

## Note S2, Notes of parameter usage and selection for the $p\text{CO}_2$ calculation

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Using the carbon isotope of pedogenic carbonates, usage and selection of parameters in the Cerling (1999) empirical equation of calculation of  $p\text{CO}_2$   $C_a = S_{(z)}(\delta^{13}\text{C}_s - 1.0044\delta^{13}\text{C}_r - 4.4)/(\delta^{13}\text{C}_a - \delta^{13}\text{C}_s)$  are noted and discussed below.

In the parameters,  $\delta^{13}\text{C}_s$  is the carbon-isotope composition of pedogenic carbonate and is often calibrated by temperature-dependent fractionation factor  $-8.98\text{‰}$  with the formula  $-8.98\text{‰} + \delta^{13}\text{C}_c$  (Ekart et al., 1999), with which  $\delta^{13}\text{C}_c$  is the measured result of pedogenic calcrite. Or,  $\delta^{13}\text{C}_s$  can be replaced by  $\delta^{13}\text{C}_{sc}$ , which is calibrated by the carbon-isotope composition of pedogenic carbonate at 25 °C based on paleomagnetic data and latitude–temperature correlations (Besse and Courtillot, 1988; Ekart et al., 1999) following the equation  $\delta^{13}\text{C}_{sc} = (\delta^{13}\text{C}_c + 1000)/((11.98 - 0.12 \cdot T)/1000 + 1) - 1000$  (Romanek et al., 1992). We used both  $\delta^{13}\text{C}_s$  and  $\delta^{13}\text{C}_{sc}$  to calculate the  $p\text{CO}_2$ , respectively in this article (Table S4).

$\delta^{13}\text{C}_r$  represents carbon-isotope ratio of average bulk C3 vascular land-plant tissue (Arens et al., 2000), reflecting atmospheric  $\delta^{13}\text{C}$  linearly across  $p\text{CO}_2$  levels (Jahren et al., 2001, 2008). So, the  $\delta^{13}\text{C}_{om}$  of organic matter within paleosols, which is the carbon-isotope ratio of soil organic matter based on the range of modern C3 ecosystem fractionations (Buchmann, et al., 1998; Ekart et al., 1999), is commonly thought to be representative of  $\delta^{13}\text{C}_r$  in the model equation (Cerling, 1999). However, the  $\delta^{13}\text{C}_r$  could be not almost applied in the measurement of the fossil soils due to oxidation and metabolism of organic matter after burial, so that the remaining organic matter would not represent the original materials (Nadelhofer and Fry, 1988). A proxy model of carbon isotope was developed between the marine carbonate, atmosphere, and soil organic matter, and values of the  $\delta^{13}\text{C}_{om}$  through the Phanerozoic can be generally read from the model curve of Ekart et al. (1999). Another way to obtain  $\delta^{13}\text{C}_r$  is from the  $\delta^{13}\text{C}_a$  by calibration of marine foraminiferal carbon-isotope composition (e.g., Arens et al. 2000; Passey and Cerling, 2002; Nordt et al., 2002; Breecker and Retallack, 2014. Details refer to Zhang et al., 2018). Nevertheless,  $\delta^{13}\text{C}_a$  is also an uncertain parameter, and the Cerling (1999) model curve is a highly smoothed and does not capture the variability in  $\delta^{13}\text{C}$  of the ocean–atmosphere carbon reservoir on a short-term scale. Therefore, we used well-dated estimates of  $\delta^{13}\text{C}_r$ . We partly used the carbon-isotope values of organic matters for the Sinemurian-Pliensbachian  $\delta^{13}\text{C}_r$  from the Paris Basin (Bougeault et al., 2017; Peti et al., 2017) and for the Toarcian one from Cardigan Bay, UK (Xu et al., 2018).

$\delta^{13}\text{C}_a$ , the carbon isotopic composition of the atmosphere, was about  $-8\text{‰}$  in the 1980s, being depleted relative to the pre-industrial atmosphere which was around  $-6.5\text{‰}$  (Friedli et al., 1986). The average value of  $-6.5\text{‰}$  has been chosen as the  $\delta^{13}\text{C}_a$  for acquiring  $\delta^{13}\text{C}_r$  and  $S_{(z)}$  (e.g., Ekart et al., 1999; Lee and Hisada, 1999; Robinson et al., 2002; Li et al., 2014), and the

$\delta^{13}\text{C}_a$  was generally calibrated as  $\delta^{13}\text{C}_{ac}$  from  $\delta^{13}\text{C}_r$  of  $S_{(z)}$  using the formula  $(\delta^{13}\text{C}_r+18.67)/1.1$  (Arens et al., 2000). Herein we used both calibrations to calculate the  $\delta^{13}\text{C}_a$  (Table S4).

Though  $S_{(z)}$  is the largest source of uncertainty in  $p\text{CO}_2$  estimates (Breecker et al., 2013) and the large uncertainty arises primarily from their sensitivity to soil-respired  $\text{CO}_2$  ( $S_{(z)}$ ) (Montañez, 2013). It is a function of depth and effectively constant below 50 cm, where carbonates are precipitated (e.g., Cerling, 1991; Cerling and Quade, 1993). In earlier publications, a value of 5000 ppmV was often adopted for  $S_{(z)}$  in paleo- $p\text{CO}_2$  reconstructions. However, this value is much higher than those estimated by examination of plant stomata (e.g. Royer, 2006; Fletcher et al., 2008). Breecker et al. (2010) interpreted the large discrepancy of  $S_{(z)}$  as due to large variations in atmospheric  $p\text{CO}_2$  over relatively short time periods as well as inaccurate proxy estimates. Instead they used carbon-isotope ratios of Holocene calcic soil to recalculate and calibrate atmospheric  $p\text{CO}_2$  from a value of  $S_{(z)}=2500$  ppmV. In detail,  $S_{(z)}=2500$  ppmV is suggested for the sub-humid temperate and tropical climates (Breecker et al., 2010), 2500-5000 ppmV for higher moisture and productivity soil (Montañez, 2013), 2000 ppmV for semi-arid areas (Breecker et al., 2009), 1500-2000 ppmV for aridisols and alfisols (calcisol-argillisol) and  $2000\pm1000$  for paleo-vertisol (Montañez, 2013), and 1000 ppmV in desert areas (Breecker et al., 2010) or  $400 \pm 200$  ppm V for immature soil (Montañez, 2013). In this context, we chose the  $S_{(z)}=2000$  ppmV for calculating  $p\text{CO}_2$  at  $25^\circ\text{C}$  as the paleosols are reddish-brownish (arid-semiarid) calcisols recognized in the studied sections and we also compared the results with that by  $S_{(z)}=2500$  ppmV. Additionally, we took samples at the middle and lower Bk horizon (often  $> \sim 20\text{-}30$  cm to the BK top ). That means the depth of calcrete samples in the examined palaeosols was generally deeper than 50 cm below the paleosol surface, meeting the requirement for a constant value of  $S_{(z)}$ .

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