinemurian Age

The early Sinemurian Zhenzhuchong Member is a combination of riverine flood plain and lacustrine facies (supplementary Note S1). 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. Differences in the color appearance show that the reddish color started in the middle member in the central basin (Location A6. Fig. S2) but almost developed through the whole member in the western basin (Location A4. Fig. 6).

Within reddish mudrocks of the flood plain facies, multiple calcisols were observed at the Shaping section, Ya'an (Location A4. Figs. 1, 4, and 7), including a strongly leached calcisol horizon (Fig. S3c). We also interpret the reddish muddy sediments with abundant calcretes as the calcisol at sections of Dafang (Location A8. Zhang et al., 2016), Tianzhu (Location A9. Li and Chen, 2010), and Weiyuan (Location A10. SBG, 1980a). The calcisols indicate a (semi-) arid climate in the Sinemurian.

This climate change, interpreted from reddish mudrocks and paleosols, is consistent with the floral fossils (e.g., Huang, 2001; Wang et al., 2010), suggesting the decreasing humidity and increasing temperature from the Late Triassic epoch and the Hettangian age into the Sinenmurian age in the southern GSB. However, in the northern GSB there are few proxies for climate change, and alluvial fan and lacustrine delta facies common in the middle Baitianba Fm (Fig. S6. e.g., Qian et al., 2016) do not give us information on climte.

The late Sinemurian Dongyuemiao Member also has reddish mudrocks and calcisols, similar to the Zhenzhuchong Member.
Pedogenic calcretes were reported at Dafang (Location A8. Zhang et al., 2016), Tianzhu (Location A9. Li and Chen, 2010),

and Yunyang (Location A15. Meng et al., 2005) and in the central and southern GSB (Figs 4 and 7 and Table S2), indicating
 continued arid climate conditions at the time.

297 The interpreted Sinemurian (semi-) arid climate from reddish mudrocks and calcisols is supported by the flora (Li and Meng, 298 2003) and the mudrock geochemistry (Guo et al., 2017). Few records of coeval terrestrial climate are known from other 299 continents or regions in the literature. The Whitmore Point Member of the Moenave Fm deposited in dryland lakes (Tanner 300 and Lucas, 2008) and the upper part of eolian Navajo Sandstone (Blakey et al., 1988) could represent the coevally similar 301 climate in Colorado Plateau although relatively cool (~9 to 18 °C) continental climate was inferred from oxygen and 302 hydrogen isotope composition of chert precipitated in interdune, freshwater lakes in the Navajo Sandstone (Kenny, 2015). 303 With a difference, in eastern England, the co-occurrence of the acmes of thermophilic pollens Classopollis classoides and 304 Liasidium variabile indicates the warm-humid climate in the late Sinemurian (Riding et al., 2013).

# 305 5.1.3 The Pliensbachian Age

The Ma'anshan Member of the Pliensbachian displays a prominent change in the distribution and extent of red color 306 307 sediment and pedogenesis. The reddish sediments extend through the entire member (comp. Figs. 6 and S2) and can be 308 observed across most of the GSB. Calcisols are documented in both the western and central GSB (Figs. 6, 7, S1, and S2). 309 Ten calcisol horizons were recognized at the Shaping section, Ya'an (Figs. 6 and S1). Strongly leached pedogenic structures 310 and mudcracks are seen in Bed H8 of the Tanba section, Hechuan (Fig. 3a and 3d). Abundant calcretes within terrestrial red 311 mudrocks are widely described at Gaoxian of Dafang (Location A8. Zhang et al., 2016), Hulukou of Weiyuan (Location A10. 312 SBG, 1980a), Geyaoguan of Gulin (Location A13. SBG, 1976), Taiyuan of Fengdu (Location A16. SBG, 1975), and Yaxi of Zunyi (Location A17. Yang, 2015). The widespread distribution of redbeds and calcisols (Figs. 4 and 7) denotes an 313 314 intensification of the (semi-) arid climate.

Plant and sporopollen fossils also show a change to drier climate in the Pliensbachian. Compared to the Sinemurian members, more plant fossils are reported in this member (e.g., Meng and Chen, 1997; Wang et al, 2010). The Pliensbachian-Toarcian sporopollen assemblages are dominated by sporomorph genera assemblage *Dictyophyllidites-Cyathidites-Classopollis*, in which the dry-type gymnosperm spore *Classopollis* is more prevalent than in the Hettangian-Sinemurian (Zhang and Meng, 1987).

Similar dry temperate / subtropical climate is interpreted for the upland coniferous forest in Qaidam Basin, Northwest China
(Wang et al., 2005) and by interdune playa mudstones of the Kayenta Fm in Colorado Plateau (e.g., Bromley, 1992) albeit it
was a cool-humid climate in South Kazakhstan, central Asia (Tramoy et al., 2016).

# 323 5.1.4 The Toarcian Age

In spite the fact that the Da'anzhai Member was deposited in the largest lacustrine transgression period (Fig. 7. details see

supplementary data Note S1), abundant evidence for arid conditions, including backshore reddish mudrocks with calcisols,
lacustrine micritic dolomites and / or gypsum, and stable isotopic geochemistry of lacustrine carbonate, indicate that the
Toarcian aridification could be the most intensive of the late Early Jurassic in the GSB.

Redbeds with abundant calcretes are well developed in this member (Figs. 4 and 7). Four calcisols in the Shaping section (Figs. 6 and S1) and the leaching/illuvial structure (Bed H13) in the Tanba section (Fig. 3c) were observed. Calcisols with calcretes also occur at sections of Dafang (Location A8. Zhang et al., 2016), Nanxi (Location A11. SBG 1980a), Gongxian (Location A12. Liang et al., 2006), and Yunyang (Location A15. Meng et al., 2005). The widespread occurance of calcisols within the lacustrine facies reveals that subaerial exposure of sediments often interrupted the lake environment, illustrating dynamic lake level fluctuations and an arid climate.

334 Gypsum and micritic dolomites are reported in the western and southern GSB (SBG, 1980a; Mo and Yu, 1987; Peng, 2009; 335 and this work) (Figs. 1, 4, and 7). Though there are a number of hypothesies on the dolomite formation in deep time, such as 336 authigenic origin, diagenetic replacement, microbial mediation (e.g., Vasconcelos et al., 1995; Mckenzie et al., 2009; Petrash et al., 2017), a high abundance of dolomite was interpreted to form during greenhouse periods, characterized by warm 337 338 climates, probably reflecting favourable conditions for evaporite deposition and dolomitization via hypersaline reflux 339 (Warren, 2000). Dolomites are aslo thought the results of interplay of climate and sea-level / base-level change (e.g., 340 Newport et al., 2017) or are interacted with climatic regimes (Vandeginste et al., 2012). The widespread micritic dolomites 341 in the Da'anzhai Member, which are associated with gypsum (Fig. 3f), likely indicate an arid climate in the central and 342 western GSB (Fig. 1b). Gypsum occasionally occurs at Maliuping of Hechuan (Fig. 3f) and Wujiaba of Zigong (SBG, 343 1980a), showing a possible evaporitic climate in the early Toarcian in the central GSB.

- Carbon and oxygen isotopes of lacustrine carbonates further support the interpretation of an aird climate in the Toarcian age in the GSB. The mainly positive  $\delta^{13}$ C values 0 to 2 ‰ (Fig. 5) from Hechuan (Wang et al., 2006) indicate the lakes were brackish or even saline. The relatively heavy negative  $\delta^{13}$ C values -1‰ to -3.5 ‰ (Fig. 5) from Zigong (Wang et al. 2006) and Ya'an (this work) denote low depletions of <sup>13</sup>C during calcite/aragonite precipitation and mean that the lakes were possibly brackish. Lightly negative  $\delta^{18}$ O values -5‰ to -12 ‰ (Fig. 5) of the lacustrine carbonates, suggest closed lacustrine, palustrine and pond systems formed in a regional arid-semiarid climate with evaporation exceeding precipitation.
- The covariance of  $\delta^{13}$ C and  $\delta^{18}$ O is a criterion to distinguish closed or open lakes (e.g., Talbot, 1990; Li and Ku, 1997). Pronounced positive covariances (R<sup>2</sup>=0.44-0.96) between carbon and oxygen 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 gymnosperm spore *Classopollis* is more abundant than in underlying strata (e.g., Zhang and Meng, 1987; Wang et al., 2010), supporting the aridification indicated by climate–sensitive sediments and stable isotope ratios of lacustrine carbonates aforementioned.

Coastal Cheirolepidiacean (gymnosperm) forests indicate (temperate to subtropical) warm-humid climate punctuated by locally dry and/or arid events in the Toarcian in Qaidam Basin, Northwest China (Wang et al., 2005). In Inner Mongolia of 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., 2017). The warm-wet climate was also indicated by assemblages of sporomorph and vegetation in the late Early Jurassic in 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).

364 Climate-sensitive sediments, carbon and oxygen isotope values and covariance, and palynoflora, together indicate that an overall (semi-) arid climate dominated the GSB during the Early Jurassic, possibly accompanied by occasional evaporitic 365 366 climate. Relatively abundant calcisols suggest that the GSB was in a subtropical arid zone based on the paleoclimatic 367 zonation model of paleosols (Mack and James, 1994) during the middle-late Early Jurassic. Through the Early Jurassic, this (semi-) arid climate in GSB is thoroughly comparable with the simultaneous arid climate recorded in dryland lacustrine and 368 369 eolian facies in Colorado Plateau (e.g., Blakey et al., 1988; Bromley, 1992; Tanner and Lucas, 2008; Parrish et al., 2017), but 370 distinct from the relatively warm-humid climate indicated by sedimentological and floral characteristics in North China (e.g., 371 Wang et al., 2005, Deng et al., 2017) and in the relatively high latitudes of Southern Hemisphere (Pole, 2009).

In summary, the increasing aridity and warming in the GSB and arid climate in the Corlorado Plateau could have been consecutive through the Early Jurassic, and seems not harmonizaed with the global fluactueated climate that could be imprinted by two large volcanic eruptions of the Central Atlantic magmatic province and Karro-Ferrar Large Igneous Province. The secular arid climate in the two areas might be more possibly constrained by paleotopography, where both were laid in the relatively low latitudes 15-30°N (Fig. 1a).

## 377 **5.2.** *p*CO<sub>2</sub> perturbations and events

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). 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., Tanner et al., 2001; Schaller et al., 2011). Herein we present a  $pCO_2$  estimate based on data from the GSB at ~1.0 Myr age resolution for ~20 Myr (199-179 Ma) interval of the Early Jurassic (Figs. 6 and 8a).

# 383 **5.2.1.** *p*CO<sub>2</sub> perturbation

Results of model estimates show that the  $pCO_2$  values range 980-2610 ppmV with a mean 1660 ppmV in the Early Jurassic post the Hettangian and can be divided into three intervals (Figs. 6 and 8a): phase I, stable 1500-2000 (mean ~1700) ppmV in the Zhenzhuchong and Dongyuemiao members (Sinemurian age); phase II, main 1000-1500 (mean ~ 1300) ppmV in the 387 Ma'anshan Member (Pliensbachian age); and phase III, great fluctuation 1094-2610 (mean ~1980) ppmV in the lower

388 Da'anzhai Member (early Toarcian age).

- The evolution and level of  $pCO_2$  estimated by carbon isotope ratios of the pedogenic carbonates from the GSB compare favorably with the global composite based on the plant stomata method (data of the composite curve see Table S6), but show significant differences relative to the global composite  $pCO_2$  based on paleosols (Fig. 8a. Suchecki et al., 1988; Cerling, 1991; Ekart et al., 1999), which may be attributed to the shortage (<4 samples) of global data and large age uncertainties (Fig. 8a and Table S5 and S6).
- The changes in  $pCO_2$  from the GSB, has a similar pattern to coeval seawater temperature estimates through the Early Jurassic although there are some discrepancies in pace and in detail (comp. Fig. 8a and 8b). That is, the relatively high  $pCO_2$ 1500-2000 ppmV approximately corresponds to the relatively high seawater mean temperature -2°C to +2°C in the Sinemurian, low  $pCO_2$  1000-1500 ppmV corresponds to low seawater mean temperature -5°C to -2°C in the Pliensbachian, and quick rising  $pCO_2$  of 1200 ppmV to ~2500 ppmV corresponds to the rapidly increased seawater temperature of -4°C to +4°C in the late Pliensbachian-early Toarcian.
- The  $pCO_2$  record roughly trends with the carbon isotope records of marine carbonates and oganic matter (comp. Fig. 8a to 8d), suggesting a possible linkage of the  $pCO_2$  record in the GSB to the global carbon cycle (see section 5.2.2). Nevertheless, it is difficult for the proxies to compare in a higher detail, making it difficult to relate the record to orbital forcing of the global carbon cycle in the Sinemurian-Pliensbachian (Storm et al., 2020).
- 404 As a greenhouse gas, atmospheric  $CO_2$  has a strong control over global temperatures for much of the Phanerozoic (e.g., Crowley and Berner, 2001; Royer, 2006; Price et al., 2013), but a decoupling of CO<sub>2</sub> and temperature has also been 405 406 suggested (e.g., Veizer et al., 2000; Dera et al., 2011; Schaller et al., 2011). The pattern of the Early Jurassic pCO<sub>2</sub> 407 reconstructed from the carbon isotope of pedogenic carbonates in GSB, Southwest China, supports the coupled relationship of CO<sub>2</sub>-temperature. Models of the coupling and decoupling of CO<sub>2</sub>-temperature and CO<sub>2</sub>-carbon cycle have to consider: 1), 408 409 age order of CO<sub>2</sub>-temperature/carbon cycle relevance, i.e. they should be related in the same age (long term or short term) 410 hierarchy; 2) precise age constraints of individual CO<sub>2</sub> and temperature data; 3) methods of CO<sub>2</sub> and temperature estimates, 411 depending on precondition, presumptions, parameters, uncertainty, sample diagenesis, etc.; 4) controls or influences of key 412 factors such ice sheet, tectonic, paleogeography, cosmic ray flux, biota, volcanic eruption, and so on.

# 413 **5.2.2. Rapid** *p*CO<sub>2</sub> falling events

The GSB Early Jurassic  $pCO_2$  curve reveals two rapid falling events (Fig. 6 and 8a). The first event ( $1E_{CO2}$ ) shows a 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 the boundary of the Dongyuemiao and Ma'anshan Members (Fig. 6), or to 1075 ppmV (sample J1z-11-02 at depth 111.7 m), which took place in the early Pliensbachian (~190.4-189.9/189.1 Ma. Fig. 8c). The extent of the rapid falling  $pCO_2$  is 418 ~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

419 the estimate of sedimentation rate (Table S4).

- 420 While the corresponding early Pliensbachian climatic and isotopic-shifting events are not observed in the smoothed curves of 421 the Early Jurassic seawater temperature and carbon cycle (Dera et al. 2011), the rapid falling event  $1E_{CO2}$  is well correlated 422 to the nearly coeval excursion events of carbon and oxygen isotopes recorded in western Tethys and North Atlantic (Fig. 8). 423 The 1E<sub>CO2</sub> compares well to: 1) the rapid carbon isotope negative excursion of (oysters, belemnites, and brachiopods) shells 424 from the Cleveland Basin, UK (Korte and Hesselbo, 2011) and northwest Algeria (Baghli et al., 2020), 2) that of organic 425 matter and marine carbonates from southern Pairs Basin (Bougeault et al., 2017; Peti, et al., 2017) and Cardigan Bay Basin, UK (Storm et al., 2020), and 3) rapid oxygen isotope negative excursion (seawater warming) of belemnites from northern 426 427 Spain (van de Schootbrugge et al., 2005). The rapid change of the stable isotope record had been called the 428 Sinemurian-Pliensbachian boundary event (SPBE) and dated in the ammonite of the upper Raricostatum - lower Jamesoni 429 zones (Bougeault et al., 2017).
- The second event  $2E_{CO2}$  displays a large drop of 2574 ppmV (sample J1z-18-01 at depth 252.7 m) to 1094 ppmV (sample 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. 8a). Following the second drop,  $pCO_2$  rises rapidly by ~1300 ppmV of 1094 ppmV to 2386 ppmV (sample J1Z-20-01 at depth 294.3 m) although only a few samples support the this cycle of  $pCO_2$  falling-rising.
- 434 Strata in western Sichuan (Xu et al., 2017), may correlate to the time interval of the T-OAE, during which  $pCO_2$  doubled 435 over background values, from ~1000 ppmV to ~2000 ppmV (e.g., Beerling and Royer, 2002; McElwain et al., 2005; Berner, 436 2006). Given that chronostratigraphical correlation is challenging, the  $pCO_2$  falling-rising cycle might correspond to the 437 quick shifting cycle of stable isotopes during the T-OAE (Fig. 8a and 8c-8d). In detail, the rapid falling-rising of  $pCO_2$  is 438 consistent with: 1) the quick negative-positive carbon isotope excursion of marine carbonates from Italy (Jenkyns and 439 Clayton, 1986; Sabatino et al., 2009), England and Wales (Jenkyns and Clayton, 1997), north Spain (van de Schootbrugge et 440 al., 2005), the Lusitanian Basin of Portugal (Hesselbo et al., 2007), Paris Basin (Hermoso et al., 2009), and Morocco (Bodin 441 et al., 2016); 2) that of invertebrate calcareous shells from the Cleveland Basin of UK (Korte and Hesselbo, 2011) and 442 northwest Algeria (Baghli et al., 2020); 3) that of marine organic matter from Morocco (Bodin et al., 2016), Yorkshire of 443 England (Cohen et al., 2004; Kemp et al, 2005), Cardigan Bay Basin of UK (Xu et al., 2018), northern Germany (van de 444 Schootbrugge et al., 2013), Alberta and British Columbia of Canada (Them II et al., 2017), northern Tibet of China (Fu et al., 445 2016), and Japan (Izumi et al., 2018); 4) that of terrestrial organic matter from Sichuan Basin, China (Xu et al., 2017); and 5) 446 quick oxygen isotope negative-positive shifting (seawater warming) of brachiopods (Suan et al., 2008) and fossil wood 447 (Hesselbo et al., 2007) from the Lusitanian Basin, Portugal.
- 448 Multiple hypotheses have been proposed to interpret the 5°–6 °C decrease of sea surface temperatures in the late 449 Pliensbachian (Bailey et al., 2003; van de Schootbrugge et al., 2005; Suan et al., 2010) and warming  $\sim 8$  °C in the early

450 Toarcian (Bailey et al., 2003; Suan et al., 2010), such as the sea level falling and rising, methane release, Karoo-Ferrar 451 eruption, Hispanic corridor opening, etc. Perhaps, these hypotheses somewhat explain the rapid change of sea surface 452 temperatures, but might not link to drastic falling of  $pCO_2$ . As we know, atmospheric  $CO_2$  is controlled by volcanism, 453 weathering, vegetation on land and phytoplankton in ocean, and orbiting forcing. The Sr isotope curve shows a rapid change in the early Toarcian but does not in the early Pliensachian (e.g., Jones et al., 1999), indicating a distinct transfer of 454 455 weathering took place on the land only at the T-OAE time. No robust evidence shows the rapid changes of terrestrial vegetation and marine primary productivy for the two intervals except for the floral change in western Tethys during the 456 457 T-OAE (Slater et al. 2019). The Karoo–Ferrar eruption could be responsible for the rapid rising of  $pCO_2$  but not for the falling. Then the orbital forcing might be an alternative. 458

To sum up, the rapid falling events of the Early Jurassic  $pCO_2$  values in the GSB, are compatible with the response of stable isotopes (carbon cycle) and seawater temperature from coeval marine sediments in a total tendency and eventful change, but not harmonized at a high-resolution time scale. Whatever caused the rapid variations of sea surface temperatures, stable isotopes, and  $pCO_2$ , their near concordance suggests that it is a positive feedback of the sea surface temperature and carbon cycle to the  $pCO_2$  in trend and event through the Early Jurassic; whereas the higher frequency changes in the Sinemurian-Pliensbachian might may support other causal driving of the climate, such as orbital forcing (Storm et al., 2020).

## 465 **6. Conclusions**

Based on analyses of climate-sensitive sediments and stable isotopes and the reconstruction of paleoclimate and  $pCO_2$ , we conclude:

468 1) An overall warm-hot and (semi-) arid climate dominated the GSB during the Early Jurassic, possibly accompanied by 469 occasional evaporitic climate in the Toarcian. This (semi-) arid climate in GSB is comparable with that in Colorado Plateau, 470 western America, but distinct from the relatively warm-humid terrestrial climate recognized in other places of Chinese 471 mainland (e.g., Qaidam, Inner Mongolia, and Lower Yangtze) and the high latitudes of Southern Hemisphere.

2) The Early Jurassic  $pCO_2$  values 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 times the pre-industrial value. Three phases of  $pCO_2$  values were distinguished: 1500-2000 (mean ~1700) ppmV in the Sinemurian age, 1000-1500 (mean ~ 1300) ppmV in the Pliensbachian age, and 1094-2610 (mean ~1980) ppmV in the early Toarcian. Two events of rapidly falling  $pCO_2$  were also recognized: ~1000-1300 ppmV drop at the Sinemurian-Pliensbachian boundary and quick falling (-rising) by ~1500 ppmV in the early Toarcian. The phases and events manifest the perturbation of  $pCO_2$  in the Early Jurassic.

3) The perturbation and rapid falling events of the Early Jurassic  $pCO_2$  from the GSB are compatible with the carbon cycle and seawater temperature from coeval marine sediments in the North Atlantic and western Tethys in a total tendency and

- 480 eventful change. The compatibility suggests that it is a positive linkage of the sea surface temperature and carbon cycle to
- the  $pCO_2$  through the Early Jurassic. On the contrary, differences at a high-resolution time sacle implies additional climate drivers, such as orbital forcing are important in the Sinemurian-Pliensbachian record.

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Figure 1 A, Global Early-Middle Jurassic climate zones (Boucot et al., 2013) laid on the Early Jurasse (~193 Ma, Sinemurian) paleogeographic map (Scotese, 2014). B, 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 km<sup>2</sup>; blue + gray region is the basin shape, eitmated ~480,000 km<sup>2</sup>. 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 of sections A5 and A8-A17 (climate-sensitive sediments) refer to supplementary data Table S1.



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 Member; *b*, Sample J<sub>1</sub>*z*-22-01, Bed B22, Da'anzhai Member. Pedogenic
calcites are mainly null to non-luminescent, minor are orange/red luminescence. Inserts are the scanned photos of thin-section, and

841 rectangles are the area under cathodoluminescence and drilling.



843 Figure 3 Field photographs of climate-sensitive sediments from the Lower Jurassic Ziliujing Fm in the GSB. a, Reddish purple 844 calcisol with strong leaching structure. Lower Bed H8 of the upper Ma'anshan Member at Tanba, Hechuan. b, Reddish purple 845 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 Member at 846 847 Maliuping, Hechuna. Pen 15 cm long. d, Mudcracks. Lower Bed H8 of the upper Ma'anshan Member at Maliuping, Hechuan. Pen 848 15 cm long. e, Brownish red calcisol with big calcretes (calcareous concretions). Arrows point to big calcretes. Calcisol horizon 849 J<sub>17</sub>-10-01, Bed B10 of Ma'anshan Member at Shaping, Ya'an. Hammer 34 cm long. f, Chicken-wire structure. Bed H12 of the 850 Da'anzhai Member at Maliuping, Hechuan.

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853 Figure 4 Diagram showing the temporal and spatial variation of climate-sensitive sediments in GSB. Section loactions and data

854 sources refer to Table S1.



Figure 5 Cross-plot and covariance of carbon and oxygen isotopic values of the Lower Jurassic pedogenic and lacustrine carbonates from the 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.

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Figure 6 Diagram of the Lower Jurassic strata and litholigcal log at the Shaping section, Ya'an with carbon and 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. Legend of lithology in log refers to supplementary Figs. S1 and S2. 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 Fig. 1. 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 875 fossils, and seawater temperature. Age model is from Cohen et al. (2013). a), pCO<sub>2</sub> values of this work and the composite pCO<sub>2</sub> by 876 paleosol and stomatal index (supplementary Table S6 and S7). Vertical bars are errors (10) of pCO<sub>2</sub> (Table S5). Errors are 877 propagated using the Gaussian approach (Breecker and Retallack, 2014). Note: 1) pCO<sub>2</sub> = 4027 ppmV (black solid diamond, 878 sample J1z-20-01) if the  $\delta^{13}C_r$  = -29.0 ‰ at 181 Ma from Xu et al. (2018) in case of other constant parameters; 2) the early 879 880 published  $pCO_2$  values from both carbon isotope of pedogenic carbonates and stomatal index of fossil plants (data refer to Table 881 S6 and S7) were awfully rough dated with the average age of a lithostratigraphic formation or group, with which the uncertainty 882 can be upto 10 Myr, leading to the difficulty of precise and accurate  $pCO_2$  correlation in pace, frequency, and event. b),  $\delta^{18}O$  and 883 seawater temperature (black dot line) of marine invertebrate fossils compiled from Rosales et al. (2001, 2004), Jenkyns et al. (2002), 884 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. (2015). c), red dot line  $\delta^{13}$ C of marine carbonates in western Tethys, 885 composed from Jenkyns and Clayton (1986, 1997), Hesselbo et al. (2000), Dera et al. (2011), Arabas et al., 2017; black dot and solid 886 line  $\delta^{13}$ C of organic matters from Paris Basin, France (Peti et al., 2017). Smoothed  $\delta^{18}$ O and seawater temperature (red curves) in 887 b) and c) are after Dera et al. (2011). d),  $\delta^{13}$ C of organic matters from North Atlantic. Composed from the Mochras borehole, 888 889 Cardigan Bay Basin, UK (Xu et al., 2018; Storm et al., 2020), seven-point average smoothing against depth (mbs).

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
Early Jurassic	Toarcian	Ziliujing Fm	Da'anzhai Mem (Bed 20-34)	Da'anzhai Mem	Da'anzhai Mem	
	Pliensbachian		Ma'anshan Mem (Bed 9-18)	Ma'anshan Mem	Ma'anshan Mem	
	Sinemurian		Dongyuemiao Mem (Bed 8)	Dongyuemiao Mem	Dongyuemiao Mem	Baitianba Fm
			Zhenzhuchong Mem (Bed 1-7)	Zhenzhuchong Mem	Zhenzhuchong Mem	
	Hettangian		Hiatus	Qijiang Mem	Qijiang Mem	?
Late Triassic	Rhaetian	Xujiahe Fm	Xujiahe Fm	Xujiahe Fm	Xujiahe Fm	Xujiahe Fm

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

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.

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## 894 Supplementary data

# 895 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).

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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).

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903 Figure S3 Field photographs of the Lower Jurassic Ziliujing Fm lithofacies in the GSB. a, Well roundness and sorting gravels in 904 the alluvial fan conglomerate. Basal and lower Baitianba Fm. Puji, Wangcang. Hammer 30 cm long. b, Large trough 905 cross-bedding with scours in the point bar and channel sandstones. Upper Baitianba Fm; Puji, Wangcang. c, Calcisol developed 906 within strong leaching overbank mudrocks on channelized sandstones. Middle of Bed B2, the Zhenzhuchong Member, Shaping 907 section, Ya'an. d, Purple red mudrocks intercalated with thin siltstones in flood plain facies. Bed H7 of the Ma'anshan Member, 908 Tanba section, Hechuan. e, Whitish medium-thick micritic dolomites in lacustrine facies. Bed H12 of the Da'anzhai Member, 909 Maliuping section, Hechuan. Hammer 34 cm long. f, Greeinsh gray lacustrine muddy dolomites and dolomitic mudrocks 910 associated with brownish / reddish purple mudrocks. Bed B21 of the Da'anzhai Member, Shaping section, Ya'an.

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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 Member, Shaping section, Ya'an. Plain-polarised light. *b*, Laminated muddy dolomite and dolomitic mudrocks. Sample J<sub>1</sub>z-21S2B, Da'anzhai Member, Shaping section, Ya'an. Plain-polarised light. *c*, Fine quartz arenite. Sample 18HC-02b3, Bed H2, Qijiang Member, Tanba section, Hechuan. Cross-polarised light. *d*, Micritic dolomite. Sample 18HC-06b, Bed H12, Da'anzhai Member, Maliuping section, Hechuan. Plain-polarised light. *e*, Coquina. Shell wall of bivalves were micritized. Mud and recrystalline calcites filled inter-shells

- and intra-shells. Sample 18HC-04b, Base of Bed H12, Da'anzhai Member, 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, Bed H12, Da'anzhai Member, Maliuping section, Hechuan. Cross-polarised light.
- Figure S5 Field photographs of the Lower Jurassic Ziliujing Fm lithofacies in the GSB. *a*, Lithofacies and stratigraphic sequence.
  Beds B8 to B10 of the lower Ma'anshan and Dongyuemiao members at Shaping, Ya'an. *b*, Karstified gravels within the limestone.
  The horizon and location is same as *a*. Pen 15 cm long. *c*, Layered dolomites with Karstified cave gravels. Bed H12 of the
  Da'anzhai Member at Maliuping, Hechuan. *d*, Karstified cave gravels. The horizon and location is same as *c*. Hammer 34 cm long.
- Figure S6 Stratigraphic correlation of the Lower Jurassic Baitianba Fm in northern GSB. Locations and sources refer to Figure
  Plant fossils and stratal thickness in the Shiguansi section, Wanyuan are cited from SBG (1980b).
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- 930 Captions of supplemenatary tables
- 931 Table S1 Occurrence list of the Early Jurassic climate-sensitive sediments in the GSB
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- 933 Table S2 Early Jurassic paleosols in Ya'an of Sichuan and Hechuan of Chongqing, Southwest China
- Table S3 Carbon-oxygen isotope composition of lacustrine carbonates from the Lower Jurassic Ziliujing Fm (Da'anzhai Mem) in
   the GSB
- Table S4 pCO<sub>2</sub> estimate by carbon isotope of pedogenic carbonates from the Lower Jurassic Ziliujing Fm at Shapingion, Ya'an
   of Sichuan
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- Table S5 Calculation of *p*CO<sub>2</sub> and Gaussian error propagation using the atmosphere determination of global organic matter isotope composition for the Early Jurassic Sichuan paleobasin
- 944 Table S6 Global *p*CO<sub>2</sub> data of the Latest Triassic Early Jurassic by stomatal method
- 946 Table S7 Global pCO<sub>2</sub> data of the Latest Triassic Early Jurassic estimated by carbon isotope of pedogenic carbonates
- Table S8 Calculation of  $pCO_2$  and Gaussian error propagation using the atmosphere carbon isotope determination of mairne fossil carbonate carbon isotope composition for the Early Jurassic Sichuan paleobasin
- 950 Captions of supplemenatary notes
- 951 Note S1, Description and interpretation of sedimentary facies and its evolution
- 952
- 953