



Accumulation, sources and pollution of heavy metals in the sediments of coastal tidal flats in the North Jiangsu Radial Sand Ridges, China

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Received: 23 May 2018 / Accepted: 5 February 2019 / Published online: 19 February 2019
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Abstract

In 2014, three sediment cores were collected from the northern [Xinyanggang (XYG)], middle [Chuangdonggang (CDG)], and southern [Xiaoyangkou (XYK)] parts of the coastal tidal flats of the North Jiangsu radial sand ridges (NJRSR), East China. The grain size and contents of heavy metals (Cu, Zn, Cr, Ni, Pb and As) in the cores were detected. The features of sediments and the accumulation of heavy metals were also investigated. Multivariate analyses (principal component analysis and correlation analysis) were applied to identify the sources of heavy metals. The enrichment factors (EF) were calculated to estimate the level of contamination stored in these sediments. The results showed that the vertical variations in the contents of heavy metals were compatible with the sedimentary features. The grain size in core XYG became coarser around the 1980s and the contents of heavy metals began to decrease. The grain size in core CDG was suddenly refined around the 1960s and the contents of heavy metals suddenly decreased. The grain size in core XYK coarsened initially before becoming finer and the contents of heavy metals were basically stable. The contents of heavy metals of the NJRSR coastal tidal flat were subject to the sedimentary parent materials of the abandoned Yellow River Delta and the offshore radial sand ridges. Several heavy metals were disturbed by anthropogenic activities after the middle and late 1980s. The content of Cr in core XYG was mainly from industries. The contents of Pb and Zn in core CDG were from agricultural, urban and industrial sources. Cu and As in XYK were from agricultural and industrial sources. The environment of sediments in the study area was generally good, but was polluted by individual elements after industrialisation. Cr in core XYG was moderately and significantly polluting, Pb and Zn in core CDG were lightly polluting, Cu was moderately and significantly polluting, and As was lightly polluting in core XYK.

Keywords Sediment cores · Heavy metals · Source analysis · Pollution · Radial sand ridges, Jiangsu Province, China

Electronic supplementary material The online version of this article (<https://doi.org/10.1007/s12665-019-8125-3>) contains supplementary material, which is available to authorized users.

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Introduction

The rapid development of China's coastal economy in the past decades has led to the continuous increase in the intensity of anthropogenic activities and the flow of a great quantity of heavy metal pollutants into the ocean through surface runoff and atmospheric precipitation. Tidal flats, which are the transitional belt between the sea and the land, bear the brunt of accommodating heavy metal pollutants that enter into the ocean. Heavy metal pollutants are adsorbed, coordinated, flocculated, and settled into sediments by particulate matter after entering marine waters; the pollutants then return into the waters through the desorption mechanism. Therefore, tidal flat sediments are not only a 'sink' of heavy metals but also a 'secondary source'. Heavy metals in sediments might cause serious and widespread environmental problems due to their

toxicity, persistence and non-biodegradable nature (Ira-bien and Velasco 1999; Wu et al. 2017). Since a series of catastrophic heavy metal pollution events occurred in the 1950s [‘minamata disease’ (Hg) and ‘itai-itai disease’ (Cd)], the adverse impact of heavy metal pollution has attracted much attention worldwide (Peerzada and Rohoza 1989; Cobelogarcía and Prego 2003; Fatoba et al. 2016; Bárcena et al. 2017; Jahan and Strezov 2018). Studies on heavy metals in tidal flat sediments began in the 1970s. Spatio-temporal distribution, pollution and the source of heavy metals have been research hotspots in recent years (Sreekanth et al. 2015; Sindern et al. 2016; Nethaji et al. 2017). Since the 1980s, heavy metals in sediments from estuaries, gulfs and offshore areas in China have been surveyed (Liu et al. 2015; Song et al. 2017; Wang et al. 2017b). Research on heavy metals in sediments from tidal flats in Jiangsu Province has focused on surface sediments (Xu et al. 2014, 2018). Sediment cores from gulfs, estuaries and other typical sea areas have also been investigated (Zhang et al. 2013; Li and Li 2017). However, few reports have systematically analysed sediments from coastal tidal flats in the North Jiangsu radial sand ridges.

The North Jiangsu radial sand ridges (NJRSR) are dominated by two major tidal wave systems, namely, the forward tidal waves of the East China Sea and the rotary waves of the South Yellow Sea. Massive radial sand ridges were developed off the coast due to a combination of the abundant sediment sources of the ancient Yellow River and the ancient Yangtze River. The Yellow River (flowing northward) and the Yangtze River (running southward) both possess substantially low terrestrial sediment input; the modern sources of sediments of the coastal tidal flats are mainly the abandoned Yellow River delta and the offshore radial sand ridges of Jiangsu Province (Xing et al. 2012; Wang et al. 2014; Zhou et al. 2014). With the development of the Jiangsu coastal economy, more than 70 drainage canals (e.g., the Sheyang River, the Wanggang River and the Bencha River), have provided a large number of land-based pollutants into the sea. Heavy metals discharged into the near-shore area by these rivers have probably affected the environment of the coastal tidal flat. The columnar sediments contain abundant and continuous sedimentary information indicative of the sources, distribution and migration of heavy metals. Therefore, this study collected three typical sediment cores (XYG, CDG & XYK) from the northern, middle and southern parts of the NJRSR coastal tidal flats, East China. Sedimentary features and the accumulation of heavy metals were explored. The sources and pollution of heavy metals were analysed. Results provide a reference for modelling and environment quality assessment and management of the NJRSR coastal tidal flats.

Materials and methods

Study area

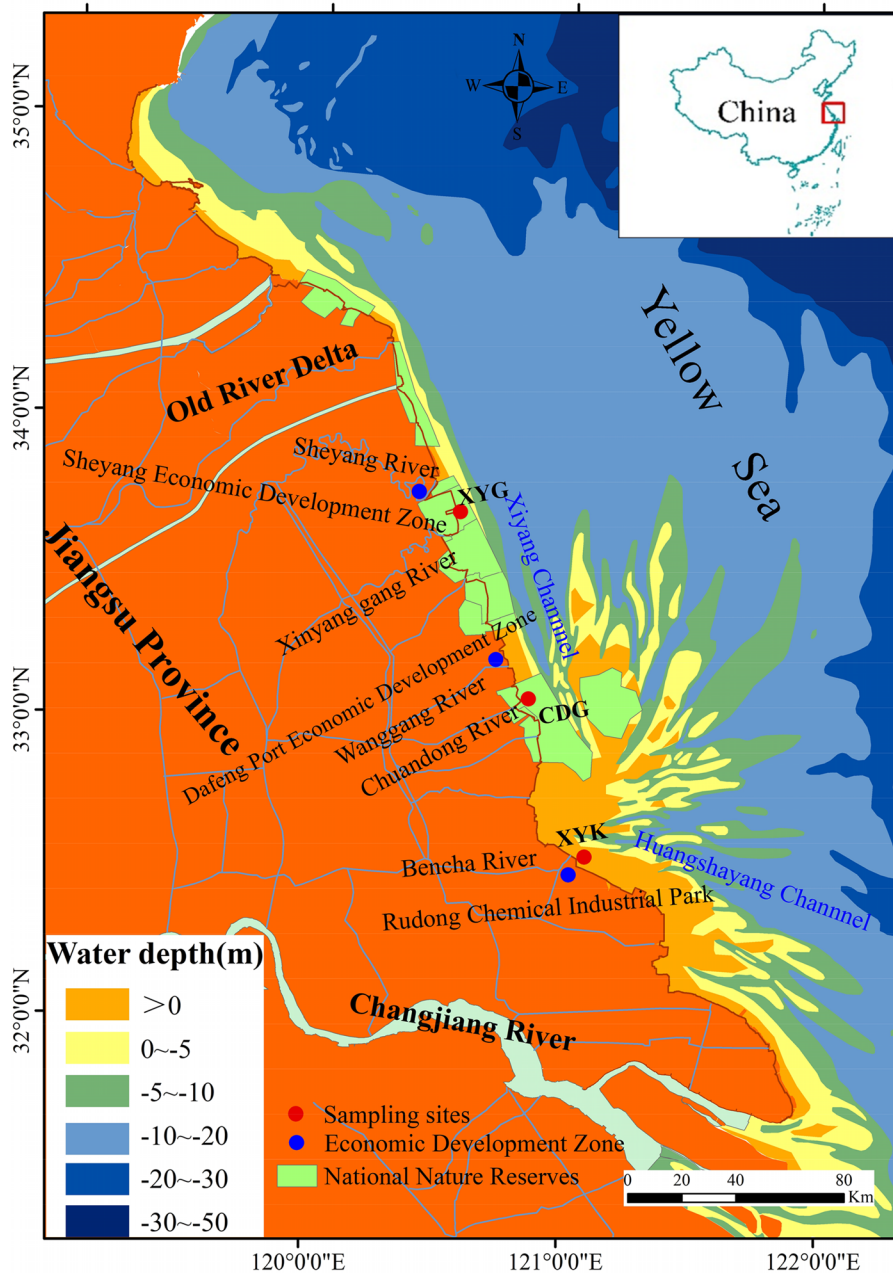
The NJRSR (32°00′N–33°48′N, 120°40′E–122°10′E) is located on the east coast of Jiangsu, ranging from the estuary of the Sheyang River in the north to Qidongzui in the south (Fig. 1). These ridges are of a unique landform type in Jiangsu Province and have an apex roughly at Jianggang, Dongtai City. These ridges consist of 10 large stripped undersea sand ridges that extend radially to the north, northeast, east, and southeast until the depth contour at –25 m below sea level. Each sand ridge is approximately 10 km wide and 100 km long. More than 70 sandbars are exposed at low tide and their total area, with theoretical depth above 0 m, is approximately 2100 km². Sandbars have a complex topography and special forms. The major tidal channels include the Xiyang Waterway off the estuary of Wanggang, the Huangshayang Waterway and the Lanshayang Waterway off XYK, and the Xiaomiaohong Waterway off Lvsi. The NJRSR is the product of interactions between matter and energy in specific environments. At present, foreign sediments minimally influence the radial sand ridges. The radial sand ridges undergo scouring outside and silting inside (Li et al. 2001). The substances scoured outside and moving towards the shore provide abundant sediment sources for the continuous deposition of the coastal tidal flats (Xing et al. 2012). A considerable amount of land reserve resources are provided every year for the lack of arable land at Jiangsu Province and a vast space is provided for future development of coastal areas.

Sampling and laboratory analysis

Three sediment cores were collected from the NJRSR coastal tidal flat using a portable gravity sampler from October to December 2014. The sampling points from north to south are located at the naked tidal flats at points including: 5 km north of Xinyanggang (XYG), Sheyang County; north of Chuandonggang (CDG), Dafeng County; and south of Xiaoyangkou (XYK), Rudong County. The 130–150 cm long sediment cores were sealed on site, placed horizontally, transported back to the laboratory, cut open longitudinally and photographed. Lithology and structures of the sediments were described. Sub-sampling was performed at every 2 cm. Subsamples were freeze dried for 24 h, processed by removing sea shells, plant roots, gravel and other impurities and used for analysis of heavy metals and grain size.

Heavy metals and grain size were analysed and tested at the Analytical Test Centre of Nanjing Normal University.

Fig. 1 Location of the study area



About 0.1 g (± 0.0001 g) of sediment sample was accurately weighed and digested in a microwave digestion system (MARS, CEM, USA), with gradually rising temperature according to 130–185 °C. 3 ml of HNO₃, 3 ml of HF and 1 ml of HClO₄ was added to the Teflon microwave digestion tank. Acid was removed using a heating plate after digestion at 200 °C, and the digestion tank was purged with a small amount of ultrapure water (Nguyen et al. 2016). The final volume was placed into a sample bottle. Each digested sample was measured using Prodigy plasma atomic emission spectrometer (ICP-AES Prodigy xp, Leeman, USA) for the geochemical elements

(Cu, Zn, Cr, Ni, Pb and Al) under the following condition: wavelength range of 165–800 nm, real-time sync background correction, resolution ≤ 0.0048 nm (in the depth of 200 nm) and precision RSD% < 1.0% ($n = 10$). As content was determined by Chinese-made Haiguang AFS-230 atomic fluorescence spectrometer, with detection limit < 0.06 µg/L and precision RSD% < 1.5%. About 0.2 g (± 0.0001 g) of sediment sample was accurately weighed into a 50 ml colorimetric tube covered with a plug. After adding 10 ml of aqua regia, the treated sample was placed in a boiling water bath for 2 h (Wu et al. 2014). During this period, the sample was shaken up, removed and cooled

to room temperature. Ultrapure water up to the volume of 50 ml was added, shaken well and settled. The supernatant was loaded for testing.

Reagents used in the analysis of heavy metals were all guaranteed reagent, and ultrapure water was used. Standard reference materials GBW-07404 (GSS-4, Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences) and blank samples were used for process quality control. The measured values of the seven elements were within the permissible error range under the national standards for reference materials, with relative test error less than 5%. The concentration determined in the extract was corrected by subtracting the corresponding blank values from the test results and expressing this as dry weight.

Grain size was analysed using British Malvern MS 2000 laser particle size analyser under the following condition: measurement range of 0.02–2000 μm , resolution of 0.1 μm and relative repeated measurement error less than 3%.

Data processes

Enrichment factor

The enrichment factor (EF) proposed by Chester and Stoner (1973) is often used to analyse the severity of heavy metal pollution (Shafie et al. 2013; Gaşiorek et al. 2017; Wang et al. 2017a). Al, an inert element during migration, is chosen as the reference element to eliminate the effects of particle size on heavy metal concentration in sediments (Soto-Jiménez and Páez-Osuna 2001; Wang et al. 2015). The calculation formula is as follows:

$$EF = \frac{M_0/Al_0^i}{M_B/Al_B^i}, \quad (1)$$

where EF is the enrichment factor, M_0 is the content of heavy metals in tidal flat sediments, M_B is the background value of heavy metals, Al_0^i is the content of Al in tidal flat sediments and Al_B^i is the background value of Al. As proposed by Sutherland (2000), the pollution severities of heavy metals can be divided into six levels based on enrichment factor: $EF < 1$, non-polluting; $1 < EF < 2$, non-polluting or lightly polluting; $2 < EF < 5$, moderately polluting; $5 < EF < 20$, significantly polluting; $20 < EF < 40$, severely polluting; and, $EF > 40$, extremely severely polluting.

The background value of a heavy metal refers to the normal content of such element in the natural unpolluted environment. This parameter indicates the chemical composition and accumulation features of sediments on a time scale and is an important basis for judging the sources of heavy metals and evaluating the severity of pollution. The content of

heavy metals in sediments before industrialisation was taken as the background value and best reflects the lithology of sediments (Cobelogarcía and Prego 2003). In this study, the background value of heavy metals in each core is the average value of heavy metal content in the deeper part of the core (Table 1).

Statistical analysis

The sources of heavy metals were determined by principal component analysis (PCA) and correlation analysis (CA) (Nguyen et al. 2016; Harikrishnan et al. 2017). In PCA, the relevant laboratory data was entered into a factor analysis template in SPSS21.0 software. The KMO values based on the element matrices were greater than 0.6. The suitability of the data for PCA was judged from the P value obtained by Bartlett's sphere test. Principal components were extracted by the eigenvalue $\lambda \geq 1$. In correlation analysis, the relevant laboratory data was entered into the Pearson's coefficient correlation analysis template in SPSS21.0 software. Statistical significance was determined with P above 0.01 or 0.05.

Results and discussion

Sedimentary features

The mean sedimentation rates of the sediment cores from XYG, CDG and XYK were 1.27, 1.75–1.85 and 2.43–3.3 cm/year (Meng 2018), respectively. According to the sedimentation rates, the corresponding depositional ages were 1900–2014 (114 years), 1935–2014 (79 years) and 1965–2014 (49 years). The industrialisation of the coastal zone of Jiangsu began in the middle of the 1980s, so the three cores could be used to analyse the impact of industrialisation of the Jiangsu coastal zone on the tidal flats.

The cross-sectional view of the cores is shown in Fig. 2. Core XYG can be divided into two layers of 43 cm depth (1980): the upper layer (43–0 cm, 1980–2014), with the mean particle size of 4.28Φ , mean clay content of 5.80% and dominated by silt; and the lower layer (145–43 cm, 1900–1980), with the mean particle size of 4.73Φ , mean clay content of 5.13% and dominated by silt. The grains significantly coarsened at 43 cm depth (1980). Sediments from the tidal flats of XYG were mainly sourced from the abandoned Yellow River delta (Zhang 1990; You et al. 1998; Zhang et al. 2012). The abandoned Yellow River undersea delta was basically levelled around 1980s (Wang 2006; Zhang et al. 2014). Low tidal flats or undersea bank slopes suffered significant erosion (Chen et al. 2010a; Zhang et al. 2016; Zhang 2016), and the erosion was coarser (Zhang and Chen 2012; Zhang 2016). The accumulation of coarse

Table 1 Summary of various data in three cores

	XYG	BG ^a	EF ^b	CDG	BG		XYK	BG	EF
					0–90 cm	91–143 cm			
Sampling time	Oct. 2014			Dec. 2014			Nov. 2014		
Core length	145 cm			143 cm			131 cm		
Time period	1900–2014			1935–2014			1965–2014		
Sand (%)	7.95–47.66 (30.11)			1.07–74.06 (31.91)			19.42–64.46 (37.5)		
Silts (%)	46.82–84.60 (64.29)			23.48–85.80 (60.35)			29.30–72.78 (54.8)		
Clay (%)	2.48–13.19 (5.60)			0–19.10 (7.74)			2.38–13.23 (7.69)		
Cu (ug/g)	1.37–20.19 (7.47)	7.08	0.20~2.47 (0.95)	0.40~13.71 (4.95)	4.07	3.10	0.13~2.94 (1.34)	0.76	0.21~6.47 (2.59)
Zn (ug/g)	14.51~72.57 (40.52)	37.49	0.50~2.85 (0.98)	5.75~55.02 (25.03)	17.24	27.20	0.45~2.34 (1.21)	5.16	0.22~2.95 (1.17)
Cr (ug/g)	1.15~107.25 (37.96)	27.25	0.04~5.34 (1.25)	13.02~51.56 (32.03)	28.58	31.22	0.59~1.97 (1.11)	27.85	0.48~1.80 (1.03)
Ni (ug/g)	9.01~27.88 (16.71)	16.58	0.46~2.45 (0.93)	10.13~35.64 (21.98)	23.83	14.72	0.72~1.66 (1.05)	19.89	0.72~1.60 (1.09)
Pb (ug/g)	13.92~40.19 (22.06)	19.26	0.72~1.89 (1.01)	0.99~27.43 (14.32)	8.47	21.29	0.17~1.80 (1.05)	3.96	0.27~1.83 (1.07)
As (ug/g)	3.61~9.02 (5.65)	5.81	0.34~2018 (0.93)	4.32~11.33 (6.69)	6.97	4.98	0.55~1.94 (1.03)	3.55	0.56~2.42 (1.33)
Al (%)	0.84~3.12 (2.26)	2.08	1.09	1.32~3.66 (2.73)	2.73	2.31	1.80~4.32 (2.94)	2.96	0.99

^aBG is background value of the average core content at deeper depths. Due to changes in the sources of CDG materials in the 1960s, two background values were used for heavy metals in the core CDG

^bEF is enrichment factors and average

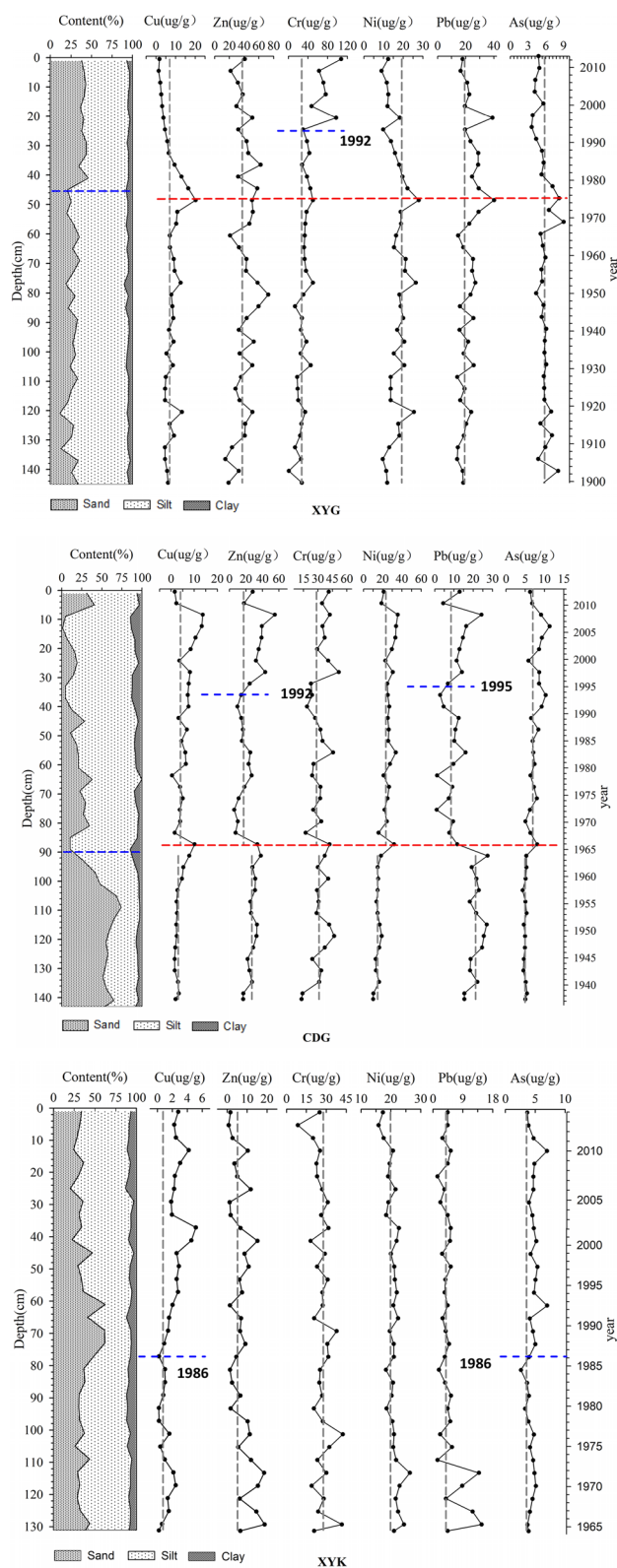


Fig. 2 Vertical distribution of heavy metal contents and sediment components in sediment cores. Vertical dashed lines show background concentration value

particles on the coast resulted in the coarsening of sediments in core XYG above 43 cm (1980).

Core CDG can be divided into two layers of 90 cm depth (1965): the upper layer (90–0 cm, 1965–2014), with the mean particle size of 5.21Φ , mean clay content of 9.11%, mainly composed of silt with significant fluctuations in the grain size; the lower layer (143–90 cm, 1935–1965), with a mean particle size of 4.12Φ , mean clay content of 5.41% and composed mainly of silt. The grain size of sediments refined and fluctuated as a whole above 90 cm (1965). Core CDG is near the Xiyang Waterway, which ran through the vicinity of the core CDG in the 1950s–1960s (Zhang and Chen 1992; Xu et al. 2012; Chen et al. 2017) and separated the coastal tidal flat from the offshore radial sand ridges. The Xiyang Waterway is an important channel for the transport of eroded sediment of the abandoned Yellow River Delta, and carries sediment towards the shore (Chen et al. 2007; Liu et al. 2011). Therefore, the Xiyang Waterway changed the material source of the sampling point from the offshore radial sand ridges into the abandoned Yellow River delta. However, the grain size of sediments from the abandoned Yellow River delta was smaller than that of the offshore radial sand ridges (Wang et al. 2012a; Zhang and Chen 2012; Rao et al. 2015; Lv 2015). Thus, the grain size in core CDG was refined above 90 cm (1965).

Core XYK can be divided into three layers of 40 cm (2001) and 80 cm (1985): the upper layer (40–0 cm, 2001–2014), with a mean particle size of 4.72Φ , mean clay content of 7.54% and composed mainly of silt; the middle layer (80–40 cm, 1985–2001) with the mean particle size of 4.47Φ , mean clay content of 6.87% and composed mainly of silt; and the lower layer (131–80 cm, 1965–1985), with the mean particle size of 4.88Φ , mean clay content of 8.45% and composed mainly of silt. Sediments significantly coarsened in the 80–40 cm layer (1985–2001). With the basic leveling of the abandoned Yellow River undersea delta around 1980s (Wang 2006; Zhang et al. 2014), tidal power in the offshore radial sand ridges has been strengthened (Chen et al. 2009, 2010b, 2013) and resulted in coarsening of sediment, the coarsening sediment were transported directly to the coastal tidal flat (Gao and Zhu 1988; Xing et al. 2012; Chen et al. 2015). The sediments became significantly fine in the layer above 40 cm (2001–2014), which was due to reclamation activities near core XYK around 2001. Reclamation activities can increase the elevation of the tidal flat (Zhu et al. 2014; Li et al. 2016b), and the surficial sediment tend to become finer on the higher tidal flat (Wang et al. 2012b; Zhu et al. 2014; Zhou et al. 2015).

Accumulation of heavy metals

The concentrations of heavy metals in the three cores are summarised in Table 1. The vertical distribution of heavy

metals (Cu, Zn, Cr, Ni, Pb and As) is shown in Fig. 2. Heavy metals in core XYG were generally stable in the 145–48 cm (1900–1975) layer, whereas the contents of Cu, Zn, Ni, Pb and As decreased in the 48–0 cm (1975–2014) layer. Cr content slightly decreased in the 48–25 cm (1975–1992) layer and increased above 25 cm (1992). The vertical distribution of heavy metals in core XYG changed in 1975, which may be related to the coarsening of sediment around the 1980s.

The vertical distribution of heavy metals in core CDG was stable and showed a slight increase in the 143–91 cm (1935–1966) layer, which suddenly changed to a decreasing trend with significant fluctuations at 91 cm depth especially for Cu, Zn, Cr and Pb (Fig. 2). According to the previous analysis of sedimentary features in core CDG, the source of sediments changed at the 90 cm depth. The change of sediment source may lead to heavy metal content recombination.

The vertical distribution of Cr, Ni, and Zn in core XYK was basically stable except for bottom disturbance with Cu basically stable below 81 cm (1986), showing an obvious increasing trend above 81 cm (1986). As was stable below 81 cm (1986) and showed a slight increase above 81 cm (1986) (Fig. 2). The contents of heavy metals in core XYK were basically stable except for Cu and As, which may be related to the trend of sediment grain size coarsening initially and then recovering.

Source analysis of heavy metals

As multivariate statistical techniques, PCA and CA were extensively used to analyse the sources of heavy metal (Nguyen et al. 2016; Harikrishnan et al. 2017). Therefore, we applied PCA and CA to identify the sources of heavy metals in this study using the SPSS statistics software version 21.0. The obtained results are presented in Tables 2 and 3.

The cumulative contribution of the first two components in core XYG reached 78.29%, which reflected most of the primary data. The contribution of F1 (the first principal component) was 53.47%, which was higher than those of the

others. Since Pb, Ni, Cu, Zn and As had the highest positive loads on F1, it was supposed that F1 played a major role as the source of these elements into the sediments. Correlation analysis showed that these elements were significantly related to Al and had similarities in the homology and the migration path and process. Al is considered to be one of the most well conserved elements as it is extremely resistant to weathering and erosion (Wei et al. 2008). If there is a strong positive correlation between certain elements and Al, it is determined that these elements are mainly controlled by natural sources, otherwise they may be anthropogenic sources (Summers et al. 1996). Therefore, F1 mainly reflected the impacts of natural sources on the contents of heavy metals. The tidal flat sediments of XYG were sourced from the abandoned Yellow River delta (Zhang 1990; You et al. 1998; Zhang et al. 2012), and F1 revealed the effects of the sedimentary parent materials of the abandoned Yellow River delta on the contents of heavy metals. The contribution factor of the second principal component (F2) was 24.82% and mainly characterised by the high positive load of Cr. Based on the correlation analysis, the contribution factor was related to the low content of Al. As such, F2 represented the impacts of anthropogenic activities on the contents of heavy metals. Cr derived mainly from the electroplating and tanning industries (Ouki and Neufeld 1997; Chang and Kim 2007; Rumpa et al. 2011). According to environmental survey data near core XYG, the core is located in the state-level nature wetland reserves (Fig. 1) in Yancheng, Jiangsu Province; these reserves are poorly developed and have few sources of industrial pollutants. The Sheyang River, north of core XYG, is a larger sewage river. Cr-containing wastewater was discharged by industries of electroplating and tanning in the Sheyang Economic Development Zone into the Sheyang River, which ran into the sea eastward. Cr-containing sediments in wastewater migrated southwards to the waters of XYG under the tidal power and enriched the tidal flat sediments. This assumption was consistent with the severe pollution of Cr in the Sheyang River estuary (Gao 2011; Wu et al. 2017). Therefore, F2 represented the effects of

Table 2 Principal component of analysis of heavy metals of the sediments in three cores

Variables	XYG		CDG		XYK	
	F1	F2	F1	F2	F1	F2
Pb	0.889	0.367	-0.369	0.861	0.819	-0.137
Ni	0.861	-0.253	0.966	0.055	0.893	-0.029
Cu	0.839	-0.397	0.865	0.248	0.028	0.994
Zn	0.768	0.170	0.253	0.849	0.922	0.120
Cr	0.112	0.848	0.616	0.422	0.508	-0.274
As	0.500	-0.678	0.914	-0.166	0.155	0.839
Characteristic root	3.743	1.733	3.562	1.887	2.604	1.575
Contribution factor	53.47%	24.82%	50.88%	26.96%	43.40%	26.25%
Cumulative contribution factor	53.47%	78.29%	50.88%	77.84%	43.40%	69.65%

Table 3 Pearson's correlations matrix of the heavy metals in three cores

XYG	Cu	Zn	Pb	Al	Cr	Ni	As
Cu	1						
Zn	0.538**	1					
Pb	0.590**	0.657**	1				
Al	0.474**	0.586**	0.929**	1			
Cr	-0.216	0.138	0.355**	0.339**	1		
Ni	0.814**	0.574**	0.635**	0.504**	-0.045	1	
As	0.604**	0.176	0.198	0.540**	-0.343**	0.155	1
CDG	Cu	Zn	Pb	Al	Cr	Ni	As
Cu	1						
Zn	0.509**	1					
Pb	-0.037	0.616**	1				
Al	0.541**	0.306**	0.085	1			
Cr	0.345**	0.476**	0.219	0.574**	1		
Ni	0.821**	0.283*	-0.312**	0.690**	0.447**	1	
As	0.796**	0.173	-0.420**	0.449**	0.180	0.829**	1
XYK	Cu	Zn	Pb	Al	Cr	Ni	As
Cu	1						
Zn	0.113	1					
Pb	-0.062	0.630**	1				
Al	-0.151	0.628**	0.735**	1			
Cr	-0.291*	0.054	0.088	0.416**	1		
Ni	-0.017	0.780**	0.562**	0.595**	0.356**	1	
As	0.441**	0.117	0.013	-0.114	-0.136	0.208	1

*Significantly correlated at the level of 0.05 (bilateral)

**Significantly correlated at the level of 0.01 (bilateral)

industrial sources on the contents of heavy metals. However, the influence of anthropogenic sources on heavy metals did not occur all the time. The vertical distribution of Cr showed that Cr content varied around the background value below 25 cm (1992) and exceeded the background value above 25 cm (Fig. 2). The enrichment factor also acted as a useful tool in distinguishing the sources of heavy metal (Zhang and Liu 2002; Han et al. 2006). The enrichment factor of Cr increased significantly after 1992, exceeding 1.0 and reaching a maximum of 5.3 (Fig. 3), and the pollution was light to significant. Therefore, the impact of human activities on heavy metals has been evident since the 1990s in core XYG.

The contribution of F1 in core CDG was 50.88%, playing a major role in the source of heavy metals particularly. Cu, Ni, Cr and As had the highest positive loads on F1. Based on the correlation analysis, these elements were significantly related to Al. Therefore, F1 represented the effects of natural source and was subject to the sedimentary parent materials of the abandoned Yellow River delta. The contribution of F2 was 26.96% and was positively related to Pb and Zn. The load values suggested that F2 dominated the sources of Pb and Zn, which were rarely or poorly related to Al.

F2 represented the impacts of anthropogenic sources. Core CDG is near the estuary of Chuandong River, which receives urban domestic sewage from both sides. China's water supply and drainage pipes were galvanised water pipes and PVC pipes (containing lead salt stabiliser) (Li et al. 2016a), which led to an increase in Pb and Zn content. The core was also surrounded by Shanghai Chuandong Farm, Dafeng Farm and Fengqing Farm. According to the studies of Nicholson et al. (1999) and Bolan et al. (2004), Zn was often used as an additive to animal husbandry feed, but it could not be completely absorbed by livestock and most of it was excreted with faeces. Ren et al. (2005) proved that Zn content in livestock faeces reached 1763 mg/kg. As an organic fertiliser, the application of livestock manure can increase Zn content in the environment. Many studies had also confirmed that coal combustion was an important source of Pb (Lv et al. 2015; Zhu et al. 2017). The industries in Dafeng Economic Development Zone on the north side of core CDG had settled in many thermal power plants and metallurgical industries, using coal as the main fuel. By collecting survey data of land-based sewage outfall in Jiangsu Province from 2006 to 2010, the Pb content of Wanggang River in Dafeng

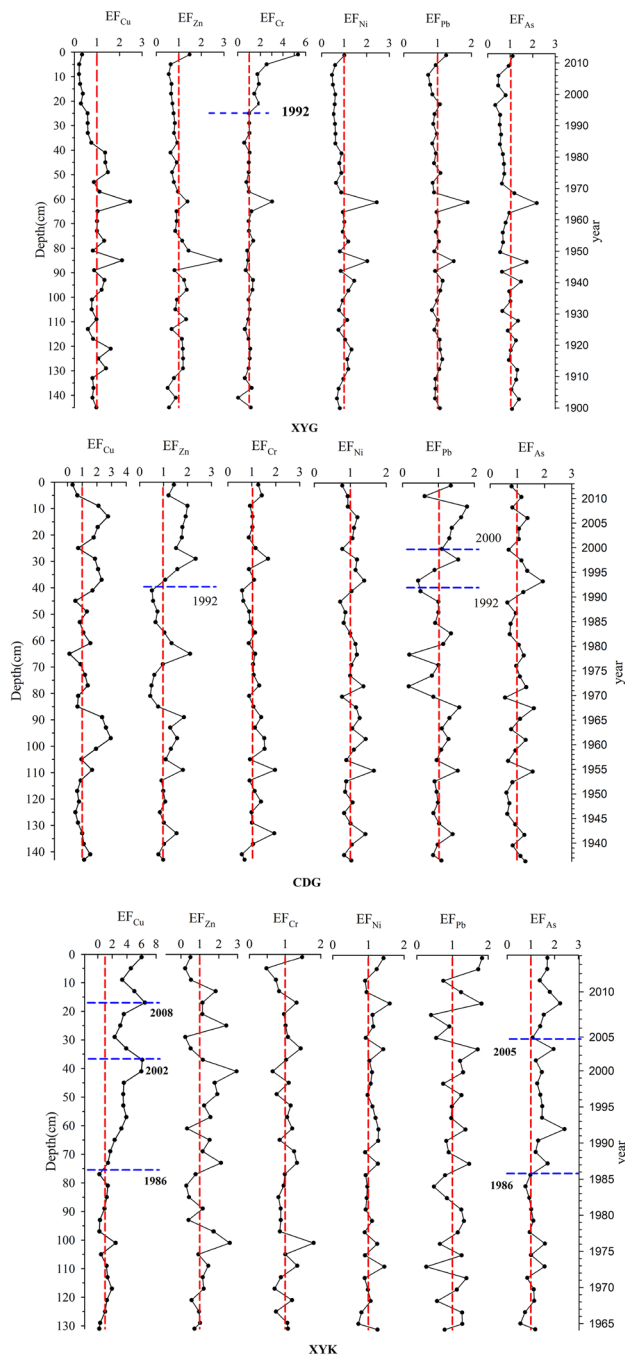


Fig. 3 The vertical distribution EF of heavy metals in three cores

Port Economic Zone ranged from 14.7 to 89.2 $\mu\text{g/g}$ (averaged 54.96 $\mu\text{g/g}$), which was higher than other estuaries. The analysis of heavy metal content in Wanggang coastal zone by Yu et al. (2007) also showed that Pb pollution was serious. Therefore, F2 may further represent urban, agricultural, and industrial sources. Pb and Zn content exceed the background value above 40 cm (1992) (Fig. 2) and the enrichment factors of Pb and Zn increased after 1992, reaching 1–2 (Fig. 3), the pollution being light. The impact of

human activities on heavy metals has been evident since the 1990s in core CDG.

The contribution of F1 in core XYK was 43.40%, playing a major role in the source of heavy metals. Zn, Ni, Pb and Cr had the highest positive loads on F1 and were significantly related to A1; hence, F1 represented natural sources and was attributed to the sedimentary parent materials of the offshore radial sand ridges. The contribution factor of F2 was 26.25%. Cu and As had high positive loads on F2 and were not related to A1. Therefore, F2 was defined as an anthropogenic source. Studies have shown that the continuous use of fertilisers and pesticides may cause the accumulation of Cu and As (Chen et al. 1999; Sutherland 2000; Barringer et al. 2007). In addition, the use of concrete releases of Cu (Sutherland 2000), and the chemical industry (e.g., pesticides plants) may emit As-containing pollutants (Barringer et al. 2007). Rudong County, a large traditional agricultural county, experiences more extensive use of fertilisers and pesticides. The amount of fertiliser used in Rudong County was 35,600 tons in 1985 and 41,200 tons in 1987 (Summary of Rural Economic Statistics by County in China 1989), ranking first among the coastal counties in Jiangsu Province. Rudong County also ranked first for pesticide application intensity in 1996 in coastal counties in Jiangsu Province (Marine Pollution Baseline Survey Report of Jiangsu Province 2001). Core XYK is closer to the Bencha River estuary and Rudong Chemical Park, where the river brings in agricultural and industrial pollutants and has certain influences on the sources of heavy metals. Therefore, F2 can further represent agricultural and industrial sources. The contents of Cu and As were higher than background value above 81 cm (Fig. 2). The enrichment factor of Cu was within 2–6 (Fig. 3), which belonged to moderate and significant pollution. The enrichment factor of As was within 1–2, which belonged to light pollution. Therefore, the impact of human activities on heavy metals has been evident since the 1980s–1990s in core XYK.

Evaluation of the pollution of heavy metals

The results of the enrichment factors of heavy metals in three cores are shown in Table 1 and Fig. 3. Al is considered to be one of the most well conserved elements as it is extremely resistant to weathering and erosion (Wei et al. 2008). The average enrichment factors of Al in core XYG, CDG and XYK were 1.09, 1.0, and 0.99, close to the background value, indicating that Al can be used as a reference element for enrichment factor in this study.

The enrichment factors of Pb, Ni, Cu, Zn, and As were relatively low and fluctuate around 1.0. The value of EF_{Cr} below 25 cm (1992) basically maintained about 1.0 and increased above 25 cm (1992). After 2006, the value reached 4–5.3, and the pollution was moderate to significant. The

industrial development of the Sheyang County in the 1990s was related to Cr. The Sheyang Economic Development Zone, constructed in the 1990s, discharges Cr-containing wastewater from factories of electroplating, tanning, etc., into the Sheyang River, which runs into the sea migrating sediments south to the tidal flat of XYG. Cr was a toxic heavy metal that was harmful to humans and should be considered as the priority control object of environmental protection in the tidal flat of XYG. Electroplating was a phase-out industry. Scholars proposed that the Sheyang Economic Development Zone should adjust the industrial structure to reduce the Cr-containing pollutants.

The enrichment factors of Ni, Cr and As in core CDG were low and fluctuated around 1.0. EF_{Cu} ranged from 0.2 to 2, which was non-pollution or light pollution. EF_{Pb} and EF_{Zn} fluctuated around 1.0 from the bottom up to 40 cm (1992) and basically remained within 1–2 above 40 cm (1992), which belonged to light pollution. Since the 1990s, the enrichment factors of Pb and Zn have shown an increasing trend and were related to urbanisation, husbandry and industrial development in Dafeng. From 1992 to 2011, China's urbanisation rate has increased from 27.63 to 51% (Bureau of Statistics of China 1992–2011). With an increase in food required to meet the population growth, China implemented a policy called "Vegetable Basket Project" and animal husbandry entered a period of rapid development in the 1990s. Dafeng is a major county for animal husbandry and has a large population in China, its development will also adapt to China's economic development. Urbanisation and agricultural development increase the content of Pb and Zn. The Dafeng Port Economic Development Zone was established at the beginning of the twenty-first century. Thermal power plants and metallurgical industries in Dafeng County mostly used coal as fuel, resulting in the pollution of Pb. Pb and Zn should be considered as the priority control elements of environmental protection in the tidal flat of CDG. Wastewater and waste gas treatment capacity must be improved to reduce the emissions of heavy metals.

The enrichment factors of Zn, Cr, Ni, and Pb in core XYK were relatively low and fluctuated around 1.0. EF_{Cu} and EF_{As} above 75 cm (1986) increased significantly; EF_{Cu} was within 2–6, which fell in the range of moderate and significant pollution, and EF_{As} was within 1–2, which belonged to light pollution. The development of agriculture and industry in Rudong County had a good response to Cu and As. In the late 1980s, to meet the demand of population growth for food, China's agriculture began to use fertilisers and pesticides in large quantities. The amount of fertiliser used nationwide was 8.84 million tons in 1978, increasing to 25.903 million tons in 1990 (China Statistical Yearbook 1999). The amount of pesticide used in China was 117,000 tons in 1983, increasing to 268,000 tons in 1995 (Ge et al. 1997). The heavy use of chemical fertilisers and pesticides

will increase the content of Cu and As in the environment. Moreover, local reclamation activities were significantly related to the enrichment factor of Cu. The value of Cu was high, indicating highly intensive reclamation activities in 2002 (at the 38 cm depth) and 2008 (at the 18 cm depth). Rudong Chemical Industrial Park, constructed in 2004, was related to the pollution of As. The enrichment factor of As above 30 cm (2005) increased obviously. Cu and As should be the priority control elements of environmental protection in the tidal flat of XYK. Ecological agriculture must be developed to reduce heavy metal pollution.

Conclusions

1. Sediments in the study area were mainly composed of silt. Sediments in core XYG began coarser in 1980, whereas those in CDG were suddenly fined in the 1960s. The grain size in core XYK coarsened in 1985 and refined in 2001. The depositional environment of the investigated region was constantly adjusted under the tidal power, causing changes in the material source and the recombination of sediments.
2. The vertical distribution in the contents of heavy metals was consistent with the sedimentary features. The contents of heavy metals in XYG were generally stable before decreasing slightly after the 1980s, whereas those in CDG suddenly decreased in the 1960s. The contents of heavy metals in core XYK were basically stable. The vertical distribution in the contents of heavy metals was complicated because of changes in the material source, swinging tidal creek and anthropogenic activities. However, for more accurate evaluation, continuous research is necessary.
3. Natural sources dominate the contents of heavy metals in the coastal tidal flats of the NJRSR. The sedimentary parent materials of the abandoned Yellow River delta controlled sediments in XYG and CDG, and those in XYK were subject to the sedimentary parent materials of the offshore radial sand ridges. However, individual elements were affected by anthropogenic source. The content of Cr in core XYG had significantly contributed to anthropogenic activities, which may be industrial processing since 1992. The contents of Pb and Zn in core CDG have been mainly contributed to anthropogenic sources since 1992 and the anthropogenic source can be subdivided into agricultural, urban and industrial sources. The contents of Cu and As in core XYK have been mainly anthropogenic activities since 1986, whereas those in XYK were defined as agricultural and industrial sources.
4. The environment of sediments in the study area was generally good, but the pollution of individual elements

existed after coastal industrialisation. Cr had become moderately and significantly polluting in XYG since the 1990s. Cr was a major contributor to the environmental pollution of tidal flats in XYG. Pb and Zn have become lightly polluting in CDG since the 1990s and were the major contributors to the environmental pollution of the tidal flats in CDG. In the middle and late 1980s, Cu was moderately and significantly polluting and As was lightly polluting in XYK. Cu and As were the main elements that contribute to the environmental pollution of tidal flat sediments in XYK.

Acknowledgements This work was financially supported by the National Natural Science Foundation of China (No. 41373112).

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