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Linkages between the spatial toxicity of sediments and sediment dynamics in the Yangtze River Estuary and neighboring East China Sea☆

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ABSTRACT

Anthropogenic activities are driving an increase in sediment contamination in coastal areas. This poses significant challenges for the management of estuarine ecosystems and their adjacent seas worldwide. However, few studies have been conducted on how dynamic mechanisms affect the sediment toxicity in the estuarine environment. This study was designed to investigate the linkages between sediment toxicity and hydrodynamics in the Yangtze River Estuary (YRE) area. High sediment toxicity was found in the Yangtze River mouth (Region I), the depocenter of the Yangtze River Delta (Region II), and the southeastern area of the adjacent sea (Region III), while low sediment toxicity was found in the northeastern offshore region (Region IV). A spatial comparison analysis and regression model indicated that the distributed pattern of sediment toxicity was likely related to hydrodynamics and circumfluence in the East China Sea (ECS) shelf. Specifically, high sediment toxicity in Region I may be affected by the Yangtze River Pump (YRP) and the low hydrodynamics there, and high toxicity in Region II can be influenced by the low sediment dynamics and fine sediment in the depocenter. The high sediment toxicity in Region III might be related to the combination of the YRP and Taiwan Warm Current, while the low toxicity in Region IV may be influenced by the local coarse-grained relict sand with strong sediment dynamics there. The present research results further suggest that it is necessary to link hydrodynamics and the spatial behavior of sediment and sediment-derived pollutants when assessing the pollution status of estuarine environments, especially for those mega-estuaries and their neighboring ocean environments with complex waves, tides and ocean currents.

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1. Introduction

Estuaries are important interfaces between continents and oceans where material fluxes have global impacts on marine biogeochemical cycles (Bianchi and Allison, 2009). As the transition zone between a river and an ocean, estuarine areas host two-thirds of the world's large cities, with intense human occupation. In addition, estuaries also shelter and nurse a diversity of species that benefit from their high primary productivity (Breukelaar, 2015; Machado et al., 2016). However, estuaries are also characterized by complicated physical processes and hydrodynamics, which could determine the dispersal pathways and distribution patterns of materials in aquatic environments (Wang et al., 2015; Yang et al., 2017).

Estuaries are also exposed to and polluted by significant anthropogenic inputs from upstream sources, metropolitan areas and industries located on or near estuaries (Duan et al., 2013; Wiederhold et al., 2014). The contaminated sediments can act as a secondary contamination source and be re-suspended in the water column when they are disturbed, moved and relocated (Droppo et al., 2011; Kalnejais et al., 2010; Roberts, 2012). Therefore, sediments in estuarine environments and adjacent seas could be net

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sinks and sources for natural and anthropogenic contaminants and could have a range of toxicological effects on benthic fauna and associated species (Roberts, 2012; Kalnejais et al., 2007).

Estuarine sediment is a major pollutant enrichment medium, with valuable geologic and environmental information records, and can serve as important evidence in the assessment of ecological environmental quality (Dai et al., 2016; Bi et al., 2017). Studies have been widely conducted to assess the pollution status in estuarine and coastal areas based on sediments. The ecological risk assessments of polycyclic aromatic hydrocarbons (PAHs) in estuarine sediments was investigated in the Tiber River Estuary (Italy) and the Brisbane River Estuary (Australia) (Montuori et al., 2016; Duodu et al., 2017). Heavy metal pollution in sediments was studied in the Loire Estuary (France) and the Vellar-Coleroon Estuary (India) (Coynel et al., 2016; Sundaramanickam et al., 2016). Since there are a wide range of pollutant species in the sediment, it hard to assess the comprehensive pollution in estuarine areas, especially for those estuaries with complex hydrodynamics and significant human impacts.

The Yangtze River Estuary is the largest estuary in China. Affected by anthropological activities from the populated regions in upstream regions and metropolitan Shanghai in the estuarine area, the YRE is exposed to various types and huge amounts of pollutants (Dai and Liu, 2013; Chen et al., 2017a). Extensive research has been conducted to detect the species of pollutants in the aquatic ecosystem in the YRE and the ECS coastal areas and found that toxic substances, including heavy metals (i.e., Pb, Zn, Cu, Cr and Ni), organochlorine pesticides and persistent organic pollutants are significant hazards for aquatic organisms and can pose a threat to the YRE ecosystem (Hu et al., 2011; Yu et al., 2016; Chen et al., 2014; Zhou et al., 2014). Moreover, the spatial distributions of pollutants in the YRE have been widely investigated, and the results indicated that pollutants in the sediment were strongly influenced by terrigenous input (Zhou et al., 2014; Wang et al., 2017). In addition, previous studies also suggested that the impoundment of the Three Gorges Dam (TGD) had an influence on the sedimentation of the pollutants in YRE (Chen et al., 2017b; Li et al., 2017). However, few studies have determined the role of sediment dynamics in the expansion, deposition, resuspension, and redistribution of pollutants in the YRE due to its complicated hydrological system. Therefore, the aims of this study are (1) to explore the spatial distribution of sediment toxicity in the YRE and (2) to investigate relationships between sediment dynamics and the spatial distribution of sediment contaminants.

2. Study area and methods

2.1. Study area and setting

The Yangtze River originates on the Tibetan Plateau and drains into the East China Sea (ECS) after flowing eastward for more than 6300 km (Zhu et al., 2011). The ECS continental shelf is the largest marginal sea in the western Pacific Ocean, with a vast continental area of 0.5×10^{12} m^2 (Yu et al., 2016) that receives large quantities of particulate and dissolved terrestrial materials from the YRE (Liu et al., 2007; Zhu et al., 2011). The circulation system of the YRE and the adjacent area is complicated by the Yangtze River Plume (YRP), the Jiangsu Coastal Current (JCC), the Zhejiang–Fujian Coastal Current (ZFCC), and the Taiwan Warm Current (TWWC), all of which occur along the continental shelf near the estuarine area (Fig. 1) (Liu et al., 2007). The Kuroshio Current (KC) flows northward along the outer break of the shelf, and as the outer boundary of high production, it can act as a barrier for the deposition of fine-grained terrigenous sediments (Lie and Cho, 2002; Lin et al., 2002). Furthermore, mud depocenter and upwelling areas (the circled area in Fig. 1) in the YRE and adjacent shelf area of the ECS (Liu et al., 2007; Zhu et al., 2011) further complicate the sedimentary environments and hydrodynamic conditions.

2.2. Sample collection

Sediment samples (0–3 cm of the surface layer) were taken in March 2013 from the YRE and the adjacent continental shelf area (28°58′–32°15′ N, 122°00′–124°45′ E; Fig. 1). All of the sediment samples were collected using a box sediment sampler with a tight seal. Once on the deck, the box sediment sample was sub-sampled to take the surface layer only (0–3 cm) using a stainless-steel spoon. Then, the surface sediment sample (approximately 150 g) was packaged in labeled polyethylene bags. Fifty-nine sediment samples were collected, frozen and stored at −20 °C for the analysis of grain size and developmental toxicity.

2.3. Grain size analysis and sediment classification

Each sediment sample was treated with a 30% hydrogen peroxide solution for 24–48 h to eliminate organic materials, and then a grain-size analysis for each sediment sample was conducted in the laboratory using an LS-100 particle size analyzer. The sediments were classified according to the method established by Folk et al. (1970), and sand–silt–clay ratios of sediments were plotted on a ternary diagram in order to diagnose the sediment type in the study area. In addition, the sediment components for all samples were calculated according to the Udden-Wentworth grain-size classification scheme (Wentworth, 1922). In this study, clay (>8 φ, <3.9 μm), silt (4–8 φ, 3.9–62.5 μm), sand (<4 φ, >62.5 μm) and the median diameter were selected to examine the influence of grain size on sediment toxicity.

2.4. Assessment of sediment developmental toxicity

A wide variety of fish and amphibian species have been used as model organisms in sediment toxicity research (Munoz et al., 2011; Zhang et al., 2014; Guo et al., 2016). The embryo Teratogenesis Assay–*Xenopus* (FETAX) is a rapid, cost-effective, and powerful bioassay for evaluating developmental toxicants (Dumont et al., 1983; Bacchetta et al., 2008). *Xenopus tropicalis* can lay a large number of eggs (approximately 5000–6000) at a time, and the hatching time is quite short (24 h). Therefore, the developmental analysis using this method is quite easy and rapid (Zhang et al., 2014). In this study, the FETAX with some modification was used to evaluate the developmental toxicity of sediments, and the exposure experiments were conducted using our previous methods (in supplemental materials) (Gao et al., 2015).

2.5. Regional Ocean Model System (ROMS)

ROMS is a three-dimensional primitive equation ocean model that includes several submodels, such as wave, sea ice and sediment models and is widely used among ocean researchers (Haidvogel et al., 2000). Previous studies have shown that the high-resolution ROMS showed a remarkable prediction ability for Isabel-generated surface stress, storm surges and currents in the Chesapeake Bay (Li et al., 2006). Klinck et al. (2014) used ROMS to investigate the delivery of ocean heat to the base of the ice shelves in the entire Southern Ocean. In addition, ROMS was also used to investigate the ecosystem response to phosphorus limitation in the Pearl River plume (Gan et al., 2014).

In this study, the ROMS model was used to calculate the mean bottom shear stress according to the method described by Warner et al. (2008), since the bottom shear stress is considered the main

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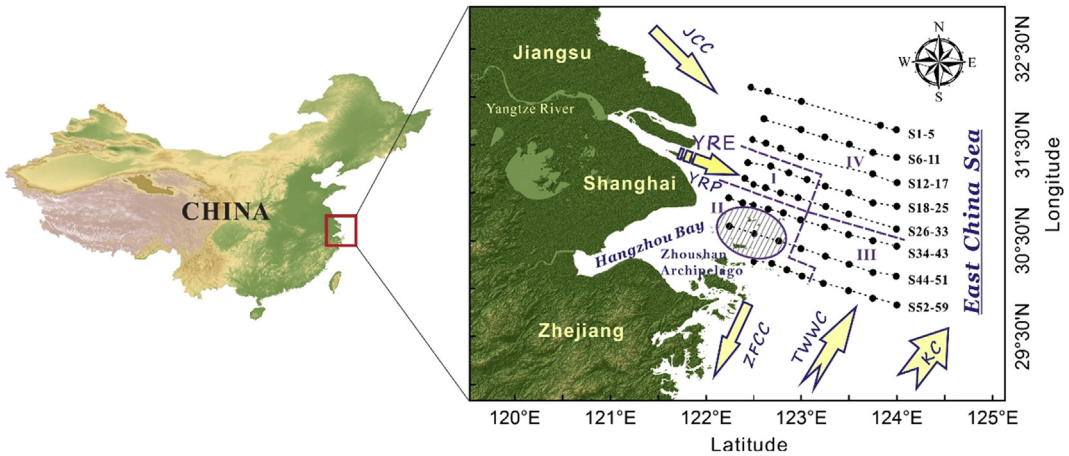

Fig. 1. Locations of sampling sites and regional circulation system in the Yangtze River Estuary and the adjacent continental shelf. The circled area is the sediment depocenter of the Yangtze River Delta. (YRP: the Yangtze River Plume. JCC: the Jiangsu Coastal Current; ZFCC: the Zhejiang–Fujian Coastal Current; TWWC: Taiwan Warm Current; KC: the Kuroshio Current).

hydrodynamic parameter that has the largest impact on sediment transport. In addition, the sediment module of ROMS was also used to model the current-induced sediment transport (both direction and flux) in the YRE and neighboring ECS. In this study, the model domain covers the YRE and its neighboring ECS area. The designed grid for the model is shown in Fig. S1, and the daily surface elevation, currents, and salinity from the HYCOM global 1/12° analysis model (see details at https://hycom.org/dataserver/glb-analysis) were applied as open boundary forcing. The model grid resolutions are approximately 800–1200 m (according to the longer side of the grid cell) in the YRE regions, and in the other sampling regions, model grid resolutions vary from 1200 m to 4500 m. In the model, the bottom shear stress was calculated as:

$$\frac{\rightarrow_{\tau_b} = \kappa^2 \rightarrow_u \left| \rightarrow_u \right|}{ln^2\left(\frac{z}{z_0}\right)}$$

where τ_b is the bottom shear stress, κ is the von Karman's constant, u is the bottom current velocity and z_0 is the bottom roughness height (Warner et al., 2008).

3. Results

3.1. Spatial characteristics of sediment composition

Sediment grain size varied greatly in the YRE and its adjacent sea (Fig. 2). The median diameter ranged from φ<2.80 to φ>7.54, with an average of φ = 4.47 (Table S1). The Folk's triangular classification of sediments revealed that surface sediments in the YRE area mainly consisted of muddy sand and mud (Fig. S2). Clay, silt and sand contents were 3–41%, 7–71% and 0.4–91%, with averages of 20%, 36% and 44%, respectively. Sediments samples were poorly sorted, and the sediments showed a band-type distribution, with fine-grained sediment around the mouth area and the depocenter (the circled area in Fig. 1) of the Yangtze Delta and coarse-grained sediment on the outer shelf (Fig. 2, Table S1).

3.2. Phenotypic characteristics of malformations in embryos induced by the extracts

Multiple malformations were observed in the embryos of the experimental groups treated with sediment extracts. The phenotypes of malformations included hypopigmentation, delayed development of the gut, abnormal proctodeum, narrow fin, abnormal eyes, edema of the heart, bent tail, bent notochord and enlarged truck (Fig. S3). The main phenotypes of malformation were dominated by hypopigmentation and delayed development of the gut, followed by abnormal proctodeum and narrow fin (Fig. S3 and Table S2). High percentages of delayed development of the gut and narrow fin were distributed along the coastal area (Fig. 3B and D), and the peaks of percentages of hypopigmentation and abnormal proctodeum were observed in the southeast of the study area (Fig. 3A and C).

3.3. Sediment toxicity

The toxicity of the sediment displayed spatial disparity. The mean percentage of survival was 80%, with several low-value regions outside of the delta and in the southeastern part of the YRE (Fig. 4B). The mean percentage of total malformation was 39%, with three distinct high value regions in the Yangtze River mouth (Region I), the depocenter of the Yangtze River Delta (Region II), and the southeastern part of the study area (Region III) (Fig. 4A). Distinct high malformations in Region I mainly consisted of hypopigmentation, delayed development of the gut and narrow fin; high malformations in Region II mainly consisted of delayed development of the gut and narrow fin; and high malformations in Region III mainly consisted of hypopigmentation, delayed development of the gut and abnormal proctodeum (Figs. 3 and 4). The northeastern region of the study area (Region IV) was characterized by high survival and low malformation, showing low sediment toxicity.

4. Discussion

4.1. Impacts of sediment source

Due to the low water solution, most organic contaminants and toxic metals are prone to combine with particles and tie their fate to sediment in aquatic systems (James et al., 2010; Tuikka et al., 2011). Therefore, a sufficient understanding of sediment sources and transportation can provide effective information for estimating the fate of sediment-related contaminants.

The frequency curves of sediment samples in the study area indicate that there are different sediment sources in the ECS shelf (Fig. 5). Fine sediments with unimodal curves along the inshore

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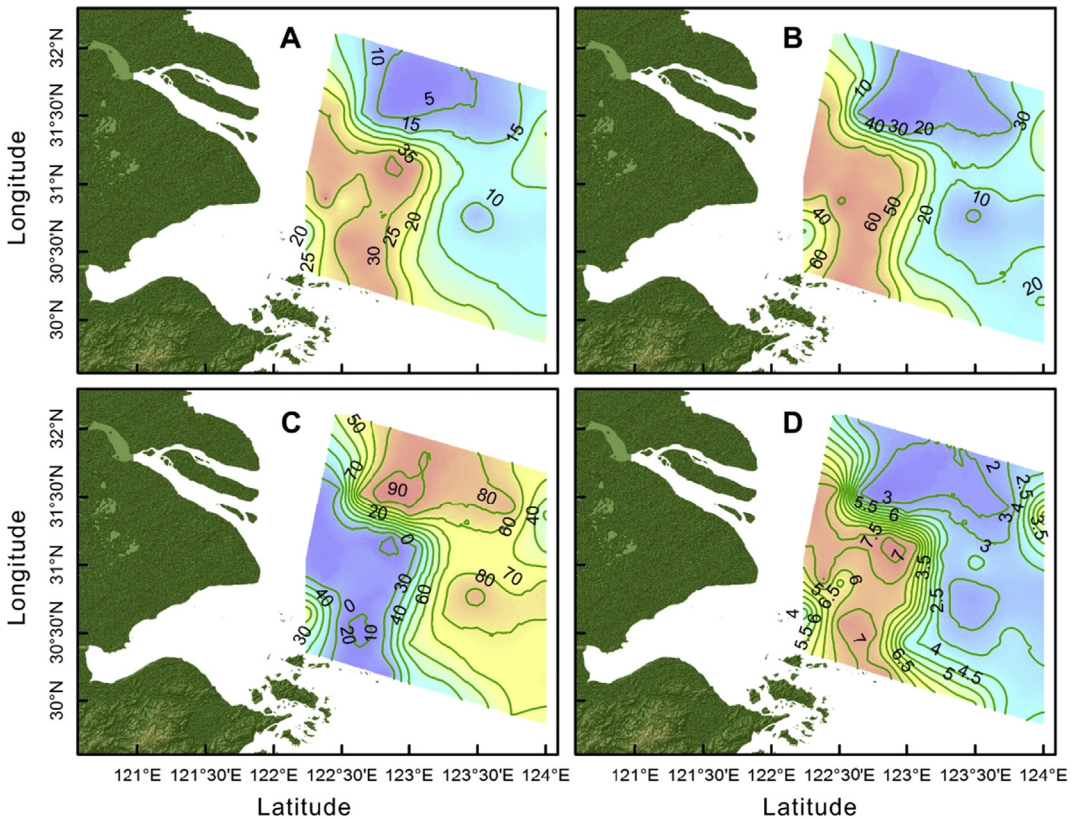

Fig. 2. Distributions of clay (a), silt (b), and sand percentages (c) (%) and the median diameter (d) (φ) of sediments in the Yangtze River Estuary and neighboring ECS.

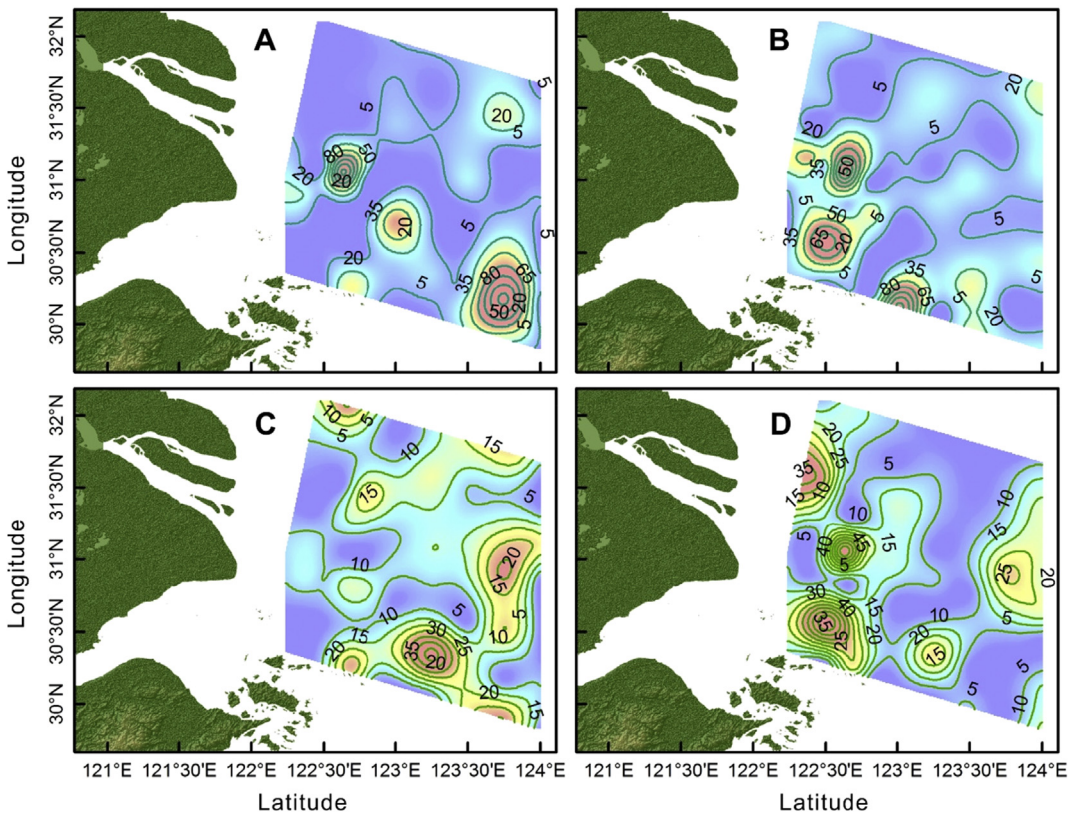

Fig. 3. Spatial characteristics of main phenotypic malformations in percentage. (A: hypopigmentation; B: delayed development of the gut; C: abnormal proctodeum; D: narrow fin).

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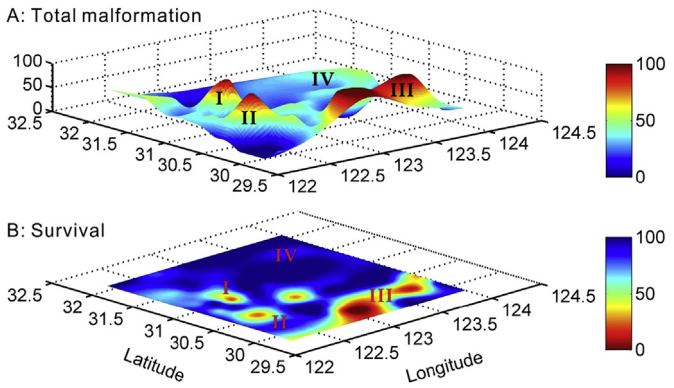

Fig. 4. Spatial distributions of sediment toxicity indicated by the indices of teratogenicity. A: total percentage of malformation; B: survival percentages.

area come from the Yangtze River (Fig. 5) (Zhou et al., 2014). As the largest river on the Eurasian continent, the Yangtze River is the main sediment source in the study area, delivering ca. 0.43 × 10^9 ton/yr (1950–2000) of suspended sediment to the YRE and ECS shelf (Dai et al., 2014, 2016). Along with the suspended particulate matter (SPM), huge amount of pollutants can also be delivered to the YRE and ECS coastal areas (Wang et al., 2017; Chen et al., 2017b). According to the result of the sediment transport model, the Yangtze River delivered suspended sediment that was mainly transported south and southeast towards along the nearshore area (Fig. 6). Thus, it is logical to deduce that sediment-bound pollutants from the mainland can also be carried by and delivered with the suspended sediment southward along the ECS inner shelf (Floehr et al., 2013; Yu et al., 2016; Chen et al., 2017b). In this study, low percentages of survival and high percentages of malformations occurred in the inshore areas (Region I, II), implying that high sediment toxicity could be caused by the pollutants with terrestrial inputs (Figs. 3 and 4).

Coarse sediments with unimodal curves in the northeast of the study region (Region IV) are relict sediments (Fig. 5). The relict sediments from the last glacial period are characterized with abundant biogenic carbonate and quartz sand, which are two primary diluting components for the contaminants from the mainland (Lin et al., 2002). Therefore, we suggest that the low sediment toxicity in Region IV was likely attributed to the dilution effect of the relict sand. In addition, the middle shelf is a transition zone, and

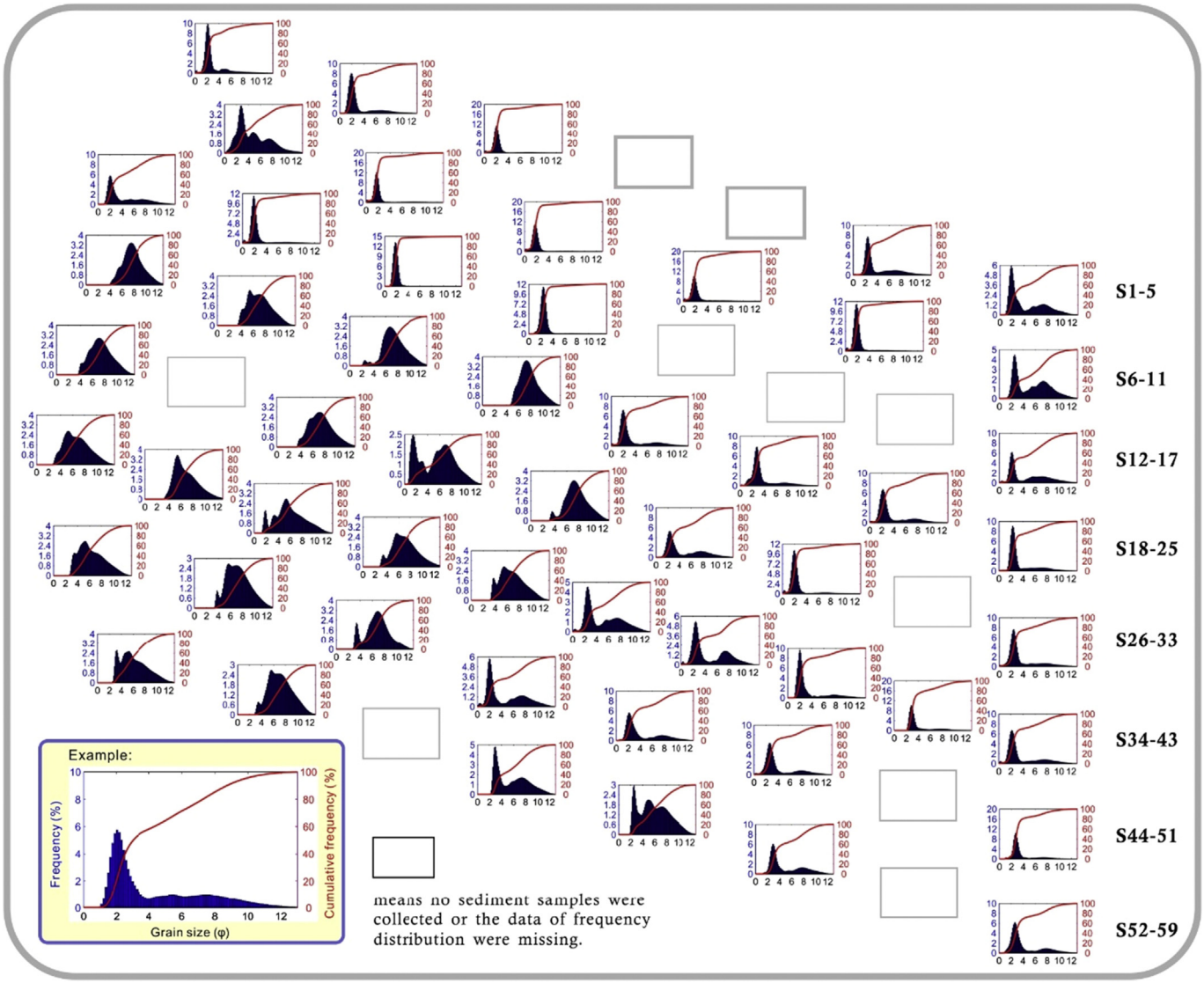

Fig. 5. Frequency distribution curves of sediment samples in the study area. The frequency curves in this figure correspond to the position of each sediment sample collected.

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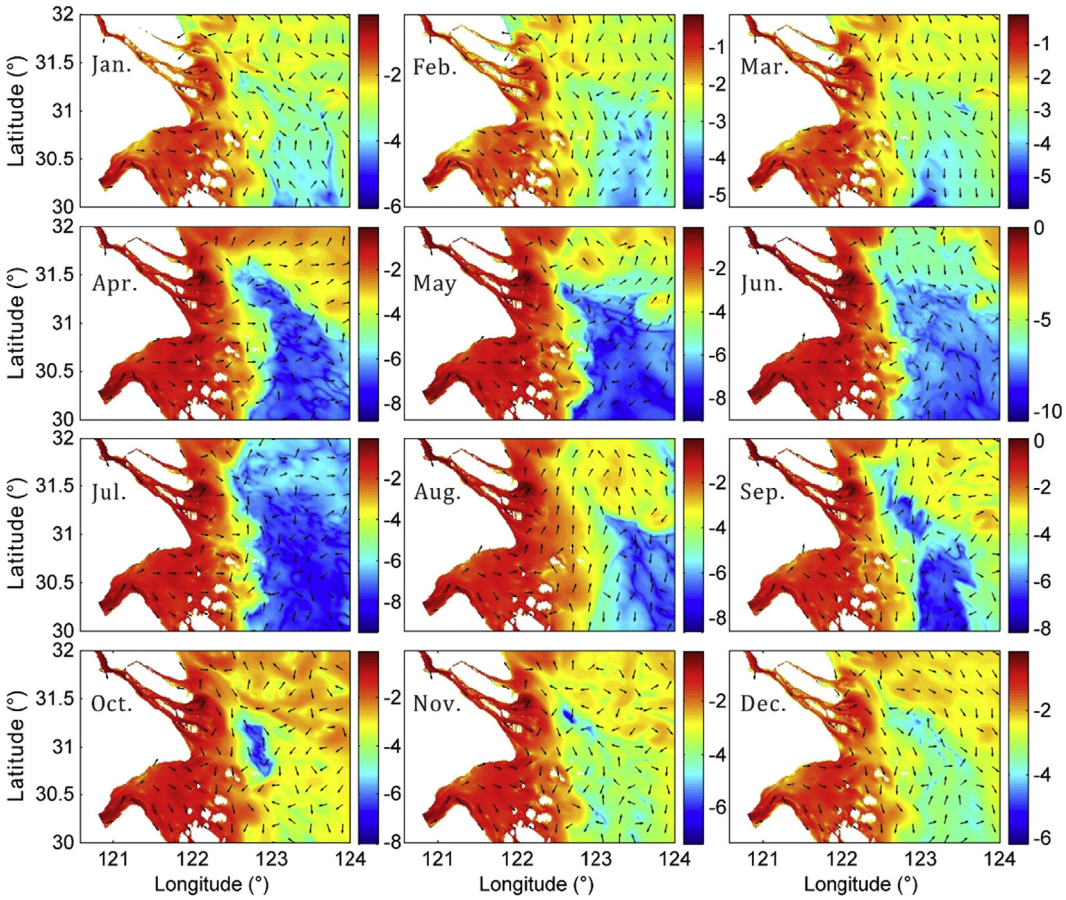

Fig. 6. Sediment direction (the arrow) and flux (the color bar shows log10 (sediment flux)) in the Yangtze River Estuary and neighboring East China Sea area (sediment flux was calculated using the formula: sediment flux = velocity × suspended sediment concentration). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the bimodal or trimodal frequency curves of sediment there result from the expansion or exchange of sediment from the Yangtze River and from the relict sand area, which can make the spatial pattern of sediment toxicity more variable and complicated (Fig. 5).

4.2. Impact of sediment grain size

The grain size is also a factor that plays an important role in the distribution of sediment-related contaminants in the YRE and neighboring sea. It is widely accepted that fine-grained sediment with high cohesion and adhesion has a high affinity to many organic and inorganic contaminants in the aquatic environment (Droppo et al., 2015; Righetti and Lucarelli, 2010).

Influenced by different sediment sources (the Yangtze River and the relict sand), the mean grain size of sediment samples from the YRE and ECS areas showed clustered trends (Fig. 7B). The content of fine-grained sediments (clay and silt) showed a decreasing trend from the inshore towards offshore areas, with higher contents of clay and silt in the coastal areas of the ECS and in the southeastern part of the study area (Region IV) (Fig. 2). The spatial pattern of sediment toxicity showed a very similar trend, with higher toxicity in the nearshore zone (Region I-III) and low toxicity in the outer shelf (Region IV) (Figs. 3 and 4). The spatial patterns of sediment grain size and the percentage of malformations indicate that sediment grain size could influence the spatial variations of sediment toxicity.

Furthermore, sediment toxicity in terms of total malformation was linearly correlated with the mean grain size of sediment ($P < 0.05$) and the content of clay ($P < 0.01$) (Fig. 7). This result means that with higher clay content and finer sediment, there is higher sediment toxicity in the study region, which is consistent with the spatial patterns of sediment grain size and sediment toxicity. These results indicate that sediment toxicity in the YRE and the ECS shelf was significantly influenced by the fine-grained sediment content.

4.3. Impact of hydrodynamics

Hydrodynamics play an important role in the transport, deposition and resuspension of sediments and pollutants in the aquatic environment (Chen et al., 2017b; Yang et al., 2017). The bottom shear stress is considered to be the main physical parameter that has the largest impact on sediment transport, since it controls erosion, resuspension and deposition in the water column (Soulsby and Clarke, 2005). Therefore, the mean bottom shear stress based on the ROMS model was selected as an indicator of hydrodynamics in the study area.

The spatial distribution of bottom shear stress is shown in Fig. 8A. Hydrodynamics were the strongest in the branches of Yangtze River Estuary and the Hangzhou Bay because of the strong interactions of rivers and tides, as well as the effect of the complex bathymetry there (Fig. 8A). In the study area, bottom shear stress showed the opposite spatial pattern to that of sediment toxicity. Specifically, bottom shear stress was low in Regions I-III and high in Region IV, while sediment toxicity was high in the Regions I-III and low in Region IV (Figs. 3, 4 and 8A). Furthermore, sediment toxicity

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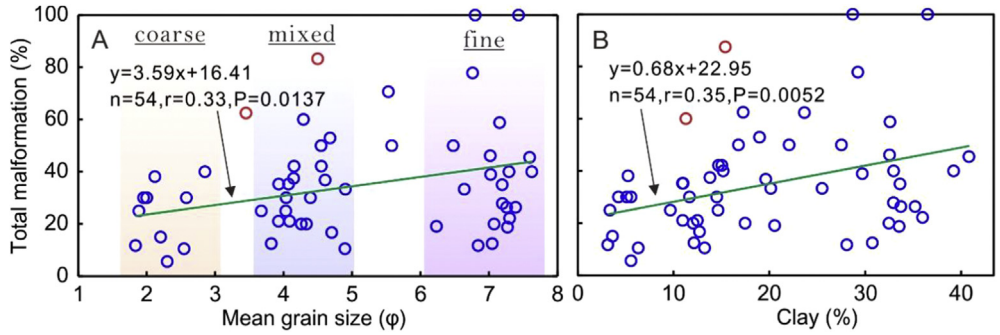

Fig. 7. Correlations of the total malformation with clay (%) and sediment mean grain size (φ). Samples marked in red and those with the percentage of survival <20% were considered outliers and not included when conducting the correlation analysis. On average, coarse samples from relict sand consist of <10% clay and >80% sand with unimodal frequency curves; fine samples from the Yangtze River are composed of 30% clay and <10% sand with unimodal curves; mixed samples, which have different sediment sources, contain 15% clay and 60% sand with bimodal frequency distributions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

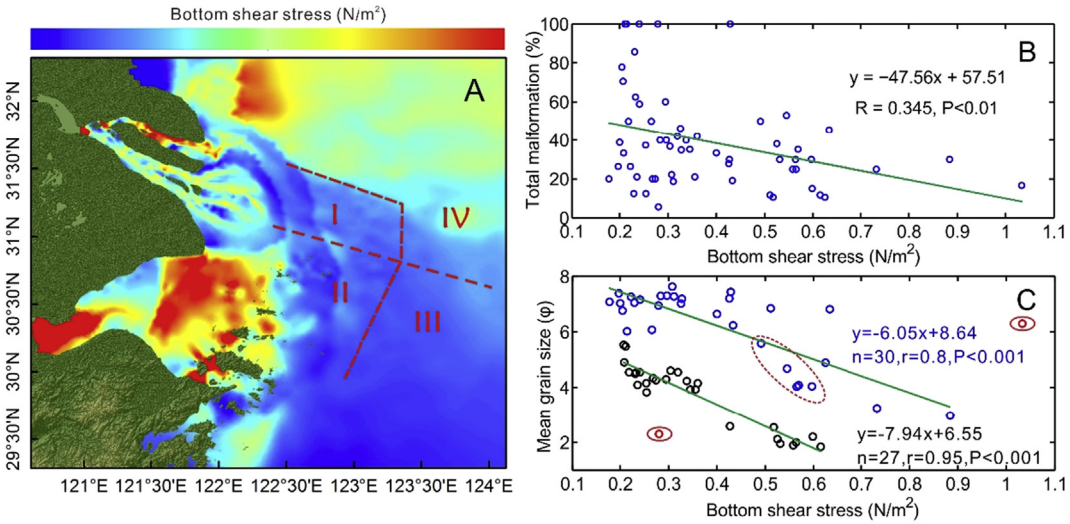

Fig. 8. Bottom shear stress and the sediment dynamic mechanism of sediment toxicity in the Yangtze River Estuary and the neighboring ECS. A. spatial distribution of bottom shear stress; B. correlation between bottom shear stress and percentage of total malformation; C. correlation between bottom shear stress and mean grain size of sediment. The black dots are samples with coarse grain sizes from the relict sand area, while the blue dots (except the ones in the dashed circle) are samples with fine grain sizes from the Yangtze River. Samples within the red circles were excluded when conducting the correlation analysis. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).

was linearly correlated with bottom shear stress at the significant level of $\alpha = 0.01$ (Fig. 8B). These observations imply that hydrodynamics can influence the spatial variation of sediment toxicity.

Hydrodynamics in the estuarine and coastal area can affect the behavior and fate of pollutants in the aquatic environment. Pollutants in the sediment can be released and resuspended into the water column when sediments are disturbed, moved and relocated (Kalnejais et al., 2010; Droppo et al., 2011; Roberts, 2012). Therefore, the high dynamics in Region IV may cause the release and resuspension of pollutants, resulting in low sediment toxicity, whereas the low bottom shear stress in Regions I-III could be beneficial for the deposition and enrichment of harmful substance and lead to high sediment toxicity (Figs. 3, 4 and 8A, 8B).

In addition, higher bottom shear stress could induce the resuspension of fine-grained particles, resulting in erosion and the coarsening of surface sediment, while lower hydrodynamics creates a deposition environment, and the surface sediment becomes finer (Zheng et al., 2011; Chen et al., 2017a, 2017b). This can be shown by the significant negative correlation between hydrodynamics (or bottom shear stress) and sediment grain size (Fig. 8C). Then, the redistribution of sediment grain size caused by erosion or deposition can have a further influence on sediment toxicity, as discussed in section 4.1.

4.4. Impact of circumfluence

In addition to the local hydrodynamics, the sediment toxicity in the coastal area is also influenced by circulation systems in the ECS shelf (Liu et al., 2007; Zhu et al., 2011). The YRP delivered materials towards the south and southeast (Fig. 6). The velocity of the YRP slows down in the river mouth and ca. 40% of the fine-grained sediments delivered by the Yangtze River is deposited in the estuarine area (Milliman et al., 1985; Dai et al., 2014). The low velocity and high content of fine sediments could facilitate the deposition and enrichment of sediment-associated contaminants around the estuarine region (Han et al., 2016), which can contribute to the high toxicity in Region I (Figs. 3 and 4). The rest of the sediments was carried by the YRP and the ZFCC southward, and then part of the sediment can be trapped on the inner shelf (Fig. 6) (Lin et al., 2002). Blocked by the Zhoushan Archipelago, sediments were deposited in the mouth of the Hangzhou Bay and formed the depocenter of the Yangtze Delta (Fig. 1) (Liu et al., 2007; Dai et al.,

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2014). Low dynamics and the fine contents of the depocenter may be responsible for the high sediment toxicity in Region II (Figs. 3 and 4).

Affected by the East Asia Monsoon, the TWWC is weaker in winter (from late September to early April) when a strong NE monsoon is dominant, and stronger in the summer (from April to September) when the southwest monsoon prevails (Liu et al., 2006; Zhu et al., 2011). The TWWC flows to the north throughout the year, which could slow down the spread of terrestrial sediments and pollutants into the ocean, especially in the summer monsoon season (Fig. 6) (Lie and Cho, 2002; Qiao et al., 2006; Wang et al., 2017). Consequently, large deposits of pollutants (i.e., PFAAs, PCBs) exist in the interface of the YRP and TWWC (Wang et al., 2017; Chen et al., 2017b). The 'barrier' effect of the TWWC and the low hydrodynamics can contribute to the high sediment toxicity in Region III (Figs. 3, 4 and 8A). In contrast, the intensive hydrodynamics and coarse sediment grain size in Region IV resulted in low sediment toxicity in Region IV. Taken together, the spatial distributions of sediment toxicity in the YRE and neighboring ECS were influenced by the sediment sources, grain size, local hydrodynamics and circumfluence in the YRE and ECS shelf.

5. Conclusions

Sediment in the estuarine and coastal areas is a good record that allows for a sound understanding of the pollution status in the aquatic environment. In this study, the spatial variations and influential factors of sediment toxicity were investigated based on surface sediment samples collected from the YRE and the neighboring ECS.

The results showed that sediment toxicity had a seaward decreasing trend, with three high toxicity regions in the nearshore area (Region I-III) and a low toxicity region in the offshore area (Region IV). As discussed, sediment sources, grain size, hydrodynamics and circumfluence in the YRE and ECS coastal area can have an influence on the spatial distribution of sediment toxicity. Fine-grained sediments showed higher toxicity, and fine sediments and sediment-bound contaminants from the Yangtze River were the main contamination sources in the coastal area. A significant correlation existed between hydrodynamics (or bottom shear stress) and sediment toxicity ($P < 0.01$). In addition, sediment toxicity was also influenced by the circulation system in the ECS shelf.

Hydrodynamics are difficult to assess in the field or laboratory, and here, we only used the bottom shear stress as an indicator of the dynamics in this study region. In fact, other physical process such as flocculation processes also impact sediment transport, settling, and contaminant bonding in the estuarine and coastal area. Future work should include more physical processes based on field data when developing and applying physical-based models to investigate the link between hydrodynamics and sediment toxicity.

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Competing financial interests

The authors declare no competing financial interests.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2017.10.023.

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