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Organic carbon source and burial during the past one hundred years in Jiaozhou Bay, North China

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Abstract

Organic carbon (OC), total nitrogen (TN), and ^{210}Pb in core sediment were measured to quantify the burial of organic carbon and the relative importance of allochthonous and autochthonous contributions during the past one hundred years in Jiaozhou Bay, North China. The core sediment was dated using ^{210}Pb chronology, which is the most promising method for estimation of sedimentation rate on a time scale of 100–150 years. The variation of the burial flux of organic carbon in the past one hundred years can be divided into the following three stages: (1) relatively steady before 1980s; (2) increasing rapidly from the 1980s to a peak in the 1990s, and (3) decreasing from the 1990s to the present. The change is consistent with the amount of solid waste and sewage emptied into the bay. The OC:TN ratio was used to evaluate the source of organic carbon in the Jiaozhou Bay sediment. In the inner bay and bay mouth, the organic carbon was the main contributor from terrestrial sources, whereas only about half of organic carbon was contributed from terrestrial source in the outer bay. In the inner bay, the terrestrial source of organic carbon showed a steady change with an increase in the range of 69%–77% before 1990 to 93% in 2000, and then decreased from 2000 because of the decrease in the terrestrial input. In the bay mouth, the percentage of organic carbon from land reached the highest value with 94% in 1994. In the outer bay, the sediment source maintained steady for the past one hundred years.

Key words: organic carbon; source; burial; Jiaozhou Bay

Introduction

Coastal areas play a vital role in the global carbon cycle either as sources of organic matter (OM) to the open ocean or as carbon sinks due to accumulation of OM in sediments (Tesi *et al*., 2007). As an accumulation of "geochemical fossils", the organic matter content of coastal sea sediments provides information that is important to interpretations of coastal sea paleoenvironments, histories of climatic change, and the effects of human activities on local and regional ecosystems (Meyers and Vergès, 1999; Gonneea *et al*., 2004). The primary source of organic matter to coastal sea sediments is from the particulate detritus of plants; only a few percent came from animals. Plants can be divided into two geochemical distinctive groups on the basis of their biochemical compositions: (1) nonvascular plants that contain little or no carbon-rich cellulose and lignin, such as phytoplankton, and (2) vascular plants that contain large proportions of these fibrous tissues, such as grasses, shrubs, and trees. Because the reactivity of higher plant and microbial OM is quite different, assessing autochthonous marine and allochthonous terrestrial inputs in coastal areas is of great importance. The relative contributions from these two plant groups to sedimentary records are influenced strongly by morphology, watershed topography, climate, abundance of coastal sea plants and watershed plants. Studies of organic matter in the sediments from different parts of the world have been used to help reconstruct records of regional and continental paleoclimates. An important component of paleolimnologic investigations is to identify the sources of organic matter in sediments deposited at different times in the past. Variations of C:N ratios within sediments have been used to determine historical changes in sources of organic matter. Algae have a C:N ratio between 4 and 10, whereas terrestrial organic matter has a C:N ratio greater than 20 (Kaushal and Binford, 1999; Brenner *et al*., 2006). Increase in C:N ratio within sediment profiles have been interpreted to identify periods in sedimentary history when sediments received a high proportion of terrestrial organic matter. Conversely, decrease in C:N ratios have been used to identify periods when sediments have received a high proportion of algal organic matter (Kennedy *et al*., 2004).

In this study, we used C:N ratio to trace the variations of organic matter source in the Jiaozhou Bay sediment, and to examine the changes in organic matter burial rate during the past one hundred years.

1 Materials and methods

1.1 Study area

Jiaozhou Bay is a semienclosed bay situated on the south

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bank of Shandong Peninsula, China, linked with Yellow Sea with a very narrow entrance about only 3.1 km (Fig.1). It extends from 36°00′53″N to 36°02′36″N and from 120°16′49″E to 120°17′30″E. The average depth is 7 m, with the maximum of 64 m. It covers an area of 362 km^2 of seawater surrounded by Qingdao City. More than 10 small rivers empty into the bay with varying water and sediment loads, notably Yanghe, Daguhe, Moshuihe, Baishahe and Licunhe River (Li *et al*., 2007b), and the largest one is Daguhe River, with an annual average runoff of 6.61×10^8 m^3. Most of these rivers, however, have become canals of industrial and domestic waste discharge with the increased population and economic activity in the region. According to Shen (2001), from 1962 to 1998, the population of Qingdao City increased from 4.6 to 7.0 million, and the gross value of industrial output increased 80 times. The discharge of industrial wastewater and sewage in urban district is 70.2×10^6 and 14.4×10^6 t/a, respectively. The use of fertilizers has increased three times from 1980 to 1998. Overall, due to the rapid economical and social developments in this region, Jiaozhou Bay is greatly influenced by human activities, leading to increased amount of industrial, agricultural, and aquaculture waste into Jiaozhou Bay. Red tide has become a frequent event, as the bay water is eutrophied. During the past 70 years, the water area of Jiaozhou Bay has decreased by one-third mainly because of the dumping of garbage from Qingdao City (Dai *et al*., 2007).

1.2 Sampling

Three sediment cores were collected with a gravity corer on board of R/V "Jinxing II" in September, 2003 (Fig.1). Station B3 was located in the bay center, with 6 m water depth, and 94 cm long core was collected, containing yellow-grey sandy mud on the surface. Station D4 was in the south of the bay and near the bay mouth, with 21 m water depth, and 104 cm core with grey-black ooze on its surface was taken. Station D7 was situated in the outer bay with 17 m water depth, and 82 cm long core was collected with yellow-grey muddy sand on its surface. The cores were successively cut into 2.0 cm thick slices from surface to bottom. All samples were sealed in polyethylene bags and brought to the laboratory for further analyses.

For the sake of convenience and statement clarity, we use Stations B3, D4, and D7 to represent the inner bay, bay mouth, and outer bay, respectively.

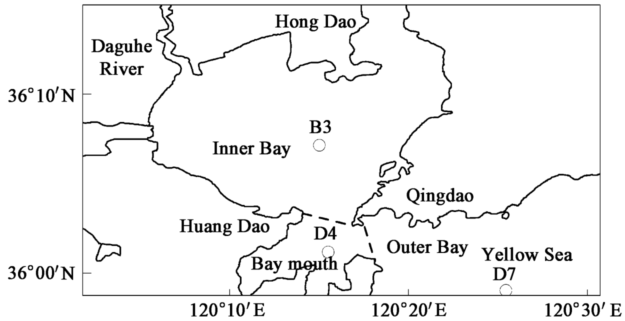

Fig. 1 Map of study area with core locations.

1.3 Laboratory analyses

Inorganic carbon was determined by a LECO CR-12 elemental analyzer with an analytical precision of 0.5% (Li *et al*., 2006b). Total nitrogen (TN) was extracted by the potassium persulfate-sodium hydroxide mixture solution in stainless steel autoclave and maintained at 124°C for 1 h, then determined by colorimetric method with an analytical precision of 0.5% (Li *et al*., 2007a). Organic carbon was determined by the revised Walkley-Black titration method (Leong and Tanner, 1999).

^{210}Pb activities were measured by alpha-counting its granddaughter ^{210}Po (i.e., ^{210}Pb $T_{1/2}$ = 22.3 years $\longrightarrow$ ^{210}Bi $T_{1/2}$ = 5.01 d $\longrightarrow$ ^{210}Po $T_{1/2}$ = 13.8 d), using a technique similar to that reported by Lu and Matsumoto (2006) and Li *et al*. (2003, 2006a). Approximately 0.5 g of ^{208}Po spike was added to 1–3 g of dried sediment prior to leaching with HCl, ammonium citrate tribasic, and hydrogen peroxide. The particles were separated from the acid solution by centrifugation, dried, and then pickled in 1 mol/L HCl. An Ag planchet (1×1 cm) was added into the solution to remove polonium isotopes. Supported level of ^{210}Pb activities was determined by measuring ^{210}Pb below the region of excess activity. Excess ^{210}Pb activity was determined by subtracting ^{210}Pb background activity (^{226}Ra-supported) from the total activity. The background activity was estimated at the core ^{210}Pb activity which had decayed to constant low levels with the depth. The excess ^{210}Pb activity was used to compute sediment age and sediment accumulation rates by the following equations:

$$t_i = \frac{1}{\lambda} \ln \frac{A_0}{A_i} \quad (1)$$

$$S = \frac{X_{i+1} - X_i}{t_{i+1} - t_i} \quad (2)$$

where, t_i is the sediment age at depth i; A_0 and A_i (dpm/g) are excess ^{210}Pb activity at the sediments surface and at depth i, respectively. S (cm/a) is the sedimentation rate of definite layer, i is the ith depth interval, X_i is the depth of the top of the ith interval, and t_i is the age of the top of the ith interval.

2 Results and discussion

2.1 ^{210}Pb chronology and sedimentation rate

One of the most promising methods for estimation of sedimentation rate on a time scale of 100–150 years is by means of ^{210}Pb, a naturally occurring radioisotope with a half-life of 22.3 years. In this article, the core sediment was dated using ^{210}Pb chronology from radioactive fallout and the sedimentation rate of definite layers were calculated and tabulated in Table 1.

Rapid increase of sedimentation rates was observed in the sediment record of Jiaozhou Bay, which is also a common phenomenon in coastal areas, especially, in recent years (Lu and Matsumoto, 2005). Intensification of land use, rapid population growth, deforestation, and urbanization has resulted in significant increase in delivery of sediments to the bay since the 1980s. Before the

Table 1 Sedimentation rates for different years in Jiaozhou Bay

Station B3 (inner bay)			Station D4 (bay mouth)			Station D7 (outer bay)		
Depth (cm)	Sedimentation rate (cm/a)	Year	Depth (cm)	Sedimentation rate (cm/a)	Year	Depth (cm)	Sedimentation rate (cm/a)	Year
0–2	0.64	2003	0–2	0.45	2003	0–2	0.49	2003
4–6	1.60	2000	4–6	2.52	1994	4–6	0.51	1997
8–10	0.60	1993	8–10	0.26	1992	8–10	1.03	1993
12–14	0.61	1987	12–14	0.26	1977	12–14	0.41	1983
16–18	0.48	1984	16–18	0.43	1962	14–16	0.36	1976
22–24	0.31	1964	22–24	0.43	1948	18–20	0.26	1960
30–32	0.31	1938	30–32	2.79	1930	24–26	0.26	1937
38–40	0.30	1912	38–40	2.79	1927	28–30	0.26	1922
48–50	0.30	1879	48–50	2.79	1923			
			58–60	2.79	1920			
			68–70	4.39	1917			

1980s, the sedimentation rates of the inner bay and outer bay maintained a relatively steady increase indicating a comparatively stable sedimentary environment. However, the sedimentation rate in the bay mouth sharply decreased from 1930s to 1940s because the Xinan River that emptied into the bay was almost dried up in the period. Climatic changes such as large flood events were not observed in the past one hundred years in Jiaozhou Bay, so the general accelerating trends in sedimentation rate from the 1980s would rather reflect the increasing effect of human impacts. With the rapid urbanization and economic growth from the 1980s, increased effluent from industry and agriculture will induce the increase of sediment discharge, which may be related to the long-term increasing trends of sedimentation rate between 1980 and 2003. Because Qingdao City was selected to be the cohost of 2008 Beijing Olympic Games, Licun, Haipohe, and Tuandao wastewater treatment plants, which are distributed on the east bank of the bay, had been built to improve the water quality, and the garbage from industry and agriculture was not allowed to be dumped into the bay directly. These measures might have accounted for the decrease in sedimentation rate since the beginning of this century. In addition, the deforestation and dam construction in the rivers entering the bay can also affect the matter inputs rivers. For example, dams have been built since 1950s, which has reduced the sand inputs to the bay to some degree. But the influence is rather weak compared with the sewage and wastewater discharge into the bay, thus, the sedimentation rates of Jiaozhou Bay increased on the whole.

2.2 Total organic carbon (TOC)

TOC in Jiaozhou Bay sediment was less than 0.5% which decreased from inner bay to bay mouth and then to outer bay in the surface sediment (Fig.2). In general, organic carbon content in core sediments decreased in the upper 30 cm, then maintained a relatively constant level below this (Ingalls *et al.*, 2004). However, the vertical distribution of organic carbon content is complicated in Jiaozhou Bay core sediment. At Station B3, the organic carbon content decreased from 2 to 0 cm (2000 to 2003), stabilized from 8 to 2 cm (1993 to 2000), and then increased from 26 to 8 cm (1951 to 1993). Thus, the highest value (0.5%) was attained between 1993 and 2000.

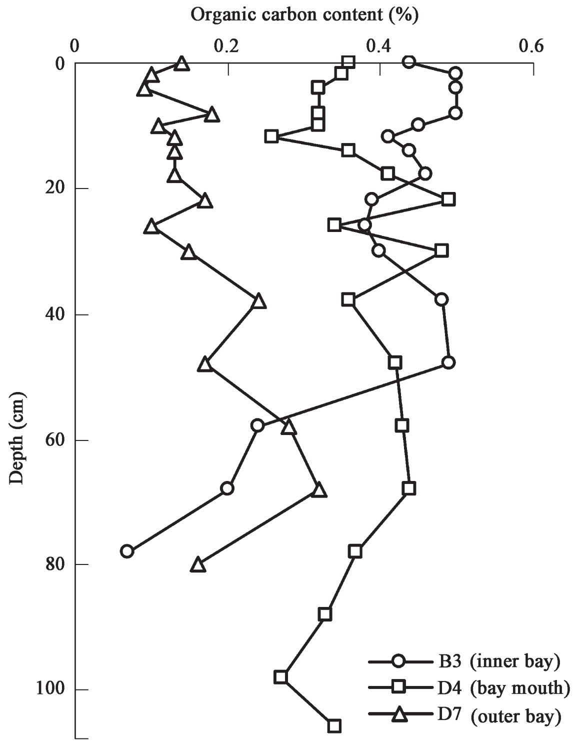

Fig. 2 Organic carbon contents in the Jiaozhou Bay sediment.

At Station D4, the organic carbon content increased from 12 to 0 cm (1977 to 2003), decreased from 22 to 12 cm (1948 to 1977), and then was maintained relatively constant prior to 1948. At Station D7, the organic carbon content increased from 4 to 0 cm (1997 to 2003) and decreased below the depth (prior to 1997). It is obvious that the organic carbon content variation was not caused by organic matter remineralization but had close relationship with the input of solid waste and sewage from Qindao City because the variation is highly consistent with the solid waste and sewage input.

2.3 Burial flux of organic carbon

The burial flux of organic carbon in marine sediments can be determined by the supply and preservation of organic matter into the sediments. However, the mechanisms that govern OC burial in margins are controversial and have historically focused on the importance of marine processes. Productivity, bottom water oxygen level, sediment accumulation rate, sediment porosity, microbial activity, bioturbation rates, and organic matter composition can all influence OC preservation in sediments (Blair *et al*., 2004). In general, three pieces of information are required to calculate the burial flux in marine sediments: the organic carbon content of sediments accumulating below a defined horizon, the recent sedimentation rate, and the dry bulk density. The burial flux (F_b) can be expressed as:

$$F_b = C_{oc} S \rho_d \tag{3}$$

$$\rho_d = (1 - C_w)/((1 - C_w)/\rho_s + C_w/\rho_w) \tag{4}$$

where, F_b (mg/(a·cm^2)) is the burial flux of organic carbon in sediments, C_{oc} (%) is the content of organic carbon in sediments, S (cm/a) is the sedimentation rate, C_w (%) is the water content in sediment; ρ_w (g/cm^3) is the water density, ρ_d (g/cm^3) is the sediment dry bulk density, and ρ_s is the sediment grain density and assumed to be 2.56 g/cm^3 (Dai *et al*., 2007) in this study.

On the basis of Eqs. (3) and (4), the burial fluxes of organic matter were calculated and described in Fig.3. The burial fluxes of organic carbon began to increase in the 1980s with a peak at the end of last century (Fig.3). At the beginning of the century, with measurements adopted to manage pollution and eutrophication, the environmental quality has been improved greatly with a drop in the organic carbon burial flux. The variation of the burial flux of organic carbon in the past one hundred years can be divided into the following three stages: (1) relatively steady stage before the 1980s, (2) rapidly increasing stage from 1980 to a peak in 1990s, and (3) decreasing stage from 1990s to the present. The change is consistent with the amount of solid waste and sewage discharged into the bay. In the middle of the 1980s, the amount of industrial solid waste emptied into the bay was approximately 290.43×10^4 t/a and municipal garbage was approximately 33×10^4 t/a. However, the total solid waste decreased to 60×10^4 t/a in recent years. The amount of sewage discharged into the bay was approximately 83.5×10^6 t in 1980, increased to 145.6×10^6 t in 1987, and 193×10^6 t in 1995. Although the sewage increased steadily from 1980, the amount of sewage cleared by wastewater treatment work increased from 2000. At present the percentage of treated sewage has reached 75.63%. To reduce the amount of waste and sewage, many techniques such as increased regulation of wastewater discharge, reduction of nitrogen, phosphorus fertilizers, increased industrial waste recycling, and cleaner industrial processes have been applied from the 1990s. Thus, the burial fluxes of organic matter decreased from 1990s to present.

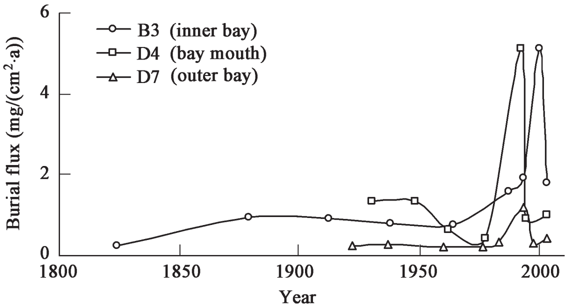

Fig. 3 Burial fluxes of organic matter during different periods in the Jiaozhou Bay sediment.

2.4 Variation of organic carbon sources in the past one hundred years

The ratio of nutrients (C:N:P) is regarded as an indicator to the source of organic carbon being autogenetic or allochthonous. If all the organic matter in sediment comes from marine phytoplankton, the ratio of C:N:P would be close to the Redfield value (106:16:1), and the ratio of OC:TN could be about 6.6. If they are from terrestrial sources, the ratio of OC:TN would be more than 20, in general. The higher the percentage of terrestrial organic matter, the higher the ratio of OC:TN. The validity of using C:N ratios to discern changes in organic carbon sources have been questioned because the C:N of terrestrial organic matter decreases during diagenesis, whereas that of algae increases. In addition, the C:N ratio record of a particular coring site may not provide an accurate representation of changes in the entire sea. However, many studies show that C:N ratios in sediments can be used reliably to identify historical sources of sedimentary organic matter, and indicate human disturbance of watersheds. Milliman *et al.* (1984) used the ratio of OC:TN to evaluate the source of organic matter in the Yangtze River estuary. It was suggested that if the ratio of OC:TN is above 12, the organic matter was land-derived, and if the ratio is below 8, the organic matter would be indicated from marine autogeny. In addition, it was also observed that the organic matter in particles came mainly from the Yangtze River estuary in winter. Subsequently, Cai *et al*. (1992) evaluated the source of organic matter in the particles in the Yangtze River estuary using the method of stable isotope (δ^{13}C) and the ratios of OC:TN. The results of the two methods lead to the same conclusion, which improved the applicability of OC:TN ratios for indicating the source of organic matter. Large diagenetic changes in C:N ratios, which would have led to an overlap of terrestrial and algal C:N ratios, were not evident in either the surface or core sediments. Therefore, the significant increase in C:N for surface sediments was most likely caused by an increase in the proportion of terrestrial organic matter in sediments. The proportion of terrestrial organic matter could have risen because of increased particulate matter loads and discharges of streams directly following deforestation.

Organic matter in the Jiaozhou Bay sediment was land-derived or marine autogenic. The former came from industrial and agricultural activities along the Jiaozhou Bay coast by surrounding rivers; the latter was mainly from primary production in the bay. Qian *et al*. (1997) brought forward a equation that was used to evaluate approximate percentage of terrestrial source or autogenic source, and then the nitrogen or phosphorus source. The ratio of TOC:TN was used in the equation to determine quantitatively whether the organic carbon and nitrogen were hydrophytic or terrestrial. With this method, it is proposed that the ratio of C:N in hydrophytic organic matter and in terrestrial organic matter is 5 and 20, respectively (Jia *et al*., 2002). The equation is given as:

$\mathrm{TOC} = C_l + C_a$
$\mathrm{TN} = N_a + N_l$
$C_a/N_a = 5$
$C_l/N_l = 20$

where, TOC and TN are the measured values, C_l is the content of organic carbon from land; C_a is the content of organic carbon from marine autogeny, N_l is the content of nitrogen from land, and N_a is the content of nitrogen from marine autogeny.

Then, the following equations can be induced from the above equation:

$$C_a = (20\mathrm{TN} - \mathrm{TOC})/3 \quad (5)$$

$$C_l = 4(\mathrm{TOC} - 5\mathrm{TN})/3 \quad (6)$$

The ratio of OC:TN in the Jiaozhou Bay sediment ranged between 6.48 and 17.92 (Fig.4). It was much greater at Station B3 (inner bay) and Station D4 (bay mouth) than that in outer bay at station D7 because the former stations had more intake of terrestrial matter in the sediments from rivers. On the contrary, the outer bay received more marine autogenic organic matter in sediment that resulted in low OC/TN ratio. The similar variation between Station B3 in inner bay and Station D4 in the bay mouth indicated they have similar sources of organic carbon in the past one hundred years. However, the ratios of OC:TN in Station D4 are slightly higher than that in Station B3. In the bay mouth, its area is small and waste water produced by industry and agriculture in Huangdao and its surrounding area was drained into this region by the Xin-an River. In addition, a number of ships shuffled through port near bay mouth might bring a mass of organic matter, and result in a higher percentage of external organic matter in the bay mouth.

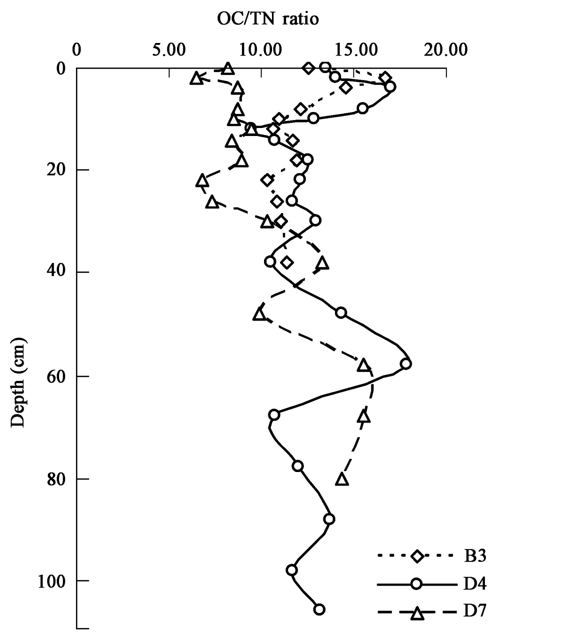

Fig. 4 Vertical distributions of OC:TN ratio in the Jiaozhou Bay sediment.

Calculations from Eqs.(5) and (6) indicate that the organic carbon from the inner bay (Station B3) and the bay mouth (Station D4) have a predominantly terrestrial source, and more than 70% terrestrial organic carbon input (Fig.5). However, the terrestrial matter is about half of the source in the outer bay (Station D7). In the inner bay (Station B3), the terrestrial source is steady with a range of 69%–77% before 1990. The percentage of organic carbon from land increased to 93% in 2000, which is consistent with the increase of solid waste and sewage discharged into the bay in this period. From 2000, the percentage of organic carbon from land decreased due to the decrease of terrestrial input. In the bay mouth (Station D4), the percentage of organic carbon from land reached the highest value with 94% in 1994. The matter source maintained steady in the outer bay. The lower percentage of organic carbon from land in 1946 and 1999 may be caused by phytoplankton bloom in this period.

2.5 Ratio of organic carbon to inorganic carbon

The ratio of organic carbon to inorganic carbon (C_{org}/C_{inorg}) is important for quantifying the efficiency of the biological pump in drawing down the $p\mathrm{CO_2}$ in the surface water because photosynthesis (assimilation) decreases the $p\mathrm{CO_2}$ of surface water, whereas more or-

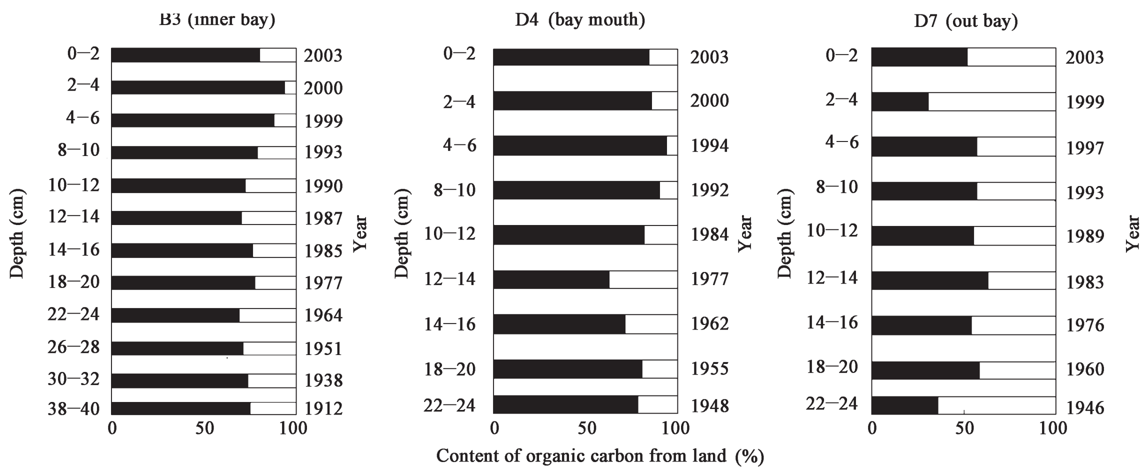

Fig. 5 Variation of organic matter source in Jiaozhou Bay in the past one hundred years.

ganic matter is produced (Honda, 2003). By drawing down the pCO_2 of the surface water, the biological pump enhances the potential uptake of atmospheric CO_2 and the production of organic matter by the ocean. Tsunogai and Noriki (1991) summarized sediment-trap data for the global ocean and found that C_{org}/C_{inorg} ratios of sediment-trap samples collected in the mesopelagic and pelagic layers (deeper than 500 m) are greater than 1 on average. On the basis of stoichiometry, Kano (1990) suggested that 0.6 is the boundary C_{org}/C_{inorg} ratio; showing that the pCO_2 of seawater increases when the particulate carbon is biologically produced and its C_{org}/C_{inorg} ratio is less than approximately 0.6. In Jiaozhou Bay, C_{org}/C_{inorg} ratio in sediment can be used to evaluate marine autogenic organic matter production because the water depth is shallow and the organic matter produced in water can be sunk to bottom before it was decomposed. The ratio of C_{org}/C_{inorg} in the inner bay is similar with that in the bay mouth with average of 0.81 and 0.88, respectively, but higher than that in the outer bay with an average of 0.17 (Fig.6). The C_{org}/C_{inorg} indicates that the inner bay and bay mouth can absorb more atmospheric CO_2 and produce more organic matter than that in outer bay. This is consistent with the conclusion that the inner bay and the bay mouth can absorb atmospheric CO_2 in winter (phytoplankton bloom season), but the outer bay can not (Li *et al*., 2007c). According to the vertical variation of the ratios in the past hundred years, the marine autogenic organic carbon is stable in the outer bay, but increases in the inner bay and bay mouth, especially after the 1980s.

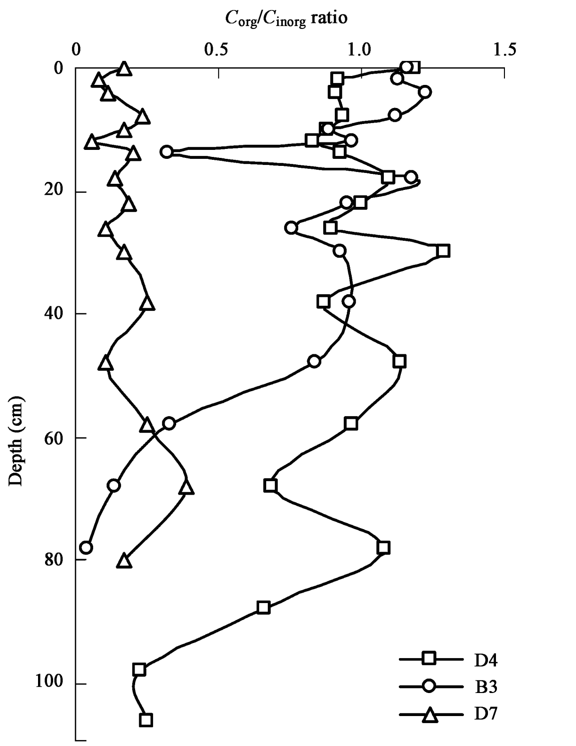

Fig. 6 Vertical distributions of C_{org}/C_{inorg} ratio in the Jiaozhou Bay sediment.

3 Conclusions

The sources and burial of organic carbon during the past one hundred years were studied with three core sediments in the Jiaozhou Bay. The variation of the burial flux of organic carbon in the past one hundred years can be divided into the following three stages: (1) relatively steady stage before the 1980s, (2) rapidly increasing stage from 1980 to a peak in the 1990s, and (3) decreasing stage from the 1990s to the present. The change is consistent with the amount of solid waste and sewage emptied into the bay. According to the ratio of OC:TN, the organic carbon mainly came from terrestrial sources in the inner bay and the bay mouth, and only about half of organic carbon was from terrestrial source in the outer bay. In the inner bay, the terrestrial source was steady with a range of 69%–77% before 1990. Then, the percentage of organic carbon from land increased to 93% in 2000. From 2000, the percentage of organic carbon from land decreased due to the decrease of terrestrial input. In the bay mouth, the percentage of organic carbon from land reached the highest value with 94% in 1994. In the outer bay, the matter source maintained steady in the past one hundred years.

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