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Organic and inorganic carbon and their stable isotopes in surface sediments of the Yellow River Estuary

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Abstract

Estuarine sediment is an important carbon reservoir, and thus may play an important role in the global carbon cycle. The Yellow River Estuary is a large estuary in northern China, having implications for the Bohai Sea's carbon cycle. However, little is known about carbon dynamics in the sediment of the transitional zone near the river mouth. In this study, we collected 15 short sediment cores from the Yellow River Estuary, and measured grain size, total nitrogen (TN), total organic carbon (TOC) and inorganic carbon (TIC) and the isotopic compositions of TOC ($\delta^{13}C_{org}$) and carbonate ($\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$). We found that TIC concentration (6.3-20.1 g kg^{-1}) was much higher than TOC (0.2-4.4 g kg^{-1}) in the surface sediment. Both TOC and TIC were higher to the north (2.6 and 14.5 g kg^{-1}) than to the south (1.6 and 12.2 g kg^{-1}), except in the southern bay where TOC and TIC reached 2.7 and 15.4 g kg^{-1}, respectively. The $\delta^{13}C_{org}$ value ranged narrowly from -24.26‰ to -22.66‰, indicating that TOC might

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be mainly autochthonous. However, C:N ratio varied from 2.1 to 10.1, with higher ratio found in the southern bay. We estimated that 60.8% of TOC might be from terrigenous OC in the southern bay. The lower TOC values in the south section were due to relatively higher kinetic energy level whereas the higher values in the bay was attributable to terrigenous matters accumulation and lower kinetic energy level. There was a significantly positive correlation between TIC and TOC, indicating that TIC was primarily from autogenic carbonate. However, the southern bay revealed the most negative $\delta^{13}C_{org}$ and $\delta^{13}C_{carb}$, suggesting that there might exist some transfer of OC to IC in the section. Our study points out that the dynamics of sedimentary carbon in the Yellow River Estuary is influenced by multiple and complex processes, and highlights the importance of carbonate in carbon sequestration.

1 Introduction

The rate of CO_2 build-up in the atmosphere depends on the rate of fossil fuel combustion and the rate of CO_2 uptake by the ocean and terrestrial biota. About half of the anthropogenic CO_2 has been absorbed by land and ocean. Large rivers that connect the land and ocean may play an important role in the global carbon cycle (Bianchi and Allison, 2009;Ran et al., 2015;Wang et al., 2016c). On the one hand, river can transport a significant amount of dissolved and particulate carbon materials from the land to the ocean, which are subject to recycling and sedimentation in the estuaries, or further transportation to the marginal seas (Cole et al., 2007;Bauer et al., 2013). On the other hand, there may be high levels of nutrients in the river waters, which could cause enhanced biological uptake of CO_2 and subsequent carbon burial in the estuaries (Cai, 2010;Raimonet and Cloern, 2017).

The Yellow River, the second largest river in China following the Yangtze River, provides approximately 50% of the freshwater discharged into the Bohai Sea every year (Wang et al., 2006). However, as the world's largest carrier of fluvial sediment, its sediment load has continually decreased since the 1950s due to changes in water discharge and sediment concentration by anthropogenic changes (Wang et al., 2016a). These changes may have profound impacts on the physical, biogeochemical and biological processes in the Yellow River Estuary.

There were some studies on sedimentary organic carbon around the Yellow River Estuary, which were mainly conducted in the Yellow River Delta (Bianchi and Allison, 2009;Ye et al., 2015;Zhao et al., 2015) and in the shelf sediments of the Bohai Sea (Hu et al., 2016;Liu et al., 2015;Xing et al., 2016;Wang et al., 2017). Limited studies showed a large spatial variability in total organic carbon (TOC, 0.7-7.7 g kg^{-1}) in the Yellow River Estuary (Li et al., 2014b), with the highest contribution (40-50%) of terrestrial organic carbon found near the delta, which might be due to the hydrodynamics constrained sedimentary environment and deposition rate and current speed (Liu et al., 2015). However, little is known about the TOC dynamics in the sediment for the transitional zone near the river mouth.

There were limited studies of inorganic carbon dynamics in the Yellow River Estuary. A field based analysis demonstrated that rate of $CaCO_3$ precipitation was modestly higher than biological production in the water columns of the estuary (Liu et al., 2014). In addition, Gu et al. (2009) found that particulate inorganic carbon (1.8%±0.2%) was also much higher than particulate organic carbon (0.5%±0.05%) in the Yellow River Estuary. These findings indicate that there might be much more inorganic carbon (TIC) than TOC in the sediment of the Yellow River Estuary. While there was evidence of high level of TIC in the sediment of the lower Yellow River Delta (Zhao et al., 2015), little has been done to evaluate the magnitude and variability of TIC in the Yellow River Estuary.

Recent studies have showed that there was a large amount of carbonate in the soils of lower part of the Yellow River Basin, and higher level of carbonate was associated with high level of organic carbon (Guo et al., 2016;Shi et al., 2017). One may expect a similar phenomenon in the sediment of the Yellow River Estuary. The objectives of this study were to investigate the magnitudes and spatial distributions of TOC and TIC in the surface sediment of the transitional zone near the river mouth, to evaluate the relationship between TOC and TIC, and explore the underlying mechanisms that regulate the carbon burial in the Yellow River Estuary.

2 Materials and Methods

2.1 Site description

The Yellow River Estuary is a typical river-dominated estuary with weak tides, showing a tidally affected zone of approximately 10-20 km upriver (Figure 1). The Yellow River Delta has a warm-temperate continental monsoon climate with distinct seasons. The annual mean air temperature and rainfall are 11.5-12.4 ℃ and 530-630 mm, respectively; approximately 70% of the total annual precipitation occurs in the summer, and the pan evaporation exceeds 1500 mm (Kong et al., 2015;Gao et al., 2016). In the Yellow River Estuary, monthly water temperature is 4.1 ℃ in January and 26.7 ℃ in July, and annual wind speed ranges from 3.1 to 4.6 m s^{-1} in the estuary (Shen et al., 2015). The estuary is characterized by a high sediment load (mainly composed of silt) in the water column, produced largely by the erosion from the China's Loess Plateau. Most of the sediments discharged from the modern Yellow River mouth are trapped in the subaqueous delta or within 30 km of the delta front by gravity-driven underflow (Zhao et al., 2015;Kong et al., 2015). In recent decades, the annual water and sediment fluxes have declined dramatically, which is caused by reginal climate change, reservoir construction, and irrigation-related withdrawals (Shen et al., 2015;Liu et al., 2014).

2.2 Field sampling and analyses

During October 2016, we collected 15 short sediment cores (H series) from the Yellow River Estuary using a Kajak gravity corer and 10 surface soil samples at 7 sites (S1-S7) along its upstream wetland (Figure 1b). Each sediment core was carefully extruded and cut into 1-cm interval, and then placed in polyethylene bags which were kept on ice in a cooler during transport. In the laboratory, we took the top 2 cm sediment and 0-5cm soil sample, and then freeze-dried for 48 h before analyses.

Grain size was determined using a Malvern Mastersizer 2000 laser grain size analyzer. According to Yu et al. (2015), each sediment sample and soil sample (~0.5 g)

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was pretreated, in a water bath (at 60-80 ℃), with 10-20 ml of 30% H_2O_2 to remove organic matter, and with 10-15 ml of 10% HCl to remove carbonates. The pretreated samples were then mixed with 2000 ml of deionized water, and centrifuged after 24 hours of standing. The solids were dispersed with 10 ml of 0.05 M $(NaPO_3)_6$, and then analyzed for grain size (between 0.02 and 2000 μm). The Malvern Mastersizer 2000 automatically outputs the median diameter d(0.5) (μm), the diameter at the 50th percentile of the distribution, and the percentages of clay (< 2 μm), silt (2-64 μm) and sand (> 64 μm) fractions.

C and N contents were measured using an Elemental Analyzer 3000 (Euro Vector, Italy) at the State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences. Freeze-dried samples were ground into a fine powder, then placed in tin capsules, weighed and packed carefully. For the analysis of TOC, a ~0.3 g sample was pretreated with 5-10 ml 2M HCl for 24h at room temperature, and then dried overnight at 40-50 ℃ to remove carbonate. TC and TN were analyzed without pretreatment of HCl, and TIC was calculated as the difference between TC and TOC.

For the analyses of ^{13}C in TOC ($\delta^{13}C_{org}$), approximately 0.2 g of the freeze-dried sample was pretreated with 5-10 ml 2M HCl for 24 h at room temperature to remove carbonate, and then mixed with deionized water to bring the pH to 7, and dried at 40-50 ℃ before analyses. Each pre-treated sample was combusted in a Thermo elemental analyzer integrated with an isotope ratio mass spectrometer (Delta Plus XP, Thermo Finnigan MAT, Germany). Additionally, ^{13}C and ^{18}O in carbonate ($\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$) were measured following reaction with 100% phosphoric acid on a stable isotope ratio mass spectrometer (Thermo-Fisher MAT 253, Germany), at the Nanjing Institute of Geology and Paleontology, Chinese Academy of Sciences. All the isotope data were reported in the conventional delta notation relative to the Vienna Pee Dee Belemnite (VPDB). Analytical precision was 0.1‰ for $\delta^{13}C_{org}$ and $\delta^{13}C_{carb}$, and 0.2‰ for $\delta^{18}O_{carb}$.

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2.3 Statistical methods and mapping

Correlation analyses were performed using the SPSS Statistics 19 for Windows. Spatial distribution maps were produced using Surfer 9.0 (Golden Software Inc.) and the Kriging method of gridding was used for data interpolation.

3 Results

3.1 Physical characteristics

The sampling sites covered most parts of the Yellow River estuary, with water depth ranging from 1.5 m to 13.5 m (Figure 2a). Dry bulk density (DBD) ranged from 0.74 to 1.55 g cm^{-3}, with an average of 1.02 g cm^{-3} (Table 1). Generally, DBD decreased with water depth, showing high values mainly occurred in the south and north sides near the river mouth (Figure 2b).

Figure 3 showed the spatial distributions of the main granulometric variables of the surface sediment. In general, clay content was low (1.4-10.8%), showing relatively higher values in the northern part than in the southern part. The highest clay content was found near the north side of the river mouth, and the lowest at the mouth section. Silt content was much high (69.4±21.1%), showing similar spatial distribution with clay. On the other hand, the highest content of sand was found at the mouth (Figure 3c), where clay and silt contents were lowest (Figure 3a-b). As expected, the spatial distribution of d(0.5) was similar to that of sand, showing the highest values in the shallow river mouth section and lowest in the southern bay, indicating strong hydrodynamic effect in the former and weak in the latter.

3.2 Spatial distribution of TOC, TN, C:N and $\delta^{13}C_{org}$

Concentration of TOC was highly variable, with higher values (3.2-4.4 g kg^{-1}) found in the northernmost section of the estuary and the north side near the mouth (Figure 4a). There was also high value of TOC in the bay south of the river mouth. On the other hand, lower

TOC concentration (0.2-1.4 g kg^{-1}) was observed near the mouth section. Similarly, TN value (ranging from 0.06 to 0.68 g kg^{-1}) was lowest at the river mouth and highest in the north section (Figure 4b). Overall, the spatial distribution of TN was similar to that of TOC, both showing higher values in the north deeper water area.

The C:N ratio ranged from 2.1 to 10.1 (Figure 4c). In general, C:N ratio was higher in the shallow water part relative to the deep water part. The highest C:N ratio (8-10) was found in the southern bay, and the lowest at the mouth (<4.5). Figure 4d showed a considerable spatial variability in the $\delta^{13}C_{org}$ values with a range from -24.26‰ to -22.66‰. The most negative value was observed at the river mouth and its adjacent south bay, and the least negative value far away from the mouth downward the sea. Overall, values of $\delta^{13}C_{org}$ were more negative in the shallow water section than in the deep area.

3.3 Spatial distribution of TIC, $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$

There was a large spatial variation in TIC, as shown in Figure 5a, ranging from 6.3 to 20.1 g kg^{-1}, with the highest concentration in the north deep sea area (>16 g kg^{-1}) away the mouth, and the lowest at the river mouth (<10 g kg^{-1}). Apparently, TIC also presented a high value in the southern bay. Overall, the spatial distribution of TIC was similar to that of TOC. The values of $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ ranged from -4.89‰ to -3.74‰ and -10.92‰ to -7.92‰, respectively (Figure 5b & 5c). Generally, the spatial distribution of $\delta^{13}C_{carb}$ was opposite to that of $\delta^{18}O_{carb}$, showing more negative values in the north deep sea area.

3.4 Relationship between TOC and TIC

As shown in Figure 5, there was a significantly positive correlation between TOC and TIC in the surface sediments in the Yellow River Estuary (r=0.97, p<0.01). Interestingly, the slope (2.93) was close to that (2.87) reported for the soils in the upper Yellow River Delta (Guo et al., 2016). However, the intercept was close to zero for the soils, but 7.17 in the surface estuarine sediments.

4 Discussion

4.1 Comparison of TOC with other studies

We first compared TOC concentration in the surface sediment near the river mouth in the Yellow River Estuary. Our value (0.2 to 4.4 g kg^{-1}) was slightly lower than the previous reports of 0.7-7.7 g kg^{-1} (Li et al., 2014b) and <1 to 6.0 g kg^{-1} (Liu et al., 2015). Concentration of TOC was lower in our study area relative to those in the other coastal areas of the Bohai Sea, i.e., north off the Yellow River Estuary (2.6-17.2 g kg^{-1}) (Yuan et al., 2004) and the Laizhou Bay (5.7-12.8 g kg^{-1}) (Wang et al., 2017).

As given in Table 2, the Yellow River Estuary also had relatively lower TOC values than other estuaries in China. There was an increasing trend in TOC from north to south (i.e., Yellow River Estuary < Yangtze River Estuary < Pearl River Estuary), which might be related to the differences in climatic conditions and estuarine sedimentary environments. The warmer and humid climate with a longer growing season to the south would enhance biological production in the water column and sedimentation of organic materials (Dong et al., 2012;Yu et al., 2015a). In addition, the vegetation (i.e., mainly mangroves) grown in the Pearl River Estuary of the South China had much higher carbon sequestration capacity than those (i.e., tidal marshes and seagrass beds) in the Yangtze River Estuary and Yellow River Estuary (Wang et al., 2016d;Pendleton et al., 2012).

Sedimentary TOC concentration in large river estuaries was relatively lower than in small river estuaries, e.g., the Luan River Estuary, Licun Estuary, Min River Estuary and GQ Estuary (Table 2), indicating that the weak hydrodynamic environment (in the small estuaries) was beneficial to the burial of organic carbon (Liu et al., 2015;Ramaswamy et al., 2008). The Yellow River Estuary, and most estuaries in China, generally showed much lower TOC values in the surface sediment than those in the South and Southeast Asia, Europe, North America and South America. The differences in TOC levels may be associated with the geomorphology of the estuary, the magnitude and stoichiometry of nutrient inputs, and other driving mechanisms (Bauer et al., 2013;Cai, 2010).

4.2 Dynamics of TOC and regulating mechanisms

Our study demonstrated large spatial variability in the TOC of the surface sediment in the Yellow River Estuary, with relatively higher values in the north section (2.6±1.5 g kg^{-1}) than in the south section (1.6±0.2 g kg^{-1}) (Table 3). The surface sediments were finer to the north than to the south (Figure 3). In general, fine-grained marine sediments contain higher organic carbon than coarse marine sediments (Canfield, 1994;Hu et al., 2016). On the other hand, coarser (finer) sediment particles usually indicated a stronger (weaker) water energy environment (Molinaroli et al., 2009;Molinaroli et al., 2014). These analyses indicated that the relatively lower TOC values to the south and in the river mouth were attributable to higher kinetic energy level.

The magnitude and spatial distribution of TOC in estuarine sediment may reflect multiple and complex processes (Hu et al., 2016;He et al., 2010). Apart from the estuary's own characteristics, such as the river plume and tidal straining effect (Wang and Wang, 2010;Xu et al., 2013), other factors may have influences on the dynamics of TOC in the surface sediment. For example, land use changes such as industrial and agricultural development would enhance the riverine input of nutrients and organic materials, leading to changes in estuary productivity and TOC burial in the sediment (Yu et al., 2014;Lin et al., 2002;Liu et al., 2012). As shown in Table 4, there was a significantly negative relationship between the $\delta^{13}C_{org}$ value and water depth (r=0.71, p<0.01), implying that the shallow sections in the Yellow River Estuary accumulated more allochthonous OC (with more negative $\delta^{13}C_{org}$ values).

Sedimentary TOC in estuaries may include autochthonous and allochthonous sources (Bianchi and Allison, 2009;Baijulal et al., 2013). Generally, aqueous organic matter has a lower C:N ratio and less negative $\delta^{13}C_{org}$ value than terrigenous source (Lamb et al., 2006;Meyers, 1997). The relatively low C:N ratio (6.3±1.7) and $\delta^{13}C_{org}$ value (-23.35±0.48‰) in our study indicated that TOC was mainly autochthonous in the surface sediment the Yellow River Estuary. However, C:N ratio was relatively higher (8.8±1.8) in the southern shallow bay (Table 3). Such high C:N ratio together with relatively more negative $\delta^{13}C_{org}$ value (-23.91±0.50‰) (Table 3) suggested that there might be some allochthonous OC sources. Using the average C:N ratio (10.8 g:g) from the soils collected near the river mouth (Table 1), and assuming 6.6 mol:mol as the marine end-member, we estimated that 60.8% of TOC was from soil OC source, indicating that the southern bay might have accumulated a significant amount of terrigenous OC.

4.3 Dynamics of TIC and underlying mechanisms

There have been only a few studies of inorganic carbon from the estuarine sediments (Table 2). According to these limited studies, the Yellow River Estuary has much higher TIC values than those (3.3-8.2 g kg^{-1}) in the Cochin Estuary, Vellar and Coleroon Estuary, and Chilika Lagoon of the South Asia. Our study showed large spatial variability in TIC of the surface sediment in the Yellow River Estuary, with relatively higher values in the north section (14.5±4.7 g kg^{-1}) than in the south section (12.2±1.2 g kg^{-1}) (Table 3), which was consistent with TOC. Our analyses revealed a significantly positive correlation between TIC and TOC ($r=0.97$, $p<0.01$) in the surface sediments, indicating that production of organic carbon might have a large influence on the formation of carbonate (Paprocka, 2007;Li et al., 2012). Accordingly, it is reasonable to believe that most TIC was from autogenic carbnate in the surface sediment of the Yellow River Estuary.

The recent study of Liu et al. (2014) demostrated that the rate of $CaCO_3$ precipitation exceeded that of OC production by a factor of <2 in the water columns of the Yellow River Estuary whereas the earlier study of Gu et al. (2009) showed a ratio of 3.6 for IC:OC in particles. We found that TIC concentration was six times of TOC concentration in the surface sediment of the Yellow River Estuary, which indicated that apart from the relatively higher $CaCO_3$ production in the upper water column, there would be a decrease of TOC and/or an increase of TIC during sedimentation and after burial. Organic matter is often subject to deocmposition process in the surface sediments, which will cause a decrease in TOC (Alkhatib et al., 2012;Koho et al., 2013;Rieling et al., 2000). Meantime, CO_2 production owing to TOC decomposition would promote carbonate formation in sediemnts (Zhao et al., 2015;Wang et al., 2016b;Gu et al., 2009). Taking the southern bay

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as an example, both TIC and TOC concentrations were relatively higher, and both $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$ were more negative than in other sections (Table 3). Such relationship suggested that there might be some transfer of OC to IC in the surface sediment in the bay.

There was a significant negative correlation between $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ in the Yellow River Estuary (r=-0.84, p<0.01), which was much different to those found in some inland waters (Yu et al., 2015b;Xu et al., 2006;Wang et al., 2002), which may reflect complex impacts of various processes in the Yellow River Estuary (e.g., riverian input, coastal runoff, ocean currents). There have been human activities over the past decades in the Yellow River Basin, including intensive irrigation and damming, and direct human regulation of water and sediment discharge (Liu et al., 2014;Gu et al., 2009;Zhang et al., 2013), which would impact on the biogeochemical processes in the Yellow River Estuary. Further studies are needed to assess the spatial and temporal variations in the carbon fields of water column and long sediment core, in order to better understand the carbon cycle in the Yellow River Estuary and the impacts of human activity and climate change.

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Table 1. Means, standard deviation (SD) and coefficients of variation (CV) of the main variables.

		DBD	d0.5	Clay	F-Silt	M-Silt	C-Silt	Sand	TN	TOC	TIC	C/N	$\delta^{13}C_{org}$	$\delta^{13}C_{PDB}$	$\delta^{18}O_{PDB}$
		g cm^{-3}	μm	%					g kg^{-1}				‰		
Sediment	Mean	1.02	34.2	6.1	34.8	13.7	20.9	24.5	0.36	2.3	14.1	6.3	-23.35	-4.36	-8.92
	SD	0.20	26.3	2.9	19.0	6.7	9.4	23.8	0.19	1.3	4.0	1.7	0.48	0.41	0.90
	CV	0.20	0.77	0.47	0.55	0.49	0.45	0.97	0.53	0.57	0.28	0.27	-0.02	-0.09	-0.10
Soil	Mean	/	28.2	4.7	31.1	25.8	25.8	12.5	0.77	8.9	12.9	10.8	-22.5	-4.0	-9.1
	SD	/	15.8	1.2	15.5	9.3	10.4	15.1	0.55	8.0	4.8	3.0	3.4	0.72	0.62
	CV	/	0.56	0.26	0.50	0.36	0.40	1.21	0.71	0.90	0.37	0.27	-0.15	-0.18	-0.07

Clay:<2 μm, F-Silt: 2-16 μm, M-Silt:16-32 μm, C-Silt:32-64 μm, Sand:>64 μm

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Table 2. Summary of TC, TOC and TIC (g kg^{-1}) values in surface sediments from estuaries in the world.

		Location	TC g kg^{-1}	Mean g kg^{-1}	TOC g kg^{-1}	Mean g kg^{-1}	TIC* g kg^{-1}	Reference
Aisia	China	Luan River Estuary			0.4-14	4.8		Li et al. (2016)
		Yellow River Estuary			0.7-7.7	4.2		Li et al. (2014b)
					<1-6.0	3.1		Liu et al. (2015)
			6.6-24.5	**16.4**	**0.2-4.4**	**2.3**	**14.1**	**This study**
		Licun Estuary			6.0-20			Yu et al. (2009)
		Yangtze River Estuary			1.0-7.0			Zhou et al. (2007)
					1.2-6.8			Li et al. (2014a)
					0.9-14.3			Liu et al. (2006)
					0.1-15.9	5.2		Wang and Xian (2011)
					6.0-15			Gao et al. (2008)
		Zhangjiang Estuary			7.4-14.9			Xue et al. (2009)
		Min River Estuary			13.6-22.1			Jia et al. (2008)
		Danshui River Estuary			2.9-17.1			Hung et al. (2007)
		Pearl River Estuary			6.0-44.1	12.1		He et al. (2010)
					10-14			Ye et al. (2012)
					0.6-10.2	5.4		Hu et al. (2006)
					0.9-28.3			Zhang et al. (2015)
		GQ Estuary			8.5-23.6			Yang et al. (2014)
	India	Mandovi Estuary			1.0-30	10.5		Alagarsamy (1991)
					1.0-32.3			Nasnolkar et al. (1996)
		Cochin Estuary	9.8-28.5	22.4	5.5-25.9	19.1	3.3	Gireeshkumar et al. (2013b)
			8.5-34.1	24.3	4.2-27.7	18.8	5.5	
			3.0-32	20	2.6-29.9	15.6	4.4	

					3.0-32.6	21		Gireeshkumar et al. (2013a)
		Kozhikode and Kannur River Estuary			18-70			Manju et al. (2016)
		Ashtamudi River Estuary			9.5-50.2	19.4		Baijulal et al. (2013)
		Kadinamkulam River Estuary			9.9-77.1	31.8		
		Godavari Estuary			35-147			Krupadam et al. (2003)
		Vellar Estuary and Coleroon Estuary	20.6-28.1	24.4	14.1-18.3	16.2	8.2	Prasad and Ramanathan (2008)
	Bengal	Chilika lagoon	3.4-19.7	12.1	2.6-16.6	8.6	3.5	Nazneen and Raju (2017)
	Malaysia	Setiu Estuary			7.0-34			Thornberg et al. (2014)
					1.0-68			Ellis et al. (2014)
Europe	Portuguese	Douro River Estuary			0.1-9.3			Dessandier et al. (2016)
		Mondego River Estuary			0-8.2			
		Estremadura River Estuary			0.1-10.7			
		Tagus River Estuary			0.1-7.0			
		Sado River Estuary			3.7-8.4			
	France	Loire Estuary			0.2-38.2	13.9		Coynel et al. (2016)
	UK	Humber Estuary			8.6-72.5	29.6		Andrews et al. (2008)
		Forth Estuary			31-61	47		Graham et al. (2001)
North America	USA	Gulf of Mexico Microtidal Estuaries			4.6-14			Darrow et al. (2017)
		Cedar and Ortega River Estuary			23-226	127		Ouyang et al. (2006)
		Hudson River Estuary			10.6-27.3			
		Mullica River Estuary			2.2-19.5			Medeiros et al. (2012)
		Pawtuxet River Estuary			0.7-44			Cantwell et al. (2016)
South America	Brazil	Caravelas Estuary			0.9-64			Sousa et al. (2016)

539 * $TIC = TC_{mean} - TOC_{mean}$

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Table 3. Means and standard deviation of the carbon variables in different water sections

Section#		**TOC**	**TIC**	**C/N**	**$\delta^{13}C_{org}$**	**$\delta^{13}C_{PDB}$**	**$\delta^{18}O_{PDB}$**
North	Mean	2.6	14.5	6.1	-23.23	-4.53	-8.84
	SD	1.5	4.7	0.7	0.32	0.43	1.08
South	Mean	1.6	12.2	6.2	-23.11	-4.03	-9.1
	SD	0.2	1.2	1.1	0.26	0.18	0.42
Bay	Mean	2.7	15.4	8.8	-23.91	-4.54	-8.75
	SD	1.4	3.8	1.8	0.5	0.36	0.67

North: H2, H3, H7, H8, H13, H14; South:H4, H6, H10, H11; Bay: H5, H26

Table 4. Correlation coefficient (r) between various variables for the sediments.

	Depth	DBD	d0.5	Clay	Silt	Sand	TOC	TIC	$\delta^{13}C_{org}$	$\delta^{13}C_{carb}$
TOC	0.54*	-0.65**	-0.94**	0.97**	0.88**	-0.90**				
TIC	0.63*	-0.70**	-0.96**	0.93**	0.93**	-0.94**	0.97**			
$\delta^{13}C_{org}$	0.71**	-0.37	-0.55*	0.55*	0.53*	-0.54*	0.50	0.47		
$\delta^{13}C_{carb}$	-0.54*	0.73**	0.90**	-0.88**	-0.85**	0.87**	-0.90**	-0.93**	-0.32	
$\delta^{18}O_{carb}$	0.63*	-0.65**	-0.98**	0.94**	0.96**	-0.97**	0.91**	0.93**	0.63*	-0.84**

Significance of Pearson correlation is marked with one ($p<0.05$) and two ($p<0.01$) superscripts.

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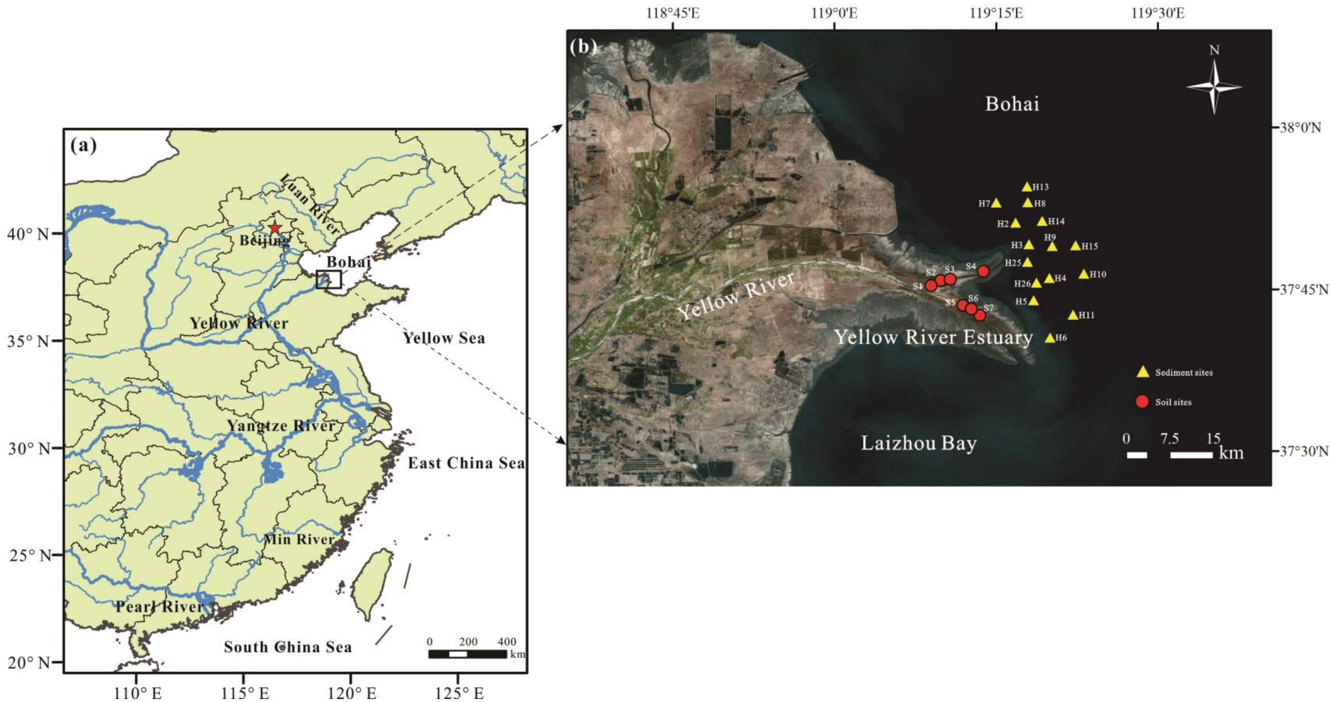

Figure 1. Map of (a) the large Chinese river-estuarine systems and (b) the Yellow River Estuary with the sampling sites. Remote sensing imagery was plotted by using software ArcGIS 10.2 and Corel DRAW X7

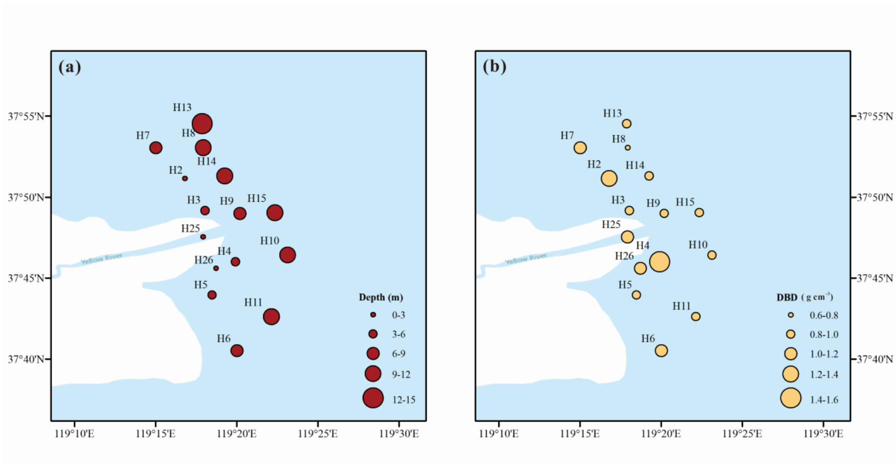

Figure 2. Spatial distributions of **(a)** depth (m) and **(b)** dry bulk density (DBD, g cm^{-3}) in surface sediments of the Yellow River Estuary.

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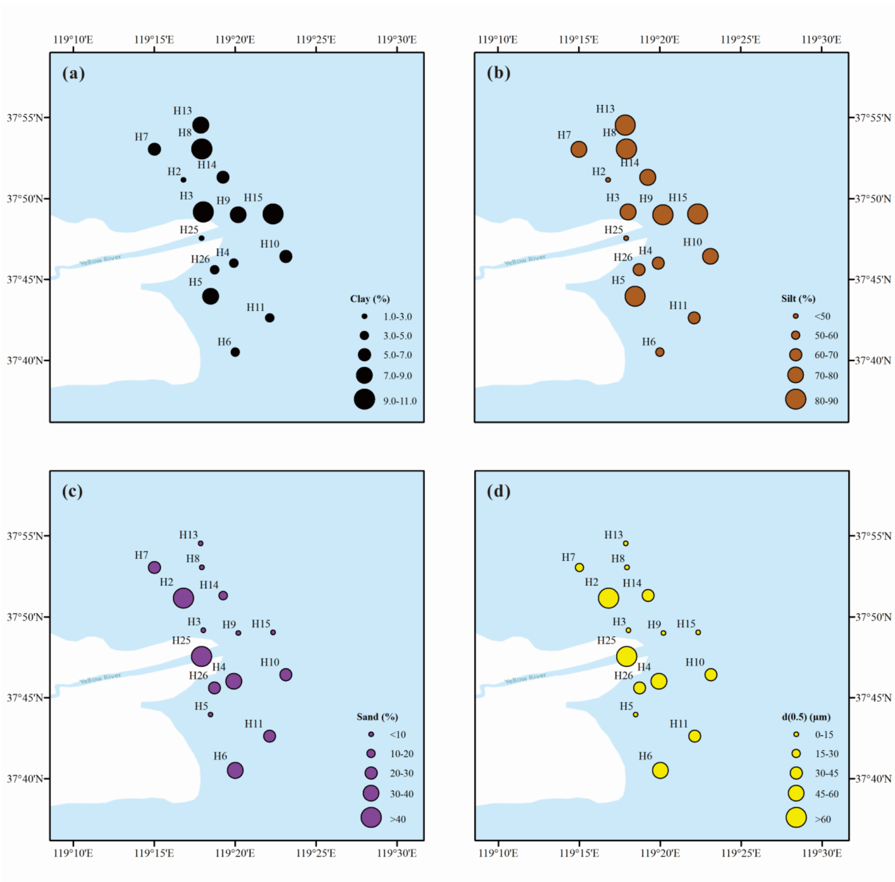

Figure 3. Distributions of **(a)** clay (%), **(b)** silt (%), **(c)** sand (%), **(d)** the median diameter (d(0.5), μm) in surface sediments of the Yellow River Estuary.

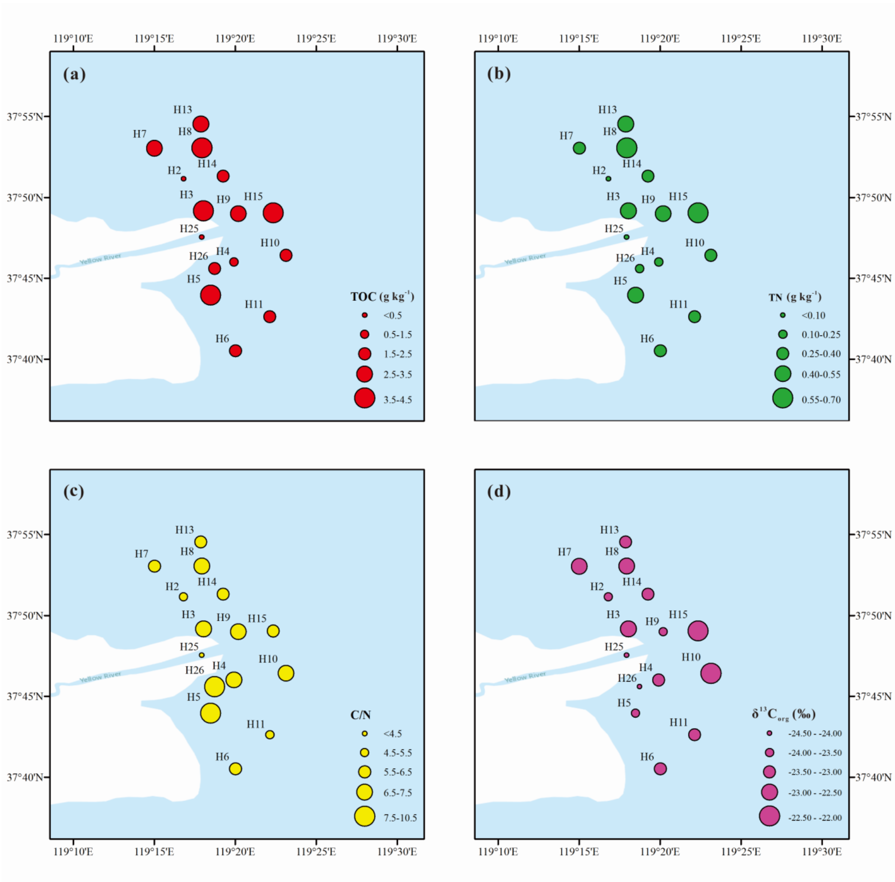

Figure 4. Spatial distributions of **(a)** TOC (g kg^{-1}), **(b)** TN (g kg^{-1}), **(c)** C/N, **(d)** $\delta^{13}C_{org}$ (‰) in surface sediments of the Yellow River Estuary.

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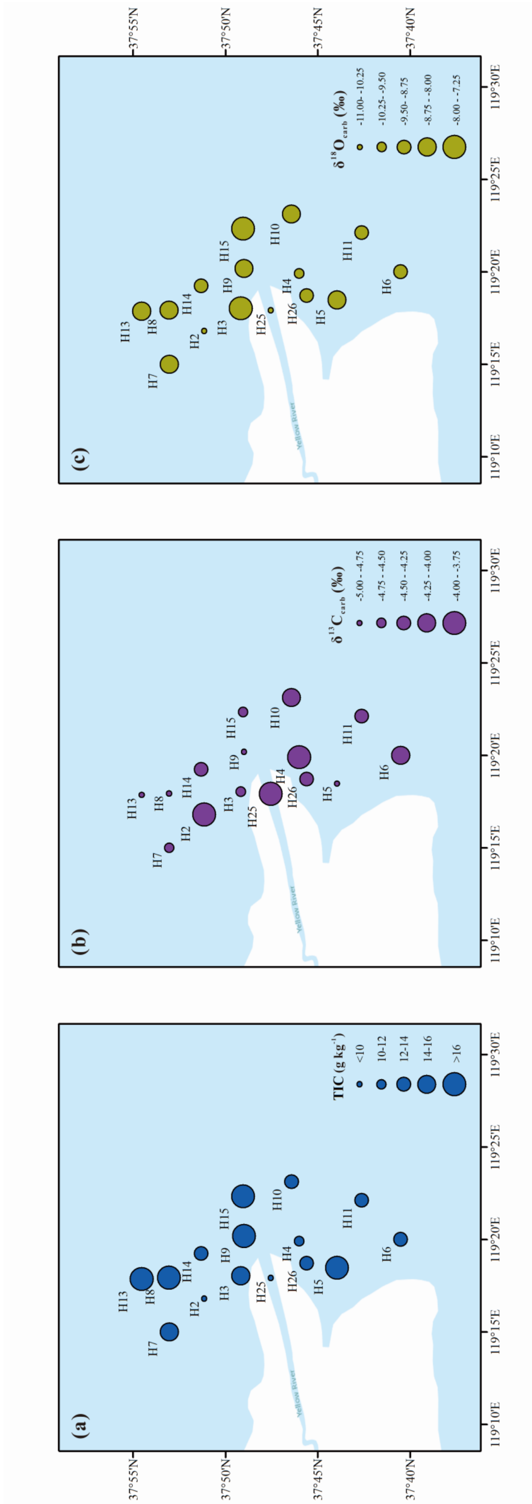

Figure 5. Spatial distributions of **(a)** TIC (g kg^{-1}), **(b)** $\delta^{13}C_{carb}$ (‰), and **(c)** $\delta^{18}O_{carb}$ (‰) in surface sediments of the Yellow River Estuary.

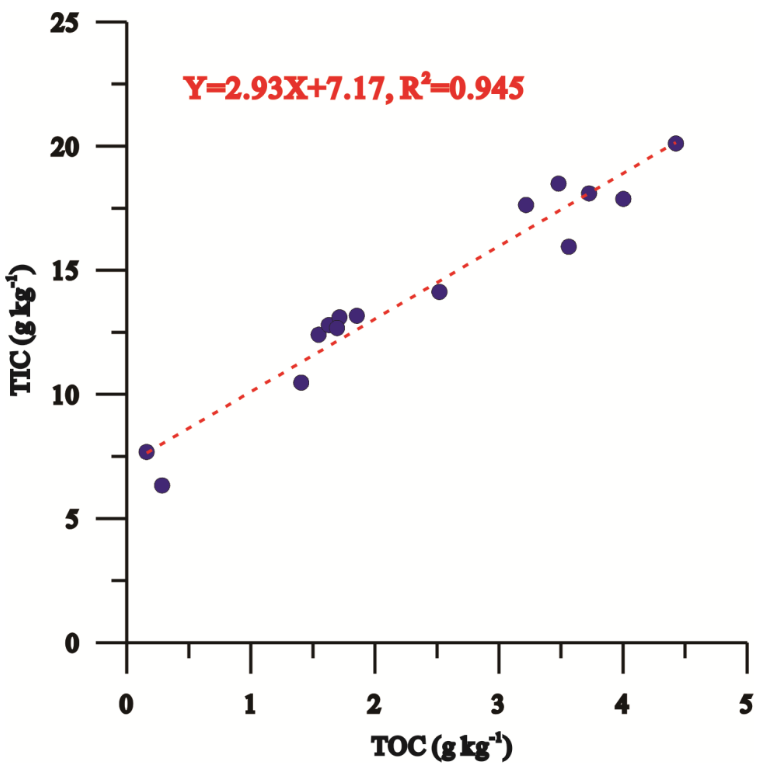

Figure 6. Relationship between TOC and TIC in surface sediments of the Yellow River Estuary.