

### Carbon isotopic composition and its implications on paleoclimate of the underground ancient forest ecosystem in Sihui, Guangdong

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We present the carbon isotopic composition of the total organic carbon (TOC) and fine roots in the sedimentary profile from the underground ancient forest in Sihui to study the climatic and environmental changes from 4.5 ka BP to 0.6 ka BP. Results show that  $C_3$  plant was the main vegetation from 4.5 ka BP to 0.6 ka BP in this region. The ancient forest began to develop in the wetland at around 4 ka BP and disappeared together with the wetland at about 3.0 ka BP, implying that the climate had changed greatly at around 3.0 ka BP. As indicated by the simulation results, the content of atmospheric  $CO_2$  increased slightly during 3.5 ka BP and 3.0 ka BP, implying climate warming during that period. The interval of radiocarbon age between 3.0 ka BP and 1.2 ka BP was possibly caused by the strong erosion when the block was lifted in the neotectonic movement. From 1.2 ka BP to 0.6 ka BP, the region remained in terrestrial sedimentary environment, and the surface plant biomass declined gradually. Drought caused by the climate change was the likely cause for the disappearance of the ancient forest. South transition of Intertropical Convergence Zone (ITCZ) was probably the main mechanism for the climate change.

Holocene, underground ancient forest, Pearl River Delta, fine root, atmospheric CO2

Studying climate variation of Holocene helps us better understand the relationship between human activities and natural factors<sup>[1–6]</sup>. At present, researches on the climate change of Holocene in China concentrate on three periods<sup>[7–10]</sup>: the early Holocene with temperature climbing, warm period in the middle Holocene, and the late Holocene with temperature decreasing. Results from the lake and cave sediments in the low latitude regions around the world showed that the climate turned cold and dry with a sharp decrease in precipitation from 3.2 ka BP to 2.7 ka BP<sup>[1,11–13]</sup>. This event may have greatly changed the ecosystem and activities of human in Pearl River Delta. It may be also one of the reasons for the termination of the Shell Mound Sites and Sand Mound Sites. Many ancient forests were found in Sihui, and the dominate tree species was hydrophilous plant *Glyptostrobus pensilis*. The evolutionary history of the ancient forests offers us an important way to reconstruct the climate and geographical environment of Pearl River Delta. However, we still know litter about the chronostratigraphy of the ancient forests and their indications on paleoclimate. Li et al.<sup>[14]</sup> has first studied the ancient

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timbers buried under Pearl River Delta, and con-sidered that the death of the timbers was associated with the Litter Ice Age. Recent study has suggested that over-cutting by the ancient people was probably one of the reasons for the death of the ancient timbers<sup>[15]</sup>.

In this paper, we try to decipher the evolutionary history of the climate and geographical environment from 4.5 ka BP to 0.6 ka BP based on the content of TOC, the carbon isotopes of TOC, and fine roots. We also discuss the mechanism for the climatic and environmental changes and their impact on the human activities.

### 1 Sampling and method

The sedimentary profile of the ancient forest is located in Longfu, the central part of Sihui, Guangdong Province. It also lies in the northwest of Pearl River Delta and lower reaches of Xijiang River, Beijiang River and Suihe River. The geographic location is 23°22'359"N and 112°42'497"E. The geography of Sihui is characterized by mountains in the north and west, alluvial plains in the south and east, and hills and valley basin in the central (Figure 1). Current mean annual temperature (MAT) is 21°C, and main annual precipitation (MAP) is 1800 mm in the region. It belongs to subtropical monsoon climate and is strongly influenced by East Asian Monsoon. The subtropical monsoon evergreen broadleaf forest is the main vegetation.

The sedimentary profile is 4.8 m high, and consists of 4 layers with clear boundary lines: the coarse aleuritic clay layer (layer A), the fine aleuritic clay layer (layer B), the humic layer (layer C) and the aleuritic clay layer (layer D). Sampling interval in layers C and D is 20 cm. Detailed distributions of the sampling sites in layers A and B are showed in Figure 2.



Figure 1 Location map of the sampling site, denoted by tree sign.

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Figure 2 Radiocarbon chronostratigraphy and distributions of the sampling sites in layers A and B.

For the method for the preparation of fine root<sup>[15]</sup>. All the preparations of sediments and fine roots were carried out at the AMS-<sup>14</sup>C laboratory in the Key Laboratory of Isotope Geochronology and Geochemistry, Chinese Academy of Sciences (CAS). The  $\delta^{13}$ C values of CO<sub>2</sub> transformed from TOC were measured by Finigan Model-251 isotopic ratio mass spectrometer facility (PDB standard) with a precision of ±0.02‰ at the State Key Laboratory of Loess and Quaternary Geology. Graphite targets for <sup>14</sup>C dating were measured at the Key Laboratory of Heavy Ion Physics, Peking University, AMS facility. The contents of TOC were determined by a microbarograph in the vacuum system in our laboratory.

### 2 Results and discussion

# 2.1 Distribution of $\delta^{13}$ C values in the sedimentary profile

Changes of  $\delta^{13}$ C values can reflect the variation of the plant types and the decomposition state of the organic carbon<sup>[16,17]</sup>, which is expressed by  $\delta^{13}$ C=(( $^{13}$ C/ $^{12}$ C)<sub>sample</sub>/( $^{13}$ C/ $^{12}$ C)<sub>standard</sub>-1)×1000‰. Based on the different pathways of photosynthesis, the plant can be divided into three different types: C<sub>3</sub>, C<sub>4</sub> and CAM. Each type

has a different range of  $\delta^{13}$ C values: -34% - 23% for C<sub>3</sub> plant, -22% - 6% for C<sub>4</sub> plant, and -20% - 10% for CAM plant<sup>[18]</sup>. In the sedimentary profile, the  $\delta^{13}$ C values varied between -29.85% and -25.58%, all falling in the range of C<sub>3</sub> plant, suggesting that the main vegetation was C<sub>3</sub> plant from 4.5 ka BP to 0.6 ka BP in this region.

Figure 3 shows that the  $\delta^{13}$ C values increased with the depth exponentially from -28.99% to -25.58% in the layers A and B, exhibiting a variation of 3.4‰, which was likely caused by the fractionation when the organic carbon was decomposed. Plant change may also lead to the variation of  $\delta^{13}$ C values, which is revealed by the great fluctuation of the  $\delta^{13}$ C values of the sediments<sup>[19]</sup>. However, the  $\delta^{13}$ C values in layers A and B had not exhibited such a fluctuation. Therefore, the variation of the  $\delta^{13}$ C values in layers A and B only reflected the decomposition degree of the organic carbon.

Given the precision of modern mass spectrograph ( $\pm 0.02\%$ ) and the error produced during the sample preparation ( $\pm 0.2\%$ ), the  $\delta^{13}$ C values remained stable in layer C: -29.04‰ and -29.85‰, suggesting that the organic carbon almost remained undecomposed, and the



**Figure 3** Variation of  $\delta^{13}$ C values with depth and age in the sedimentary profile.

sedimentary environment kept reductive from  $3813\pm31$ a BP to  $3068\pm35$  a BP. Similar to layer C, the  $\delta^{13}$ C values in layer D ranged from -29.5% to -29.3%, indicating that the organic carbon may partly come from layer C. The obviously distinction of the  $\delta^{13}$ C values among layers A, B, and C implied that great ecological change happened around 3.0 ka BP, which finally led to the disappearance of the original ecosystem.

### 2.2 Distribution of TOC in the sedimentary profile

Contrary to decreasing with age and depth in the typical soil profile<sup>[20,21]</sup>, the content of TOC increased exponentially from 0.8% to 5.5% in layers A and B (Figure 4(a) and (b)). Two causes may be responsible for it: 1) The amount of organic matters entering the layers A and B declined with time; 2) Organic matters in layers A and B were partly transferred from layer C. However, a 4.0% variation of the  $\delta^{13}$ C values between the bottom of layer B and layer C suggested that the organic matters in layers in layers A and B were from layer C. Therefore, the organic matters in layers A and B were from the surface soil of the surroundings, and the variation of the content of TOC stood for the change of the vegetation density, which suggested that the biomass of the surface vegetation had a decreasing tendency from 1.2 ka BP to 0.6 ka BP in this region.

Layer C in which the ancient forest developed had the <sup>14</sup>C chronology ranging from 3813±31 a BP to 3068±35 a BP. The content of TOC fluctuated between 33.4% and 49.0%, showing no obvious trends, which implied that the vegetation composition and sedimentary environment stayed stable and kept a rather reductive environment in this region during the period. Because



Figure 4 Distribution of TOC content in the sedimentary profile and Shuangchi Maar Lake with C/N ratio. (a), (b) Variation of TOC content with depth and age in the sedimentary profile; (c), (d) variation of TOC content and C/N ratio with depth in Shuangchi Maar Lake in Hainan.

*Glyptostrobus pensilis* tends to grow along the river bank or in wetland<sup>[22]</sup>, the region was most likely to be wetland with a warm and wet climate during that period. In layer D, the basement of the ancient forest, the content of TOC decreased from 14.5% to 6.7% from  $4544\pm30$  a BP to  $3813\pm31$  a BP. Given the combination of the sedimentary property and the probable source of organic carbon, such low content of TOC in layer D could not be formed in the wetland. This suggests that the region could not be a wetland between 4.5 ka BP and 4.2 ka BP and that the wetland should initiate between 4.2 ka BP and 3.8 ka BP.

As shown in Figure 2, the properties of the sediments in layers B and C changed obviously. The change also exhibited in the radiocarbon chronology, the content of TOC and the  $\delta^{13}$ C values, which reflected the distinguished difference of ecological environment before and after 3.0 ka BP. The cause of the great interval of radiocarbon age between layers C and B was possibly linked to the neotectonic movement. Sihui lies in the active region of the neotectonic movement, and vertical activities occurred frequently through Holocene<sup>[23]</sup>. When the block was lifted, erosion caused by exogenetic forces would lead to the loss of the stratum between 3068±35 a BP and 1250±29 a BP. In addition, the erosion intensity significantly enhanced around 3.0 ka BP in South China as indicated by the changes of the content of TOC and the ratio of C/N (Figure 4(c) and (d)) in Shuangchi Maar Lake in Hainan<sup>[11]</sup>.

## 2.3 Variation of atmospheric CO<sub>2</sub> concentration during 3.5–3.0 ka BP

Many proxies recorded the climatic variation of Holocene<sup>[24-28]</sup>. The ice core and stalagmite records are generally acknowledged relatively precise. However, in Pearl River Delta, the main proxies are corals and sediments in lakes<sup>[3,10-11]</sup>. Studies on the tree-rings in this region has not been reported yet.

The fine root can record the climate information when it grows. It is the main organ of plant for the water and nutrient absorption, in which the carbon is produced through photosynthesis<sup>[29]</sup>. The life span of fine root is short, usually ranging from days to months, years at most<sup>[30–34]</sup>. The biomass of fine root tends to concentrate in the litter layer or above 10 centimeters of mineral topsoil, and declines with depth in exponential<sup>[31,35–37]</sup>. The  $\delta^{13}$ C value of the fine root is not influenced by temperature, but mainly controlled by atmospheric CO<sub>2</sub> concentration and the precipitation. Influence of precipitation mainly shows up on the use efficiency of water: high  $\delta^{13}$ C value stands for high use efficiency of water, and the opposite is also true<sup>[38]</sup>. Because of the swampy environment, the water use efficiency of the fine root could not be affected significantly. Variation of  $\delta^{13}$ C value of fine root thus shows the variation of atmospheric CO<sub>2</sub> concentration.

Abundant fine roots with different colors and densities are preserved in layer C. In order to assure that all the fine roots belong to the same period, we collected the samples at the same position with the darkest color and diameter less than 0.5 mm from each site. Thus,  $\delta^{13}$ C values of the mixed fine roots would reflect the average  $\delta^{13}$ C values of atmospheric CO<sub>2</sub> when the fine roots grew.

 $\delta^{13}$ C values of fine roots in the profile ranged between -28.60‰ and -27.26‰ with an average value of -27.84‰ (Figure 5(a)). Marshall et al.<sup>[39]</sup> presented the relationship between the  $\delta^{13}$ C value of plant ( $\delta^{13}$ C<sub>p</sub>) and atmospheric CO<sub>2</sub> ( $\delta^{13}$ C<sub>a</sub>) as follows:

 $\delta^{13}C_p = \delta^{13}C_a - a - (b-a)C_i/C_a$ , (1) *a* stands for the fractionation factor (4.4‰) caused by the diffusion effect when CO<sub>2</sub> pass stomas of plant. *b* denotes <sup>13</sup>CO<sub>2</sub> fractionation value (29‰) when the leaf fix atmospheric CO<sub>2</sub> through photosynthesis<sup>[40]</sup>.  $C_i/C_a$ represents the ratio between CO<sub>2</sub> content ( $C_i$ ) in leaves and atmosphere ( $C_a$ ). For C<sub>3</sub> plant in general, the variation of the atmospheric CO<sub>2</sub> content has litter influence on  $C_i/C_a^{[41]}$ .

In the simulation, we applied the function:

$$\delta^{13}C_{L} = \delta^{13}C_{R.A} - 2.0\%, \qquad (2)$$

where  $\delta^{13}C_L$  stands for the  $\delta^{13}C$  value of *Glyptostrobus* pensilis leaf, and  $\delta^{13}C_{R,A}$  stands for the average  $\delta^{13}C$ value of fine root (-27.84‰). 2.0‰ is the fractional constant of <sup>13</sup>C between the leaves and fine roots of conifer plants<sup>[29]</sup>. The  $\delta^{13}C$  value of atmospheric CO<sub>2</sub> was supposed to be -6.5‰ at that time<sup>[27]</sup>. Inputing these data above into eq. (1), we finally got  $C_i/C_a = 0.77$ . Inputing all the  $\delta^{13}C$  values of fine roots into eq. (3):

 $\delta^{13}C_{R,A} - 2.0\% = \delta^{13}C_a - a - 0.77 \times (b-a).$  (3) Results are shown in Figure 5(b).

Mook et al.<sup>[42]</sup> and Keeling et al.<sup>[43]</sup> have found that fine inverse correlation existed between modern atmospheric CO<sub>2</sub> content and  $\delta^{13}$ C values, expressed by eq.



**Figure 5** Distribution of the  $\delta^{13}$ C values of fine roots, and simulation results of the atmospheric CO<sub>2</sub> content and the  $\delta^{13}$ C values during 3.5 ka BP to 3.0 ka BP. (a) Variation of  $\delta^{13}$ C values of fine roots with the depth; (b) simulation results of the  $\delta^{13}$ C values of atmospheric CO<sub>2</sub> during 3.5 ka BP to 3.0 ka BP; (c) variation of atmospheric CO<sub>2</sub> content according to different equations and the ice core record of Antarctica. A, B, C and D denote to the simulation results of the eqs. (5), (6), (4) and ice core records in Taylor Dome, Antarctica, respectively.  $\diamond$  in the curve C derived from the data of the shadow in curve D.

(4), in which m is a constant, and n denotes the  $\delta^{13}$ C value of plant leaves.

$$[CO_2] = m/(\delta^{13}C_a + n).$$
 (4)

In this paper,  $n = \delta^{13}C_L(-29.84\%)$ ; *m* is determined by the atmospheric CO<sub>2</sub> content (280 ppmv, on average) and the  $\delta^{13}C$  value (-6.5‰, on average) of pre-indus $try^{[44,45]}$ , and the calculated result is 6535.2.

The inverse relationship also showed between the paleo-atmospheric CO<sub>2</sub> content and  $\delta^{13}$ C value. CO<sub>2</sub> trapped in the ice core at Taylor Dome, Antarctica, manifested a fine inverse relationship between atmospheric CO<sub>2</sub> content and  $\delta^{13}$ C values during the past 11 ka BP<sup>[27]</sup>. In addition, the relationship had been applied to reconstruct the paleo-atmospheric CO<sub>2</sub> content successfully. White et al.<sup>[24]</sup> recovered the variation of the atmospheric CO<sub>2</sub> content since 14 ka BP using this relationship, and the results showed great agreement with the records from the ice core.

Based on eqs. (3) and (4), variation of the atmospheric  $CO_2$  content during 3.5 ka BP to 3.0 ka BP was shown in Figure 5(c)-C. In contrast, Figure 5(c)-A and B showed the simulation results of eqs. (5) and (6) given by Keeling et al.<sup>[43]</sup> and Amundson et al.<sup>[46]</sup>:

$$\delta^{13}C_a = -26.54 + 6346/(CO_2), \tag{5}$$

$$\delta^{13}C_a = -28.773 + 7128.5 \times 1/(CO_2).$$
 (6)

Simulation results showed that the  $\delta^{13}$ C values of the atmospheric CO<sub>2</sub> decreased slightly while the content increased during 3.5 ka BP to 3.0 ka BP in this region, reflecting a warming trend. That was consistent with the records in Taylor Dome, Antarctica.

## 3 The event around 3.0 ka BP and its impact on human activities

The geographical environment in this region changed greatly before and after 3.0 ka BP: from 4.0 ka BP to 3.0 ka BP, the region was a wetland and covered by the ancient forest which was dominated by the hydrophilous plant Glyptostrobus pensilis. The climate had a warming trend during that period. At around 3.0 ka BP, the wetland disappeared and the ancient forest was gone, suggesting the geographical environment likely turned dry at that time. Causes for the drought may link to the climate change and the neotectonic movement. Contemporaneous records of pollen data and  $\delta^{18}$ O values of the sediments from lakes in the low latitude areas around the world all implied that the climate changed dramatically at around 3.0 ka BP<sup>[3,11,12,47]</sup>. Haug et al.<sup>[48]</sup> confirmed that the precipitation in Cariaco Basin, South America, declined sharply at 3.0 ka BP.  $\delta^{18}$ O values of the stalagmite from Dongge Cave also recorded the less precipitation and weaker strength of East Asian Summer Monsoon at that time<sup>[49]</sup>. The Titanium content from Huguang Maar Lake in Zhanjiang, near the study area, also displayed the strengthened Winter Monsoon at 3 ka BP. Almost at the same time, the cyclicity and intensity of the ENSO enhanced distinctly, and the ENSO- teleconnected regions were characterized by an increased impact of ENSO<sup>[50]</sup>. The ancient forest and wetland were likely to disappear under such climate conditions. Vertical activity of the neotectonic movement could also lead to hydrological changes in this region. However, the ancient forest had been preserved thanks to rapid burial. If the drought was caused by vertical movement, the ancient forest could not have been buried rapidly. Therefore, the reason for the disappearance of the ancient forest was related to climate changes at around 3.0 ka BP.

Acknowledged mechanisms of the climate change around 3.0 ka BP are associated with the changes of monsoon intensity<sup>[51]</sup>, ENSO activities<sup>[52]</sup>, North Atlantic current<sup>[53]</sup>, and the annual position of Intertropical Convergence Zone (ITCZ)<sup>[3,48]</sup>. For the East Asia at that time, the climate change is likely to be driven by transition of the annual position of ITCZ. When the ITCZ towards south, the summer monsoon intensity would decline while the winter monsoon intensity will increase, and the North Hemisphere turns dry and cold. In addition, the southward transfer of ITCZ could improve the intensity of ENSO activity<sup>[48]</sup>. These are all in agreement with paleoclimate records in this region. Thus, the southward transfer of ITCZ would be the main reason for the climate change in this region.

The climate change around 3.0 ka BP may have greatly impacted on the production and life style of ancient people in this region. Along with the climate change, the sea level decreased<sup>[54,55]</sup>. Traditional gathering and fishing could not satisfy the need of life, as a result, agriculture especially rice planting flourished<sup>[4]</sup>. Maybe the same reason led to the termination of the Shell Mound Sites and Sand Mound Sites at around 3.0 ka BP in Pearl River Delta.

### 4 Results

(1) The ancient forest in Sihui began to develop at around 4.0 ka BP, and disappeared at about 3.0 ka BP. The region was a wetland, and climate stayed warm and wet during this period.

(2) The region was dominated by  $C_3$  plant during 4.5 ka BP to 0.6 ka BP, however, the biomass of the surface vegetation declined during 1.2 ka BP to 0.6 ka BP. The organic carbon in layer C almost remained undecomposed.

(3) Simulation results showed that the atmospheric  $CO_2$  content increased slightly during 3.5 ka BP to 3.0 ka BP, consistent with the records of the ice core in Antarctica, indicating a warming trend during the pe-

#### riod.

(4) Disappearance of the ancient forest and wetland was likely to associate with the drought at around 3.0 ka BP. The southward transfer of ITCZ was possibly the

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### main mechanism of the climate change.

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