



# Article Records of Organic Carbon Isotopic Composition and Its Paleoenvironmental Implications in Shengshan Island Loess Deposition in the East China Sea during the Last Glacial Period

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Abstract: Organic carbon isotopic composition ( $\delta^{13}C_{org}$ ) in loess deposits is an important indicator of terrestrial paleovegetation, and it has been widely used for paleoenvironmental reconstruction in aeolian sediments around the world. However, little research has been done on the variation and paleoenvironmental implication of  $\delta^{13}C_{org}$  from loess deposits on Shengshan Island, East China Sea, during the last glacial period (LG). In this research, we present optically stimulated luminescence (OSL) ages, total organic carbon (TOC) data and  $\delta^{13}C_{org}$  records of the loess section at Chenqianshan (CQS) on Shengshan Island. Additionally, to study the effectiveness of  $\delta^{13}$ C<sub>org</sub> in documenting paleoenvironmental changes, magnetic susceptibilities and diffuse reflectance spectra were surveyed. TOC concentration for the CQS loess section ranged from 0.11% to 0.47%, and the  $\delta^{13}C_{org}$  composition of the CQS loess section varied between -20.80% and -24.56% during the LG. The average value of C<sub>4</sub> abundance was 21.31%. TOC,  $\delta^{13}$ Corg,  $\chi_{fd}$ , and Hm/(Hm + Gt) curves for the CQS loess section showed similar patterns. The results of our study indicated that the vegetation of the CQS loess deposit was mainly  $C_3/C_4$  mixed vegetation, and  $C_3$  vegetation was the most important vegetation. The comparison between the  $\delta^{13}C_{org}$  curve for the CQS section and other existing  $\delta^{13}C_{org}$  records of the loess sections from central and northern China showed similar trends and their vegetation succession exhibited synchronous change during the LG. Based on a comparison of the  $\delta^{13}C_{org}$  record,  $C_4$  abundance and  $\chi_{fd}$  of the CQS section and other global geological records, it was concluded that the mutual effects of precipitation and temperature caused the change of paleovegetation in loess deposits on islands in the East China Sea during the LG.

**Keywords:** +30]loess; organic carbon isotopes;  $C_3/C_4$  vegetation abundance; paleovegetation; palaeoenvironment

#### 1. Introduction

Because of their different photosynthesis, terrestrial plants are commonly called C<sub>3</sub> plants and C<sub>4</sub> plants. The values of carbon isotopic composition ( $\delta^{13}C_{org}$ ) of modern C<sub>3</sub> plants range between ~-22‰ and -30‰. The  $\delta^{13}C_{org}$  composition of modern C<sub>4</sub> plants varies from ~-9‰ to -19‰ [1,2]. Pure C<sub>3</sub> plants prefer to grow in cold, humid and high concentration CO<sub>2</sub> conditions, but pure C<sub>4</sub> plants prefer to grow in hot, arid and low concentration CO<sub>2</sub> conditions [3]. The  $\delta^{13}C_{org}$  composition of the soil is controlled by vegetation and environmental conditions. Theoretically, the  $\delta^{13}C_{org}$  composition of the soil can be used to study the change of relative abundance of C<sub>3</sub>/C<sub>4</sub> plants and the reconstruction of paleoenvironment and paleoclimate in the past geological history period [4–6].



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**Copyright:** © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). Many studies have shown that, in addition to genetic factors, environmental factors, such as precipitation, temperature and CO<sub>2</sub> concentrations, could affect the  $\delta^{13}C_{org}$  composition of soil [7–9]. The climate was cold and dry, and the atmospheric CO<sub>2</sub> concentration was low during the last glacial period (LG). A relatively dry climate and low CO<sub>2</sub> concentration were beneficial to the growth of C<sub>4</sub> plants, however, low temperature limited the growth of C<sub>4</sub> plants [7]. It was demonstrated that during the LG, the values of  $\delta^{13}C_{org}$  in loess deposits have been more negative than those in paleosols [10–12]. However, in the eastern part of Europe, in the Tianshan Mountains and the northwest of the Chinese Loess Plateau, the  $\delta^{13}C_{org}$  composition in loess deposits has been more positive than those in paleosols during the LG [6,13,14]. Moreover, the vegetation was mainly controlled by C<sub>3</sub> plants, and the signal of C<sub>4</sub> vegetation was not obvious in these regions. Thus, it is necessary to further investigate the response of  $\delta^{13}C_{org}$  composition from the loess-paleosol sequences to climate or environmental conditions in different time scales and regions during the LG [7,15,16].

Marine isotope stages (MIS) are an alternate stage of cold and warm in the earth's paleoclimate, inferred from oxygen isotope data reflecting temperature changes. Modern times are marked as the first stage of the MIS. The even stage usually has a high level of <sup>18</sup>O, and it is a cold ice stage, while the odd stage is a warm interglacial stage [17]. Malan loess was formed by the accumulation of dust and the process of loess formation during the last glacial period [18]. An et al. [19] divided Malan loess into three stages, namely L1L1, L1S1 and L1L2. Among them, L1L1 and L1L2 are the early and late Malan loess deposits, and L1S1 is the paleosol in the middle of the Malan loess. Kukla et al. [20] found that the magnetic susceptibility in the middle part of the Malan loess is higher than that in other horizons, which can be compared with MIS 3. Therefore, the L1L1, L1S1 and L1L2 stages in Malan loess correspond to MIS 2, MIS 3 and MIS 4, respectively [21].

There are many islands in the eastern extension areas of China, and they are scattered with loess deposits [22]. Studies of island loess deposits mainly focused on ages, particle sizes, spectral characteristics, elements and magnetic proxy indicators [23–27]. Shengshan Island is one of the Zhoushan Islands. The hillside of the island comprises exposed granite bedrock, and it is covered with loess deposits. Until now, little study has been done on the variation and paleoenvironmental implication of loess deposits'  $\delta^{13}C_{org}$  composition on Shengshan Island, East China Sea. Therefore, the process and response mechanism of the  $\delta^{13}C_{org}$  change in this area is still unclear.

Here, we display total organic carbon (TOC) data, organic carbon isotope composition ( $\delta^{13}C_{org}$ ), frequency-dependent magnetic susceptibility ( $\chi_{fd}$ ) and diffuse reflectance spectra (DRS) for the loess section on Shengshan Island. By combining these data with the results of optically stimulated luminescence (OSL) dating, this study is intended to (1) recognize the variation characteristics of the  $\delta^{13}C_{org}$  composition from the CQS loess section in the Shengshan Island area and (2) discuss the mechanism driving the  $\delta^{13}C_{org}$  variations in Shengshan Island loess during the LG. Accordingly, the study results will enhance understanding of the paleoenvironment evolution of the East China Sea islands.

#### 2. Materials and Methods

#### 2.1. Study Site

The landscape of Shengshan Island is mountainous and hilly. The climate is mainly controlled by the East Asian monsoon. The mean annual temperature (MAT) is 15~17 °C, the mean annual precipitation (MAP) is 1072 mm, and the solar radiation rate is 48% [26]. Because of the influences of altitude and ecological environment, the vegetation of Shengshan Island is mainly coniferous forests (including mainly black pine forests and Chinese fir forests), with scattered broad-leaved forests. Chenqianshan (CQS) is the highest peak on Shengshan Island, and its average altitude is ~3400 m above sea level. The CQS loess profile is located on the northwest slope of Shengshan Island (latitude 30.73° N, longitude 122.817° E, and altitude 150 m). The 0–30 cm thick CQS loess section is a modern soil layer, which is obviously artificially disturbed. Therefore, this study does not involve a

0–30 cm layer. The lithology of the CQS loess section in this study (30–257 cm thickness) is described as follows (Figure 1): (1) 30–172 cm: this unit is composed of brown weak paleosol, multiferroic manganese film and hard texture, corresponding to the interstadial weak paleosol layer of Malan loess (L1S1) (the L1 loess layer corresponds to the Malan loess in loess stratigraphy); (2) 172–257 cm: the unit consists of brown-yellow loess, and weathered debris of granite bedrock can be seen at the layer, corresponding to the early Malan loess layer (L1L2) [7,18].



**Figure 1.** (a) Location of the Chenqianshan loess section at Shengshan Island in the East China Sea (modified from Cheng [22]) and (b) the alternation of loess and weak paleosol units in the Chenqianshan section.

#### 2.2. Sample Collection and Preparation

Samples for the OSL were light-proof samples obtained by hammering steel tubes into a fresh loess section. Then, the steel tubes with the samples were wrapped using black plastic bags and adhesive tape to avoid light and water loss before testing. A total of 227 bulk samples were obtained at a 1 cm interval. All samples were dried in an oven at 60 °C for 24 h. The samples were ground and separated into <75 µm parts with a dry sieve. They were measured for TOC,  $\delta^{13}C_{org}$ , DRS and geochemical element contents. Unscreened samples were used for magnetic susceptibility analysis.

#### 2.3. Analytical Methods

#### 2.3.1. OSL Dating

Pre-treatment of samples and OSL measurements were carried out at the Luminescence Dating Laboratory of East China Normal University. For OSL dating, quartz grains (38–63  $\mu$ m) were purified and extracted, then etched with H<sub>2</sub>SiF<sub>6</sub> in a dark room under red light and prepared according to the method of Nian et al. [28]. Quartz grains were used to determine equivalent doses by the single-aliquot regenerative dose method [29]. Neutron activation analysis (NAA) of uranium (U), thorium (Th) and potassium (K) was used to determine the radionuclide concentrations of all samples. The water content of the samples was the actual measured water content. Considering the changes in water content during the geological stage, the given absolute error is 7%.

# 2.3.2. TOC and $\delta^{13}C_{org}$ Measurements

All 227 sieved samples were used for TOC and  $\delta^{13}C_{org}$  analyses. Samples (~1.5 g) were pre-treatment with 10 mL of 10% HCl (*v:v*) to remove carbonates. Then, the samples were centrifuged and rinsed with distilled water repeatedly to neutrality, and finally, freezedried and ground again. Subsequently, a MAT-253 stable isotope ratio mass spectrometer was used to measure the TOC and  $\delta^{13}C_{org}$  composition of the samples. The standard deviations of TOC and  $\delta^{13}C_{org}$  composition were less than 0.5% and 0.15%, respectively. All  $\delta^{13}C_{org}$  compositions were reported in the V-PDB (Vienna Pee Dee Belemnite) formula:  $\delta^{13}C_{\%} = [(^{13}C/^{12}C)_{sample}/(^{13}C/^{12}C)_{standard} - 1] \times 1000\%$ .

The method used to calculate the ratio of the C<sub>4</sub> plants was to apply the measured  $\delta^{13}C_{org}$  composition to the isotope mass balance equation [30]:

$$C_4(\%) = \left[ (\delta^{13}C_{\text{org}} - \delta^{13}C_3) / (\delta^{13}C_4 - \delta^{13}C_3) \right] \times 100, \tag{1}$$

where  $\delta^{13}C_{\text{org}}$  was the carbon isotope composition of the CQS loess section,  $\delta^{13}C_3$  and  $\delta^{13}C_4$  represented the average value of the carbon isotope composition from modern  $C_3$  and  $C_4$  plants, respectively. In order to estimate the relative abundance of  $C_4$  plants in the CQS loess section, we took -25% and -12% as the  $\delta^{13}C$  composition for  $C_3$  and  $C_4$  plants, respectively [6].

#### 2.3.3. Magnetic Susceptibility Measurement

The magnetic measurement of the samples referred to the method of Lv et al. [31]. An amount of ~5.0 g of the samples was weighed with precision scales and placed into the magnetic plastic boxes (2 × 2 × 2 cm<sup>3</sup>). The low-frequency magnetic susceptibility ( $\chi_{lf}$ ) (0.47 kHz) and high-frequency magnetic susceptibility ( $\chi_{hf}$ ) (4.7 kHz) were measured by the Bartington MS2B dual-frequency magnetic susceptibility meter and calculated. Then, frequency-dependent susceptibility was defined as  $\chi_{fd\%} = (\chi_{lf} - \chi_{hf})/\chi_{lf} \times 100\%$ .

#### 2.3.4. Diffuse Reflectance Spectra (DRS) Measurement

The sieved samples were put into a plastic ring with a diameter of 4 cm (under a pressure of 15 MPa) to make the test loess piece. The reflectivities of 227 samples were measured by a Perkin Elmer Lambda 950 spectrophotometer. The measured wavelength range was 400–700 nm, and the measurement interval was 2 nm. The reflectivity percentage of the standard color bands was obtained by analyzing the measured reflectivity data [27]. The reflectance percentage of the color band referred to the ratio of the reflectance percentage in a wavelength band to the total visible wavelength reflectance in the sample [32]. We calculated hematite and goethite concentrations in loess samples according to the method of Cheng et al. [27]. The loess samples extracted by the CBD experimental method were used as matrices. Then, standard goethite (Gt) (Hover Color Corp: Arlington Heights, IL, USA, SY610), hematite (Hm) (Bayferrox Corp: Köln, Germany, R4399) and Al(NO<sub>3</sub>)·9H<sub>2</sub>O powders with known contents were artificially added into the matrices, respectively. Finally, a series of aluminum substituted standard samples were synthesized (n = 20). After calculation, the linear regression models for the hematite (Hm) and goethite (Gt) contents and characteristic peak intensities were:

$$Hm\% = 2.766 \times (I_{575} + 0.007 \times Hm(Al \ mol\%) - 0.002) - 0.077$$
(2)

$$Gt\% = 13.404 \times (I_{535} + 0.002 \times Gt(Al \ mol\%) + 0.071 \times Hm\% + 0.002) - 1.096$$
(3)

where  $I_{575}$  and  $I_{535}$  were the peak intensities corresponding to the characteristic peak for Hm and the main peak for Gt in the first derivative diffuse reflectance spectra of natural samples, respectively. Hm (Al mol%) and Gt (Al mol%) were the aluminum substitution amounts of Hm and Gt in loess samples obtained by XRD tests and calculations. Finally, the ratio of Hm/(Hm + Gt) was calculated according to the values of Hm and Gt.

#### 2.3.5. Geochemical Elemental Composition Measurement

A total of 227 sieved samples were used for the geochemical elemental composition measurement. The contents of Fe<sub>2</sub>O<sub>3</sub>, Na<sub>2</sub>O, CaO, K<sub>2</sub>O and Al<sub>2</sub>O<sub>3</sub> in the samples were determined with a SHIMADZU1800 X-ray fluorescence spectrometer. The analytical uncertainties of all elements are within 5%. The chemical index of alteration (CIA) was calculated as CIA =  $[Al_2O_3/(Al_2O_3 + CaO^* + Na_2O + K_2O)] \times 100$  (molar ratio), where CaO\* represents the amount of CaO in the silicate fraction of the sample [33,34].

#### 3. Results

#### 3.1. Chronology of the CQS Loess

The dating results for the six loess deposit samples obtained from different depths of the CQS section are shown in Table 1. The ages of OSL in the CQS loess profile indicated that the age ranges of the loess deposition section were from 75.9  $\pm$  7.5 ka (depth ~ 230 cm) to 39.7  $\pm$  3.4 ka (depth ~ 50 cm). The generalized age-depth model adopted in the whole loess section is presented in Figure 2. It demonstrated different sedimentation rates during the LG. The sedimentation rate from 75.9  $\pm$  7.5 ka to 67.8  $\pm$  6.2 ka was calculated to be ~ 3.70 cm·ka<sup>-1</sup>. It was observed that the sedimentation rate increased significantly to over ~ 10.26 cm·ka<sup>-1</sup> between 67.8  $\pm$  6.2 ka and 63.9  $\pm$  5.8 ka (depth ~ 200 to ~160 cm), and then it decreased from ~ 5.08 cm·ka<sup>-1</sup> between 63.9  $\pm$  5.8 ka and 58.0  $\pm$  5.3 ka to ~3.23 cm·ka<sup>-1</sup> from 48.9  $\pm$  4.6 ka to 39.7  $\pm$  3.4 ka. No obvious erosion-deposition discontinuity was found in the CQS loess section during the field investigations; therefore, it was inferred that the loess stratigraphy with a thickness of 257 cm constituted aeolian deposition, and accumulation was continuous.



Figure 2. Age distribution of the CQS loess section.

Sample	Depth (cm)	K (%)	Th (ppm)	U (ppm)	DOS (Gy/Ka)	De (Gy)	OSL Age (ka)
CQS-1	50	$1.69\pm0.04$	$14.44\pm0.80$	$2.71\pm0.40$	$3.29\pm0.25$	$130.5\pm5.1$	$39.66\pm3.4$
CQS-2	80	$1.68\pm0.04$	$14.43\pm0.80$	$2.38\pm0.30$	$3.16\pm0.23$	$154.4\pm9.0$	$48.9\pm4.6$
CQS-3	130	$1.69\pm0.04$	$13.65\pm0.80$	$2.31\pm0.30$	$3.05\pm0.23$	$177.3\pm9.4$	$58.0\pm5.3$
CQS-4	160	$1.72\pm0.04$	$14.14\pm0.80$	$2.62\pm0.40$	$3.17\pm0.24$	$202.5\pm9.9$	$63.9\pm5.8$
CQS-5	200	$1.80\pm0.04$	$14.14\pm0.80$	$2.67\pm0.40$	$3.19\pm0.24$	$216.4 \pm 11.0$	$67.8\pm6.2$
CQS-6	230	$1.82\pm0.04$	$15.46\pm0.80$	$2.77\pm0.40$	$3.22\pm0.24$	$244.3 \pm 15.8$	$75.9\pm7.5$

Table 1. Results for OSL ages.

# 3.2. TOC, $\delta^{13}C_{org}$ and $\chi_{fd}$ Characteristics

Figure 3 shows the results for the  $\delta^{13}C_{org}$  composition, TOC concentration,  $C_4$  abundance and  $\chi_{fd}$  of the CQS loess section. The values of all indicators showed similar variations with depth. In the weak paleosol layer (L1S1), the values of all indicators were higher than in the loess layer (L1L2) (Figure 3). The TOC concentration for the loess section ranged from 0.11% to 0.47%, and the average was 0.24% (Figure 3a). The  $\delta^{13}C_{org}$  composition varied between 24.56‰ and -20.80%, and the average was -22.19% (Figure 3b). The value of  $\delta^{13}C_{org}$  in the weak paleosol layer (L1S1) gradually increased with increasing depth, while the value of  $\delta^{13}C_{org}$  in the loess layer (L1L2) gradually decreased with increasing depth. The minimum value of  $\delta^{13}C_{org}$  is displayed in the loess layer (L1L2). Figure 3c shows the variation of the relative abundance of  $C_4$  plants in the CQS loess section. The value of  $\chi_{fd}$  for the CQS loess deposits showed fluctuation (Figure 3d). The variation range of  $\chi_{fd}$  for the CQS loess deposits showed fluctuation (Figure 3d). The variation range of  $\chi_{fd}$  was from 3.40  $\times 10^{-8}$  m<sup>3</sup>·kg<sup>-1</sup> to 13.92  $\times 10^{-8}$  m<sup>3</sup>·kg<sup>-1</sup>, and the average value was 9.70  $\times 10^{-8}$  m<sup>3</sup>·kg<sup>-1</sup>. The highest value of  $\chi_{fd}$  appeared in the L1S1 layer.



Figure 3. Variations in (a) TOC, (b)  $\delta^{13}C_{org}$  composition, (c)  $C_4$  abundance and (d)  $\chi_{fd}$  of the CQS section.

#### 3.3. Geochemistry and Diffuse Reflectance Spectra (DRS) and Hm/(Hm + Gt) Ratio Characteristics

Figure 4 shows that the value range for the Fe<sub>2</sub>O<sub>3</sub> concentration in the CQS loess section was 6.04% ~ 6.60%, with an average of 6.25%. CIA did not change much in the whole section; it ranged from 0.81 to 0.83 with an average of 0.82. The Fe<sub>2</sub>O<sub>3</sub> and CIA of the weak paleosol layer (L1S1) were basically the same as those of the loess layer (L1L2), indicating that the Shengshan Island area was weakly pedogenic in the later stages and had underdeveloped paleosol horizons [35]. The formation of hematite (Hm) and goethite (Gt) indicated that some dry and wet climates prevailed at that time [36–38]. The Hm/Gt ratio in the CQS loess section was 0.37~0.47, with an average of 0.42 (Figure 4c). The Hm/(Hm + Gt) ratio in the CQS loess section was 0.27~0.34, with an average of 0.29 (Figure 4d). The curves for

Hm/Gt and Hm/(Hm + Gt) of the L1S1 layer from the CQS loess section were higher than that of the L1L2 layer. The curves for Hm/Gt and Hm/(Hm + Gt) of the CQS loess section were similar to those for the  $\delta^{13}C_{org}$  composition, TOC concentration, C<sub>4</sub> abundance and  $\chi_{fd}$  (Figures 3a–d and 4c,d).



Figure 4. Vertical distribution characteristics of (a)  $Fe_2O_3$  concentration, (b) CIA, (c) Hm/Gt ratio and (d) Hm/(Hm + Gt) ratio for the CQS section.

#### 4. Discussion

### 4.1. Glacial-Interglacial Variability in $\delta^{13}C_{org}$

As an index of vegetation coverage, TOC was often used to reflect the variation of vegetation biomass from the study area in loess studies [39]. The  $\delta^{13}C_{org}$  composition obtained from the loess-paleosol sequences derived from terrestrial higher plants has been used to reconstruct the variation in  $C_3/C_4$  relative abundance in past geological history periods [4–6]. The values of  $\delta^{13}C_{org}$  composition and  $C_4$  abundance indicated that the climate was colder and drier during the L1L2 (MIS 4) stage than during the L1S1 (MIS 3) stage [19–21]. The growth tendency of  $C_4$  plants was more advantageous in the humid and warm climate and increased rainfall conditions [6,11,39–41]. Magnetic susceptibility was commonly used as the proxy index for precipitation [42,43]. Previous studies have confirmed that there was a positive correlation between rainfall and frequency-dependent susceptibility ( $\chi_{fd}$ ) through experiments on modern surface soil [44]. The  $\chi_{fd}$  for the CQS loess section was taken as an indicator of the precipitation change over the past 76~39 ka (Figure 3d). Previous studies have shown that there was a positive correlation between the Hm/(Hm + Gt) ratio and temperature for soils in the study area [45]. The  $\delta^{13}C_{org}$  and  $C_4$ abundance in the CQS profile revealed similarities with changes of  $\chi_{fd}$  and Hm/(Hm + Gt) (Figures 3b–d and 4d). The change of  $\delta^{13}C_{org}$  composition recorded the regional differences of local plants and climate change.

Previous studies have shown that the distribution ranges of the  $\delta^{13}C_{org}$  of  $C_3$  plants and  $C_4$  plants were ~ 22‰ to -30‰ and ~ -9‰ to -19‰, respectively [1,2]. The average value of the  $\delta^{13}C_{org}$  in the CQS loess section was -22‰. This indicated that  $C_3$  plants were one of the vegetations in the CQS area. Rao et al. [12] studied the modern surface soil  $\delta^{13}C_{TOC}$  data of eastern China, Australia and the Great Plains of North America around the North Pacific, and found that  $C_4$  plants could grow when the mean annual temperature was higher than 12 °C. The current mean annual temperature of CQS in Shengshan Island is 15~17 °C, which is significantly higher than 12 °C. This showed that the vegetation of CQS contained  $C_4$  vegetation. The average value of  $C_4$  relative abundance in the CQS area was only 21.31%, and it indicated that  $C_4$  plants were not the main vegetation in this area. To sum up, the  $\delta^{13}C_{org}$  composition for the CQS loess section revealed that  $C_3/C_4$  mixed vegetation was the main vegetation in this study area, among which  $C_3$  plants were the dominant vegetation during the LG.

# 4.2. Comparison between $\delta^{13}C_{org}$ Record of the CQS Loess Section and Other Existing $\delta^{13}C_{org}$ Records from Central and Northern China

Figure 5 presents the comparisons of the  $\delta^{13}C_{\rm org}$  records for the CQS loess section with other existing  $\delta^{13}C_{org}$  records for the China Loess Plateau (CLP) [7,37,46] and Tianshan Mountains [16] and also displays the marine oxygen isotope stages record [47]. Except for the Yuanbao loess section west of the CLP and the Axike loess section north of the Tianshan Mountains, the curves of  $\delta^{13}C_{org}$  composition for the CQS, Luochuan, Lantian and Xunyi loess sections showed a similar trend (Figure 5a–d). These results indicated that central and eastern China have been equally affected by the cyclic changes during the LG. Notably, during the MIS 4 stage, the values of  $\delta^{13}C_{org}$  composition for the CQS, Luochuan, Lantian and Xunyi loess sections were more negative than those for the MIS 3 stage (Figure 5a–d). The values of  $\delta^{13}C_{org}$  composition for the Yuanbao loess section west of the CLP and the Axike loess section north of the Tianshan Mountains in the L1S1 (MIS 3) stage were more negative than those in the L1L2 (MIS 4) stage (Figure 5e,f). The values of  $\delta^{13}C_{org}$  composition of the CQS, Luochuan, Lantian and Xunyi loess sections were different, however, the  $\delta^{13}C_{org}$  curves in the four loess sections are similar during the LG (Figure 5a-d,g). This indicated that the vegetation succession in the Shengshan Island area, under the impact of climatic change during the LG, was synchronized with the existing records previously reported from central China [40,46].



**Figure 5.** Vertical distribution characteristics of (a)  $\delta^{13}C_{\text{org}}$  records of CQS section; (b) Luochuan loess section [40]; (c) Lantian loess section [40]; (d) Xunyi loess section [46]; (e) Yuanbao loess section [7]; (f) Axike loess section [16]; (g) Marine oxygen isotope records [47].

Generally, the variations of the  $\delta^{13}C_{org}$  records in six loess profiles also reflected the cycle changes of the climate during the LG. At the same time, when discussing the variations of the  $\delta^{13}C_{org}$  composition in different areas during the LG, the differences in regional climate in different regions should be considered, such as elevation, seasonal precipitation and temperature [48,49]. Climatic change has a significant impact on regional vegetation and the relative biomass of C<sub>3</sub> and C<sub>4</sub> plants.

# 4.3. Environmental Factors Driving $\delta^{13}C_{\text{org}}$ Changes during the Last Glacial Period

As is known to all, the most important external factors, such as precipitation, temperature and atmospheric CO<sub>2</sub>, affect changes in the  $\delta^{13}C_{org}$  composition of loess [50–52]. As shown in Figure 6, in order to discuss the environmental mechanism driving the variations of  $\delta^{13}C_{org}$  composition for the CQS loess section from the perspective of external environmental factors, we compared the  $\delta^{13}C_{org}$  records of the CQS section, C<sub>4</sub> abundance in the CQS section, the  $\chi_{fd}$  in the CQS section, the oxygen isotope record from Chinese stalagmite [53], the East Asian Summer Monsoon record [54], ocean temperature records from Antarctica Dome C ice cores [6,55], the Greenland Ice-core Project (GRIP) marine oxygen isotope records [56] and the atmospheric CO<sub>2</sub> concentration recorded in the Antarctic ice cores [57].



**Figure 6.** (a) The  $\delta^{13}C_{org}$  record of the CQS section; (b) C<sub>4</sub> abundance in the CQS section; (c)  $\chi_{fd}$  of the CQS section; (d) oxygen isotope records from Chinese stalagmite [53]; (e) the East Asian summer monsoon records [54]; (f) ocean temperature records from Antarctica Dome C ice cores [6,55]; (g) GRIP marine oxygen isotope records [56]; (h) atmospheric CO<sub>2</sub> concentration recorded by Antarctic ice cores [57].

Precipitation played an important role in the paleovegetation of the East China Sea islands. The  $\chi_{fd}$  of the CQS loess section was higher in the MIS 3 stage than in the MIS 4 stage (Figure 6c,e), suggesting that precipitation was significantly strengthened in the MIS 3 stage. The high precipitation level inferred from  $\chi_{fd}$  was positively correlated with the  $\delta^{13}C_{org}$  composition of the CQS loess section during the warm-humid stage (MIS 3) (Figure 3). The  $\chi_{fd}$  was mainly affected by iron oxide minerals, which were usually generated in the warm, humid with strong pedogenesis and weathering environment. The precipitation in the CQS study area could increase with the strengthening of the East Asian summer monsoon during the L1S1 (MIS 3) stage. Some researchers considered that the  $\delta^{18}$ O values of Chinese stalagmites could indicate the summer monsoon precipitation and monsoon intensity [58–60]. The more negative  $\delta^{18}$ O values for stalagmites indicated stronger Asian monsoons and more rainfall [53]. The  $\delta^{18}$ O record of the stalagmites was basically consistent with the  $\delta^{13}C_{org}$  composition of the CQS loess section (Figure 6a,d). Thus, precipitation was one of the main driving factors of variations of  $\delta^{13}C_{org}$  composition in the CQS loess section.

The effect of temperature on the  $\delta^{13}C_{org}$  composition was also very complex. High temperature could advance the activity of plant enzymes, enhance the photosynthetic efficiency of plants, and thus promote the growth of plants [1].  $C_4$  plants generally had a relatively competitive growth advantage in high-temperature environments [3]. The curve changes of the Hm/(Hm + Gt) ratio, the  $\delta^{13}C_{org}$  composition and C<sub>4</sub> abundance from the CQS loess profile were similar, and it indicated that temperature had a significant impact on the change of relative abundance of C4 plants on the East China Sea Island (Figure 6a–c). Moreover, the positive similarity among  $C_4$  abundance of the CQS loess section, the ocean temperature records of the Antarctica Dome C ice cores and the GRIP marine oxygen isotope records, indicated that the change of  $C_4$  abundance was closely related to temperature (Figure 6b,f,g). Zhang et al. [61] considered that a low temperature was beneficial to the growth of  $C_3$  plants by studying the  $\delta^{13}C$  composition of n-alkanes of CLP. Rao et al. [62] studied the differences in the soil organic carbon isotope records of three typical regions in the middle latitudes of the northern hemisphere during the LG and considered that the high temperature in the growing season had an obvious effect on the growth of C<sub>4</sub> plants, and at the same time, the temperature range had a certain threshold. To sum up, the temperature was also one of the important factors to control the  $\delta^{13}C_{org}$ composition and C<sub>4</sub> abundance of the CQS loess deposition.

Carbon isotope composition was the result of  $C_3$  and  $C_4$  photosynthesis of atmospheric  $CO_2$  by plants [7]. Previous studies indicated that a higher atmospheric  $CO_2$  concentration was more beneficial to the growth of  $C_3$  plants, while a lower atmospheric  $CO_2$  concentration was more beneficial to the growth of  $C_4$  plants [6,7,63]. Feng and Epstein [64] quantitatively reconstructed the atmospheric  $CO_2$  concentration by using the  $\delta^{13}C$  composition of tree rings; the  $\delta^{13}C$  composition of tree rings decreased by  $2.0\% \pm 0.1\%$  for every 100 ppm increase in atmospheric  $CO_2$  concentration. It indicated that the  $\delta^{13}C$  composition decreased with an increase in atmospheric  $CO_2$  concentration. Figure 6a,h shows that there was no negative correlation between atmospheric  $CO_2$  concentrations and the  $\delta^{13}C_{\text{org}}$  record of the CQS loess profile. In general, the change of atmospheric  $CO_2$  concentration was unlikely to drive the change of  $\delta^{13}C_{\text{org}}$  composition in the CQS loess deposition during the LG.

#### 5. Conclusions

In this paper, we investigated the optically stimulated luminescence (OSL) ages and the variations in the frequency-dependent susceptibility, diffuse reflectance spectra, TOC concentration and  $\delta^{13}C_{org}$  composition of the Chenqianshan loess section on Shengshan Island, East China Sea, and discussed the paleoenvironmental significance of these indices and environmental influencing factors. The results of TOC,  $\delta^{13}C_{org}$ ,  $\chi_{fd}$ , and Hm/(Hm + Gt) for the CQS profile displayed a similar pattern. The TOC concentration for the CQS section varied between 0.11% and 0.47%, and the  $\delta^{13}C_{org}$  composition of the CQS loess

section varied between -20.80% and -24.56% during the last glacial period. The  $\delta^{13}C_{org}$  composition of the loess layer during the L1L2 (MIS 4) stage was lower than those of the weak paleosol layer during the L1S1 (MIS 3) stage, showing the transition of climate from dry-cold to warm-humid. The average value of C<sub>4</sub> abundance was 21.31%. It indicated that the vegetation of CQS loess deposits was mainly C<sub>3</sub>/C<sub>4</sub> mixed vegetation, and C<sub>3</sub> vegetation was the primary vegetation. The  $\delta^{13}C_{org}$  composition of the CQS loess section could more likely indicate the evolution of vegetation and climate change than other proxy indicators. The comparison between the  $\delta^{13}C_{org}$  curve of the CQS section and other existing  $\delta^{13}C_{org}$  records of the loess sections from central and northern China showed similar trends, and their vegetation successions exhibit synchronous changes during the LG. Based on a comparison of the  $\delta^{13}C_{org}$  record, C<sub>4</sub> abundance and  $\chi_{fd}$  of the CQS section and temperature caused the change of paleovegetation in loess deposits on islands in the East China Sea during the LG.

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