# Variations in radiocarbon ages of various organic fractions in core sediments from Erhai Lake, SW China

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Radiocarbon dating was performed for the extracted organic fractions (cellulose-rich and humic acid fractions of plant fragment; fulvic acid, humic acid and humin fractions of humus substance) and shell from core sediments of the Erhai Lake, SW China. The <sup>14</sup>C dating results reveal that there are considerable differences, but there apparently is a humic acid  $\leq$  humin < fulvic acid fraction sequence of <sup>14</sup>C age increase. The variability in radiocarbon ages of organic fraction of lake sediment suggests that special caution is necessary when radiocarbon ages of bulk sediments are used.

The linear correlation between <sup>14</sup>C age of allochthonous terrestrial macrofossil (plant fragment and shell) and depth indicates roughly a constant sedimentation rate of *ca*. 0.7 mm yr<sup>-1</sup> in central Erhai Lake since 4500 yr BP. The <sup>14</sup>C ages of the autochthonous humic acid fraction are 210~4800 yr shift from "the true <sup>14</sup>C age" obtained by interpolating the corresponding horizontal level to the above <sup>14</sup>C age-depth correlation. Such age difference may be alternatively attributed to a uniform reservoir effect (most likely *ca*. 300 yr). The period with large <sup>14</sup>C age shift synchronizes with the period of changes in  $\delta^{13}$ C and ARM intensity and ARM/susceptibility values.

#### INTRODUCTION

Materials for <sup>14</sup>C dating in lake sediment may include various fractions: 1) macrofossils of terrestrial plants; 2) aquatic organic matter which is formed mainly in the shallow part of the lake water and consists of phytoplankton, aquatic macrophytes and *Characeae*; 3) authigenic inorganic carbonate (marl, tufa, stromatolites) which is precipitated from the total dissolved inorganic carbon (TDIC) of the shallowest lake water. Because the source of carbon for these components is different, the corresponding initial <sup>14</sup>C values for sediment samples may deviate considerably from each other and must be interpreted separately. Among those components, macrofossils of terrestrial plants provide the most accurate <sup>14</sup>C dates without the requirement of a reservoir correction, provided the samples were not reworked. However, terrestrial macrofossils and typical tephra layer are sparse in some lake sediments, in particular lakes in China. Hence, an estimate of the radiocarbon age was generally obtained from the aquatic organic matter (bulk lake sediments) or authigenic inorganic carbonate (e.g., Zhang *et al.*, 1998; Eisma *et al.*, 2000). Although the <sup>14</sup>C method has been used successfully to date bulk lake sediments (e.g., Benson *et al.*, 1997; Lowe *et* 

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Fig. 1. Location map showing the sites of cores ER1, ER3 and ER24 in Erhai Lake, Yunnan Province, SW China.

*al.*, 1997; Sveinbjörnsdóttir *et al.*, 1998), the <sup>14</sup>C dates of bulk lake sediments may sometimes be contradictory with sedimentary sequence. In addition to the problem of contamination, there is considerable uncertainty regarding whether a particular organic fraction of sediments yields a more reliable <sup>14</sup>C age than other fractions. Most laboratories date either the NaOH-soluble (humic acid or humate), NaOH-insoluble (humin or residue),

or the total fraction. The literature affords no consensus about which fraction this is. In order to evaluate the validity of the humus substances in bulk sediment, this study essentially compares the <sup>14</sup>C results of three extracted fractions of humus substances with those of the macrofossils of terrestrial plants and shell collected from three core sediments in Erhai Lake, SW China.

The Erhai Lake (24°35'~58' N and 100°05'~17'



Fig. 2. Lithologies for sediment cores ER1, ER3 and ER24 from Erhai Lake. <sup>14</sup>C dates shown as Italic font are from Hyodo et al. (1999) and others are from this study.

E), located in the Dali basin, is the second largest lake on the Yunnan Plateau, southwest China (Fig. 1). It is a large (249 km<sup>2</sup>), shallow lake ( $z_{max} = 20.7$ m) that lies 1974 m above mean sea level (m asl). From the west and north where high mountains stand up 2800~3500 m asl, about 18 rivers and streams flow into the lake. There is only one outlet (to the west in the southwest corner of the lake near Xiaguan). Erhai Lake is fresh (degree of mineralization = 170~172 mg L<sup>-1</sup>). Its pH is generally between 8.4 and 8.5 (Zhu, 1989).

#### SAMPLES AND EXPERIMENTS

Sediment core ER1 (241 cm long) was taken from a flat-bottomed site 10 m deep in southern part of the lake (Figs. 1 and 2). The sediment material changed from clay to silt at the base. Cores ER3 and ER24 were collected from the central part of the lake, about 20 km northwest of the ER1 site. Both sites are 20 m deep, which is the maximum depth of the lake. Core ER3 was 316 cm long, consisting of clay above the 217 cm-deep level and silt below it. Core ER24 was 120 cm long. The sediment material consists of clay, corresponding to the upper part (0~217 cm) of ER3. There is a change of visible color and particle size: light brown above 70 cm and gray below it. However, such a lithological boundary is less clear in core ER1 and ER3. A total of nine bulk sediment samples were collected from different depths in the three cores. One shell sample was collected from 88.7 cm-deep of ER3 and one plant fragment from 251.2 cm-deep of ER3, too.

About 5~10 g dried bulk sediment samples were used for the extraction of the three humus

fractions (humic acid, fulvic acid, and humin). The sample was ultrasonically washed with distilled water and sifted through a sieve (106  $\mu$ m mesh). The samples passed through the sieve were mixed with 1.2N HCl solutions and heated to 80~90°C for 2~3 hours. This hot treatment was repeated 2~3 times. The acid filtrate was siphoned and the remaining sample was mixed with 1.2N NaOH solutions and heated to 80~90°C for 2~3 hours. This hot treatment was also repeated 2~3 times. The alkaline filtrate containing fulvic acid and humic acid fractions were siphoned. The residual after alkaline filtrate, the humin fraction, was washed with the distilled water and then dried by the vacuum freezing dryer. The humic acid fraction was precipitated by adding HCl solution into alkaline filtrate until pH < 1. The upper solution, which contains the fulvic acid fraction, was removed from the humic acid fraction precipitated. The precipitated humic acid was washed with distilled water and then dried by a vacuum freezing dryer. The separated solution was centrifuged under 3000~4000 rotation per minute for at least 40 minutes to get rid of any solid materials and dried by a vacuum freezing dryer to obtain the fulvic acid fraction.

In the case of plant fragment sample, it was also ultrasonically washed and then purified by the routine acid-alkali-acid (A-A-A) treatment described above to remove contamination completely. The resulting cellulose-rich and humic acid fractions were obtained. The extracted organic fractions were sealed in an evacuated glass tube with CuO and Pt catalyst, and combusted at  $850^{\circ}$ C for 2~3 hours resulting in CO<sub>2</sub> and water.

In the case of shell sample, the outer surfaces of the shell samples were etched with week (0.5N) hydrochloric acid and rinsed with deionized water and dried to remove any secondary carbonate. Shell sample was placed in 10-ml vacutainer and evacuated. After evacuation, 0.5 ml of phosphoric acid was added to the vacutainer and the shell sample was hydrolyzed for 30~60 minutes at 90°C to release  $CO_2$ .

The resulting  $CO_2$  from combustion and phosphoric acid reaction was purified cryogenically in

a vacuum line and then reduced catalytically to graphite on Fe-powder with hydrogen gas in a sealed Vycor<sup>®</sup> tube (Kitagawa et al., 1993). The <sup>14</sup>C/<sup>12</sup>C ratios of the graphite target were measured using a Tandem accelerator mass spectrometry at the JNC Tono Geoscience Center (Xu et al., 2000). The standards of HOxII oxalic acid and IAEA C-1 were used for <sup>14</sup>C normalization and the background correction, respectively. Other standard samples such as IAEA C-2 were also measured together with unknown samples for monitoring the <sup>14</sup>C accuracy, the result of which is in well agreement with the consensus values within  $1\sigma$  error margin. The carbon isotopic fractionation was corrected by using the sample  $\delta^{13}$ C measured for an aliquot of CO<sub>2</sub> gas with a conventional isotope ratio mass spectrometer (IRMS).

#### RESULTS

The measured <sup>14</sup>C ages and  $\delta^{13}$ C values for the samples are listed in Table 1 and shown in Fig. 2. The stated uncertainties (1 $\sigma$ ) of <sup>14</sup>C ages include both the statistical uncertainty of the samples and the reproducibility of the standards and blanks. Humic acid fraction in samples 97.4 cm-deep of ER1 and 88.7 cm-deep of ER3, and fulvic acid fraction in samples 58.5 cm- and 115 cm-deep of ER24 were not determined as the amount of these organic fractions extracted were too small to be detected.

#### Carbon stable isotope ratios

The carbon isotope ratios for organic fractions from the Erhai Lake sediment vary from -25.3 to -28.8% (Table 1 and Fig. 3a). Variation of  $\delta^{13}$ C values for fulvic acid, humic acid and humin fractions are from -25.3 to -28.6%, from -26.3 to -28.3% and from -26.5 to -28.8% respectively. It is clear in Fig. 3a that  $\delta^{13}$ C values below 100 cm in ER3 and ER24 are less negative than those above, although the difference is only about 1.5%for humic acid fraction, 2% for humin fraction. Such a small change in  $\delta^{13}$ C with sediment depth may be related to the change of organic source or sedimentation process.

Core No.	Depth (cm)	Material	δ <sup>13</sup> C (‰)	<sup>14</sup> C age <sup>(a)</sup> (yr BP)	Age difference <sup>(b)</sup> (yr)	Lab code (JNC-)
ER1	97.4	Fulvic acid	-27.7	$5,690 \pm 40$		2027
		Humic acid	—	—		
		Humin	-27.5	$4,970 \pm 40$		2024
		Shell		$3,030 \pm 110^{(c)}$		
ER3	88.7	Fulvic acid	-26.9	$4,490 \pm 40$		2028
		Humic acid		_		
		Humin	-26.6	$2,910 \pm 40$		2025
		Shell	-5.4	$2,210 \pm 30$		2030
	251.2	Fulvic acid	-28.6	$7,270 \pm 50$		2029
		Humic acid	-28.3	$5,900 \pm 40$	1,400	2023
		Humin	-28.5	$6,120 \pm 40$		2026
		Cellulose-rich of plant	-26.3	$5,050 \pm 40$		2031
		Humic acid of plant	-26.2	$5,070 \pm 40$		2059
		Wood		$4,\!440\pm130^{\rm (c)}$		
ER24	10	Fulvic acid	-25.8	$2,650 \pm 40$		2003
		Humic acid	-26.5	$1,410 \pm 40$	370	1726
		Humin	-27.1	$2,260 \pm 30$		2009
	25	Fulvic acid	-25.7	$5,530 \pm 40$		2004
		Humic acid	-26.6	$1,640 \pm 30$	390	1727
		Humin	-26.7	$2,570 \pm 40$		2010
	58.5	Fulvic acid	_	_		
		Humic acid	-26.2	$1,940 \pm 30$	210	1728
		Humin	-27.4	$2,740 \pm 40$		2011
	75	Fulvic acid	-25.3	$5,310 \pm 40$		2006
		Humic acid	-26.4	$2,270 \pm 30$	300	1729
		Humin	-27.2	$2,570 \pm 30$		2188
	95	Fulvic acid	-25.4	$9,230 \pm 50$		2007
		Humic acid	-26.6	$3,890 \pm 30$	1,600	1730
		Humin	-27.5	$4,\!640\pm\!40$		2013
	115	Fulvic acid	_	_		
		Humic acid	-27.9	$7,\!330\pm\!40$	4,800	1731
		Humin	-28.8	$7,\!400\pm\!40$		2014

Table 1. AMS <sup>14</sup>C ages for the cored sediments from Erhai Lake, SW China

<sup>(a)</sup>In conventional radiocarbon age based on the Libby half-life (5568 years)  $\pm 1$  standard deviation.

<sup>(b)</sup>Age difference between autochthonous humic acid fraction  ${}^{14}C$  age and "the true  ${}^{14}C$  age" calculated by Eq. (1) for each horizontal level.

<sup>(c)</sup>Hyodo et al. (1999).

The  $\delta^{13}$ C value of plant fragment sampled from 251.2 m-deep of ER3 is -26.3‰, consistent with that of humic acid fraction extracted from the same plant fragment. The  $\delta^{13}$ C value of shell sampled from 88.7 cm-deep of ER3 is -5.4‰.

#### Radiocarbon dates

As a whole, variation of apparent <sup>14</sup>C ages in this study is from 2 to 10 ka BP through the sequence from 10 to 251 cm deep. <sup>14</sup>C dates for organic fractions extracted from humus substance show a distinct discrepancy in ages for different fractions in individual horizontal level (Table 1 and Fig. 2).

Core ER1 (97.4 cm-deep) and ER3 (88.7 cm and 251.2 cm-deep) Fulvic acid and humin fractions in 97.4 cm-deep of ER1 gave <sup>14</sup>C ages of 5690  $\pm$  40 and 4970  $\pm$  40 yr BP, respectively. Similarly, fulvic acid and humin fractions in 88.7m-deep of ER3 gave <sup>14</sup>C ages of 4490  $\pm$  40 and 2910  $\pm$  40 yr BP, respectively. A shell sample gave a <sup>14</sup>C age of 2210  $\pm$  30 yr BP, 700~2200 yr younger than do



Fig. 3.  $\delta^{13}C(a)$  and  ${}^{14}C$  age (b) profile in the cores ER3 and ER24 sediment from the central Erhai Lake. The dotted line is the best fitting obtained from the accumulated terrestrial macrofossil (plant fragment and shell) measured in this study and Hyodo et al. (1999).

organic fraction extracted from humus substance in the same horizontal level.

Five organic fractions were measured for sample from 251.2 m-deep of ER3 and gave <sup>14</sup>C age distribution pattern: (fulvic acid) > (humic acid = humin) > (plant cellulose-rich fraction = plant humic acid fraction). It should be pointed out that the consistent age between cellulose-rich fraction and humic acid fraction extracted from plant fragment indicates no postburial effect and negligible vertical movement for humic acid fraction. It is also worth to note that our measured celluloserich fraction age  $(5050 \pm 40 \text{ yr BP})$  is older than previous date,  $4440 \pm 130$  yr BP, obtained in the same horizon by Hyodo et al. (1999). The reason for the difference between the two measurements remains unclear. Our plant fragment may have a relative long resident time on the land before deposited in the lake. Alternatively, it may be caused by the difference of chemical treatment between laboratories, as observed previously in Martin and Johnson (1995). However, considering the uncertainties  $(2\sigma)$  of two measurements, we found that the difference of <sup>14</sup>C dates does not change significantly the conclusion of this study and we use both dates in the following discussion.

*Core ER24* <sup>14</sup>C ages were obtained from fulvic acid, humic acid and humin fractions in six horizons. It can be found that (1) like other horizontal level in other cores, fulvic acid fraction in ER24 core is also older than other fractions in each individual horizontal level; (2) sample in 115 cmdeep has identical ages for humic acid and humin fractions; (3) normal sequence was found for both humic acid and humin fraction from surface to 115 cm deep.

#### DISCUSSION

#### Source of organic matter in Erhai Lake sediments

Organic carbon in lacustrine sediments may be considered to have two main sources: autochthonous biogenic components and allochthonous terrestrial organics. It is well known that  $\delta^{13}$ C value of terrestrial C3 plant ranges from -22 to -32‰ with average of -27‰ (Boutton, 1996). Lowe *et al.* (1997) found that the  $\delta^{13}$ C values of the Mahoney Lake sediment in southern British



Fig. 4. Variation of  ${}^{14}C$  age difference between "the true  ${}^{14}C$  age" and the observed humic acid fraction age in the Erhai Lake since 4500 yr BP.

Columbia range from -23.5 to -28.7‰. Horiuchi et al. (2000) also reported that  $\delta^{13}$ C values of organic matter in surface sediment in Lake Baikal range from -25.0 to -29.5%. Variation of these  $\delta^{13}$ C values is in the range for typical autochthonous origin in lake (Irwin et al., 1977). Therefore, it is not easy to distinguish the carbon source from  $\delta^{13}$ C value itself. Taking into account of many diatom frustules in the surface sediments in Erhai Lake (Zhu, 1989), we consider that  $\delta^{13}C$ values of the Erhai Lake sediment ranging from -25.3 to -28.8‰ may suggest mainly autochthonous origin. The apparent less negative  $\delta^{13}C$ value below 100 cm than those above observed in ER3 and ER24 may imply that small amount of terrestrial organic matter with much less negative  $\delta^{13}$ C value (i.e., <-29‰) has contributed to the lake sediment below 100 cm in the Erhai Lake. Other factors such as temperature and the fluvial environments of the lake, may have a minor influence on the change in  $\delta^{13}$ C value of sediment.

The  $\delta^{13}$ C values of sea-shells are almost constant being typical 0‰, however, those of landshells show quite various values (Boutton, 1996). One shell sample from 88.7 cm-deep of ER3 has the  $\delta^{13}$ C values of -5.4%, suggesting most likely its terrestrial origin. Hence, like terrestrial plant fragment, it can be used for discussion on sedimentation rate without reservoir effect correction.

# <sup>14</sup>C age and depth correlation of terrestrial macrofossils

The bottom slop along the sides of the Erhai Lake is in the order 2% to 3%, but in the middle and central part, where ER3 and ER24 are located, becomes less than 1% (Eisma et al., 2000). This geographical evidence together with lithological features strongly suggests that the core sediments between ER3 and ER24 can be horizontally compared. Based on this view, <sup>14</sup>C ages for each carbon fraction of ER3 and ER24 were plotted in Fig. 3b. It can be found that there is a linear correlation between <sup>14</sup>C age and depth for the terrestrial macrofossils (plant fragment and shell) in ER3 and ER24 measured in this study and Hyodo et al. (1999). Although there is a gap between 115 and 251 cm-deep, the best fitting gives the linear correlation (dashed line in Fig. 3b) between <sup>14</sup>C age (yr BP) and sampling depth d ( cm).

From Eq. (1), a constant sedimentation rate,  $ca. 0.7 \text{ mm yr}^{-1}$ , is estimated from near surface to 251 cm-depth, corresponding to the period from present to 4500 yr BP.

## Variations in <sup>14</sup>C ages of organic fractions

Like many previous studies (e.g., Kigoshi *et al.*, 1978; Martin and Johnson, 1995), our results support that there is no consistent sequence for the organic matter fractions. The same question raised as which fraction is more reliable than other fractions.

Fulvic acid fraction, extractable with an acidtreatment, has been observed to give younger <sup>14</sup>C age than do other organic components for soil profiles (Campbell et al., 1967; Miyazaki, 1971), which suggests that fulvic acid in the sediments can be transported vertically by pore water. Hence, fulvic acid fraction of humus substance at any horizon of the sediments may partly contain foreign fulvic acid transported from upper and/or lower horizons. Such a consideration may also be the case of humic acid fraction, as observed previously by Kigoshi et al. (1978). In contrast, humin fraction of the sediments, insoluble in water, as well as acid- and alkali-insoluble, are not movable and are preserved in the sediments where they accumulated, if there is no turbulent movement of the sediments after they deposited. From these views, the <sup>14</sup>C ages of humin fraction from the humus substances is generally considered to be more reliable than those of fulvic acid and humic acid. However, in the case of the central Erhai Lake as discussed below, we found that humic acid fraction may be more reliable than humin fraction.

Humic acid and humin fractions have identical <sup>14</sup>C ages in 115 m and 251.2 m-deep (Fig. 3b). However, <sup>14</sup>C ages of humic acid are younger than those of humin fraction above 115 m-deep. Normal sequence was found for both humic acid and humin fraction from surface to 115 cm-deep. It is clear that there is similar but distinct linear correlation between <sup>14</sup>C age and depth between 10 and 90 cm-deep for both humic acid and humin fraction. The <sup>14</sup>C age-depth correlation for humin fraction is apparently steeper than that for humic acid fraction. We can presumably calculate the sedimentation rate from <sup>14</sup>C age-depth correlation for humin and humic acid fraction between 10 and 90 m-deep. The result calculated from humin <sup>14</sup>C age-depth correlation gave a sedimentation rate of 2 mm yr<sup>-1</sup> whereas that from humic acid gave of 0.7 mm yr<sup>-1</sup>. It is clear that the former value since 3000 yr BP is too high while the latter value is quite reasonable (Eisma et al., 2000). Therefore, although more analysis is absolutely necessary before reaching a conclusion, we presumably consider that, in the case of Erhai Lake sediments, humic acid fraction <sup>14</sup>C age is more reliable that humin fraction. However, the mechanism is still unclear.

### $^{14}C$ age shift of humic acid fraction

"The true <sup>14</sup>C age" of each horizontal level can be calculated by interpolating the sample depth to Eq. (1) and were shown in the right ordinate in Fig. 4. Hence, comparing "the true <sup>14</sup>C age" with humic acid fraction <sup>14</sup>C age, we estimated the age shift of humic acid fraction from "the true <sup>14</sup>C age" in each horizontal level of ER3 and ER24 (Table 1 and Fig. 4). The results gave the age shift from 210 to 4800 yr since 4500 yr BP. We presumably consider the following possibilities which may be related to the <sup>14</sup>C age shift.

Lakes have two principal sources of  $^{14}$ C: atmospheric CO<sub>2</sub> and the total dissolved inorganic carbon (TDIC) of the entering groundwater and runoff. The former has values of *ca*. 100 pMC (percent modern carbon), the latter usually has a  $^{14}$ C value well below 100 pMC. As a result, the  $^{14}$ C ages of lake sediments are usually exaggerated. A shellfish alive today in a lake within a limestone catchment, for instance, will yield a radiocarbon date which is excessively old. The reason for this anomaly is that the limestone, which is weathered and dissolved into bicarbonate, has no radioactive carbon. This phenomenon is termed as "reservoir effect". One of the most commonly referenced reservoir effects concerns the ocean. The average difference between a radiocarbon date of a terrestrial sample such as a tree, and a shell from the marine environment is about 400 yr in <sup>14</sup>C age (Stuiver and Braziunas, 1993).

Some researchers have shown that the application of a uniform reservoir correction throughout a sequence may be erroneous (e.g., Turner et al., 1983). However, while agreeing with this view, MacDonald et al. (1991) presented data showing that, for the last 10,000 yr BP, bulk <sup>14</sup>C ages on calcareous lake sediment essentially parallel with <sup>14</sup>C ages from terrestrial macrofossils. If the application of a uniform reservoir correction (most likely 300 yr as shown by upper four samples) throughout the core is valid in the Erhai Lake, the age shift before 2000 yr BP could have to invoke other carbon source, for instance, such as the contribution of terrestrial organic matter to lake sediment. If terrestrial organic matter contains low or no <sup>14</sup>C, small contribution of such terrestrial carbon to lake sediment may cause large <sup>14</sup>C age shift of organic matter. Such a consideration is supported by geochemical and geophysical evidence. From  $\delta^{13}$ C values of organic matter in the Erhai Lake, less negative values below 100 cm (corresponding to 2000 yr BP) may imply small contribution of terrestrial organic matter. Hyodo et al. (1999) found that the curves of ARM intensity and ARM intensity normalized by susceptibility (ARM/susceptibility) show a sudden increase at around 100 cm (corresponding to ca. 2000 yr BP) and explained it to be related to a sudden change in grain size. Consequently, they concluded a large change in the source of sediments in the Erhai Lake around 2000 yr BP. It is clear that the period with large <sup>14</sup>C age shift in Fig. 4 synchronizes with the period of large ARM intensity and ARM/susceptibility values, given by Hyodo et al. (1999).

Alternatively, <sup>14</sup>C age shift (210~4800 yr) may be related to the temporal change of reservoir effect since 4500 yr BP, as observed in some freshwater and saline lakes (Geyh *et al.*, 1998). It was argued that the proportion of atmospheric  $CO_2$  in the total carbon balance of a lake depends on the ratio of the water volume to the lake surface area. The water surface area can be considered to be constant for lakes with an oulflow. The ratio of water volume to the lake surface area is related to the maximum water depth when the water is dammed by a barrier growing in the river bed. On the other hand, normally water of a lake stratifies in summer season. The establishment of lake-water stratification prevents vertical circulation of lake water. However, full water column mixes well vertically in winter season. Hence, from these views, it may be not expected that <sup>14</sup>C concentration of TDIC and CO<sub>2</sub> in lake water changes to as low as 45% (corresponding to *ca*. 4800 yr in <sup>14</sup>C age) of the normal <sup>14</sup>C concentration level from 4500 to 2000 yr BP, due to only the carbon reservoir effect. More  $\delta^{13}C$  and  $^{14}C$  analysis and organic geochemical research in future will be helpful to clarify the alternatives.

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