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# Phosphorus speciation and distribution in surface sediments of the Yellow Sea and East China Sea and potential impacts on ecosystem

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**Abstract**

For better understanding the phosphorus (P) cycle and its impacts on one of the most important fishing grounds and pressures on the marine ecosystem in the Yellow Sea (YS) and East China Sea (ECS), it is essential to distinguish the contents of different P speciation in sediments and have the knowledge of its distribution and bioavailability. In this study, the modified SEDEX procedure was employed to quantify the different forms of P in sediments. The contents of phosphorus fractions in surface sediments were 0.20–0.89 μmol/g for exchangeable-P (Exch-P), 0.37–2.86 μmol/g for Fe-bound P (Fe-P), 0.61–3.07 μmol/g for authigenic Ca-P (ACa-P), 6.39–13.73 μmol/g for detrital-P (DAP) and 0.54–10.06 μmol/g for organic P (OP). The distribution of Exch-P, Fe-P and OP seemed to be similar. The concentrations of Exch-P, Fe-P and OP were slightly higher in the Yellow Sea than that in the East China Sea, and low concentrations could be observed in the middle part of the ECS and southwest off Cheju Island. The distribution of ACa-P was different from those of Exch-P, Fe-P and OP. DAP was the major fraction of sedimentary P in the research region. The sum of Exch-P, Fe-P and OP may be thought to be potentially bioavailable P in the research region. The percentage of bioavailable P in TP ranged from 13% to 61%. Bioavailable P burial flux that appeared regional differences was affected by sedimentation rates, porosity and bioavailable P content, and the distribution of bioavailable P burial flux were almost the same as that of TP burial flux.

**Key words:** phosphorus speciation, burial flux, sediment, Yellow Sea, East China Sea

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## 1 Introduction

Phosphorus (P) is an essential nutrient in the oceans that can control primary production, influence ecosystem structure (Paytan and McLaughlin, 2007) and the global biogeochemical cycles (Broecker and Peng, 1982). The oceanic P level is governed by the input from fluvial and eolian sources and burial in sediments (Schenau et al., 2005). Excessive imports of P to the ocean can change the nutrients concentrations and their stoichiometric ratios, cause eutrophication (Ahlgren et al., 2006), bloom of toxic algae (Anderson et al., 2002; Kawasaki et al., 2010) and hypoxia in bottom waters (Diaz and Rosenberg, 1995), affect plankton composition and change the entire food web in a longer perspective (Humborg et al., 1997). In marginal seas, marine sediments are the major sink for P. Previous studies indicated the occurrence of significant diagenetic reorganization of P during burial in continental margin sediments (Ruttenberg and Berner, 1993; Berner and Rao, 1994; Slomp et al., 1996). Better understanding of P cycling in the oceans, especially in continental margin seas can provide an insight of the biogeochemistry in the ocean and the cycling of C, N, O and S on geological time scales (Van Cappellen and Ingall, 1996).

The Yellow Sea (YS) and the East China Sea (ECS) are the important marginal seas in the Northwest Pacific. The YS is a temperate and shallow semi-enclosed sea between China and the Korea Peninsula and its mean depth is 44 m. Previously, the YS ecosystem has been experienced so many changes in the past few decades, including the changes of nutrient concentrations and ratios (Gao and Li, 2009), increased eutrophication (She, 1999), changes in the composition of plankton communities (Lin et al., 2005), significant increase in the jellyfish blooms (Dong et al., 2010) and the recurrence of macroalgal green tides blooms (Sun et al., 2008). The spring blooms could play an important role in the structure of the YS ecosystem, affecting carbon fixation and other services (Tang et al., 2013). The ECS which links the western pacific by the Okinawa Trough and Ryukyu Islands at the southeast is connected to the YS at the north. The ECS has a continental shelf width of about 640 km and the mean depth is 72 m (Dong et al., 2011). The ECS may face the same responses of ecosystems to changes in physical variables and chemical elements as the YS in recent years (Chai et al., 2009; Wang et al., 2003). The YS and the ECS receive a huge amount of sediments discharged annually by the Changjiang River (Yangtze River) ($0.48\times10^9$ t/a; Milliman, 1991) and the erosion of the old Huanghe River (Yellow River) mouth ($0.5\times10^9$ t/a; Satio and Yang, 1994). Most of the

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suspended sediments transported by the Changjiang River is moved southward by the Jiangsu coastal current (Cao et al., 1989) and deposits in the Zhejiang and Fujian mud area, only a small portion is transported east and northeast (Sternberg et al., 1985). It is reported that the Changjiang River which is the world's fourth largest river discharged (calculated by P) about $(11.8\pm2.3)\times10^9$ mol/a and more than 95% of the riverine P is in the solid phase (Qu et al., 1993; Fang et al., 2007). The Kuroshio's incursion can also supply 0.38 kmol/s in summer and 0.92 kmol/s in winter (Zhang et al., 2007a).

In recent years, many research focused on the concentrations, distribution, biogeochemistry behavior and environmental significance of P in surface sediments or sediment cores in the YS and the ECS (Feng et al., 2001; Cao and Liu, 2010; Liu et al., 2004; Song et al., 2006; Wei and Wang, 2012; Zheng et al., 2003). For better understanding the P cycle and its impacts on one of the most important fishing grounds and pressure on the marine ecosystem in the YS and the ECS as a whole, it is essential to obtain a better insight into the behavior of P and distinguish the contents of different P speciation in sediments. The knowledge of distribution and bioavailability of P in sediments of the YS and the ECS will be significant for further study of P geochemistry. In this study, the modified SEDEX procedure (Slomp et al., 1996; Kraal et al., 2009) is employed to quantify the different forms of P in surface sediments of the YS and the ECS.

## 2 Materials and methods

Surface sediments were collected in the YS and the ECS by box sampler on R/V *Dongfanghong* 2 and *Shiyan* 3 between March and June in 2011(Fig. 1). Surface sediments (0–2 cm) were sealed in polyethylene bags and kept frozen (–20°C) for future analysis. Surface sediment samples were frozen dried using a Freeze Dryer (Alpha 1-4, Martin Christ GmbH, Germany) and grounded with an agate mortar in the home laboratory. In our previous study, P was measured using separate extractions. Inorganic P (IP) was directly measured by 1 mol/L HCl extraction (25°C for 16 h), total P (TP) was measured by 1 mol/L HCl extraction after ignition of the sediment (550°C for 2 h) (Aspila et al., 1976). In the present study, P was measured using sequential extractions. TP was divided into exchangeable P (Exch-P), Fe-bound P (Fe-P), authigenic Ca-P (ACa-P), DAP and organic P (OP). IP which is the sum of exchangeable P, Fe-bound P, ACa-P and DAP was extracted by the SEDEX procedure (Ruttenberg, 1992) as modified by Slomp et al. (1996) and Kraal et al. (2009). Exch-P was extracted with 1 mol/L $MgCl_2$ (pH 8, 30 min, 20°C). The sediment residue was extracted with citrate-dithionite-bicarbonate (CDB, pH 7.3, 8 h, 20°C) as Fe-P and the sediment residue was washed once with 1 mol/L $MgCl_2$ (pH 8, 30 min, 20°C). ACa-P was subsequently extracted with 1 mol/L Na-acetate buffer (pH 4, 6 h, 20°C) and the sediment was washed again with 1 mol/L $MgCl_2$ (pH 8, 30 min, 20°C). The rinses with $MgCl_2$ are necessary to reverse secondary adsorption. Finally the sediment residue was extracted with 1 mol/L HCl (24 h, 20°C) as DAP. OP was obtained by subtracting IP from TP. The analysis of the Chinese standard of coastal sediment (GBW 07314) gave the TP concentrations of (19.68±0.08) μmol/g, which compared well with the certified value ((20.85±1.97) μmol/g) (Liu et al., 2004). The analytical precision for the P extractions was better than 1% for TP, 1.58% for Exch-P, 7.33% for Fe-P, 1.24% for ACa-P, 1.84% for DAP and 5.00% for OP.

## 3 Results

Among the different forms of P, the P concentrations for Exch-P, Fe-P, ACa-P, DAP, and OP were 0.20–0.89 μmol/g, 0.37–2.86 μmol/g, 0.61–3.07 μmol/g, 6.39–13.73 μmol/g and 0.54–10.06 μmol/g, respectively; the average percentage of different forms of P in TP were 3%, 8%, 9%, 55% and 25%, respectively. IP was the major speciation accounting for 54%–95% in TP and had no obvious differences with that of directly measured by 1 mol/L HCl extraction in our previous work.

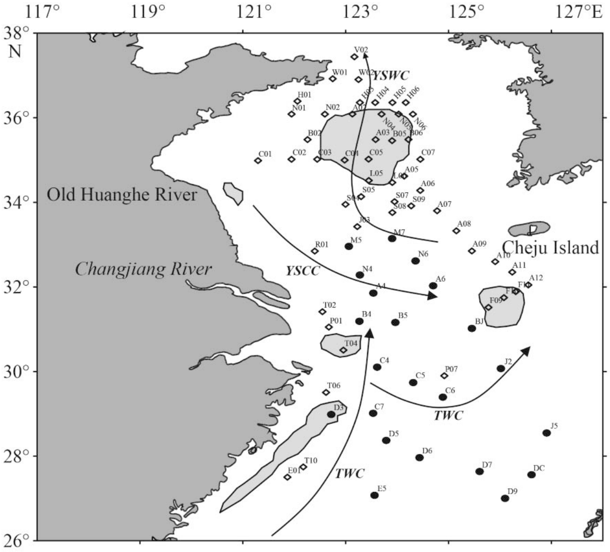

**Fig. 1.** Locations of sediment samples, currents and the distribution of fine-grain sediments in the Yellow Sea and the East China Sea. ◇stands for the locations of sediment samples sampled between March and April and ● the locations of sediment samples sampled between May and June. The currents are the Yellow Sea Warm Current (YSWC), the Yellow Sea Coastal Current (YSCC) and the Taiwan Warm Current (TWC). The gray colors indicating mud areas. The currents and mud areas are based on the studies of Bian et al. (2013) and Yuan et al. (2008).

The horizontal distributions of different forms of P in surface sediments of the YS and the ECS are shown in Fig. 2. The distribution of Exch-P, Fe-P and OP seemed to be similar. The concentrations of Exch-P, Fe-P and OP were slightly higher in the YS than that in the ECS, and low concentrations could be observed in the middle part of the ECS and southwest off Cheju Island. The distribution of ACa-P was different from those of Exch-P, Fe-P and OP. The concentrations of ACa-P decreased southward, but the maximum of ACa-P appeared in the southeast of the study area. The concentration of DAP had two high values located in the south and north parts of the Changjiang River Estuary and decreased eastward obviously. The minimum of DAP could be observed in the northern part of the southern YS.

## 4 Discussion

### 4.1 *Phosphorus forms*

With the development of industrialization and urbanization, so many sewage effluents with abundant nutrients have been discharged into the coastal ocean through different pathways. Atmospheric deposition was also the predominant pathway for P to the Yellow Sea, and was in the same magnitude as the rivers input (Zhang et al., 2007b; Shi et al., 2013). While in the ECS, at-

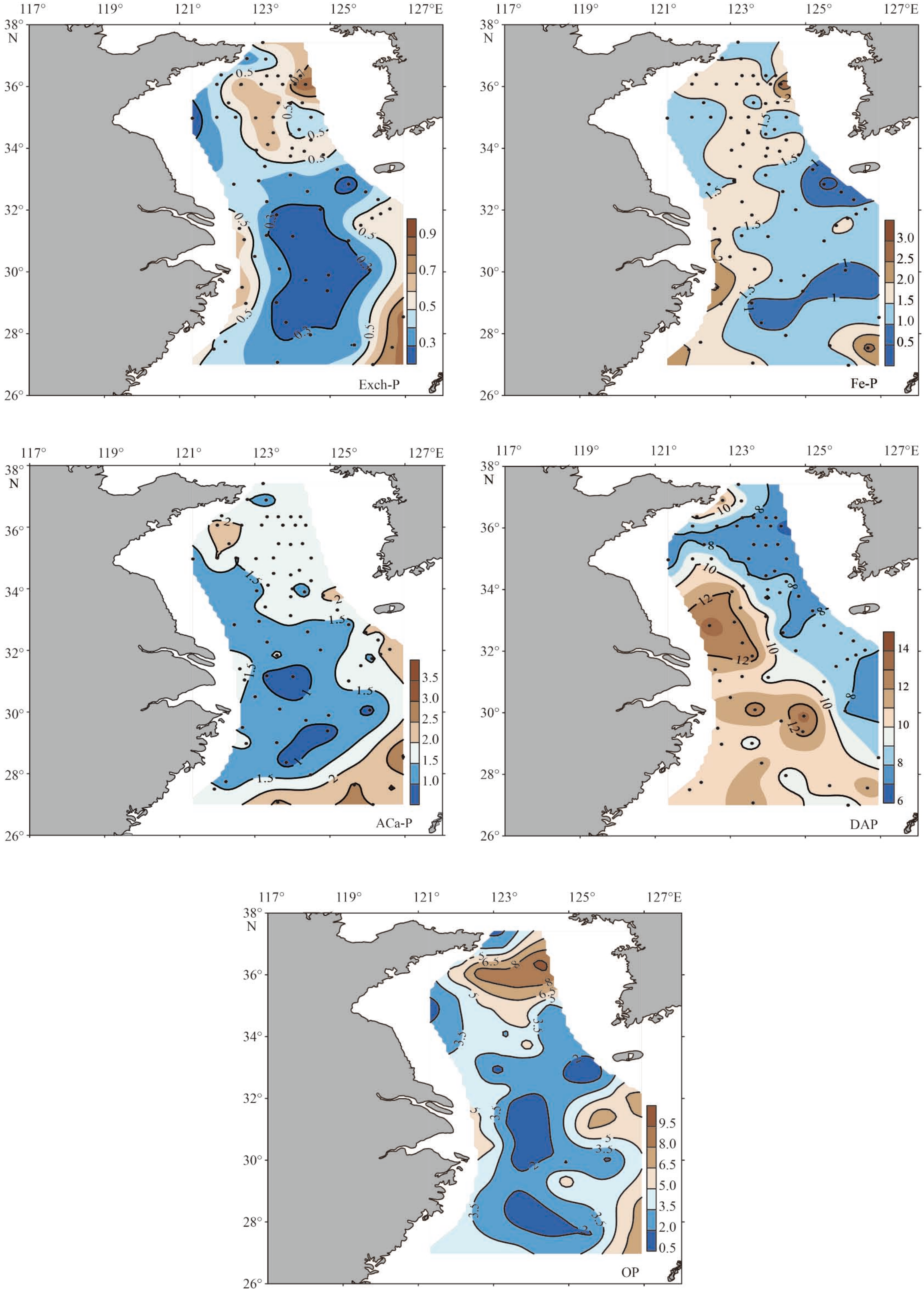

**Fig. 2.** The contours of different forms of P in surface sediments (μmol/g).

mospheric deposition was almost equal to the rivers transportation. In addition, Kuroshio's intrusion and phosphate transported through the Taiwan Strait are also important sources of P (Zhang et al., 2007a). The majority of the riverine P is retained within continental shelf sediments and cannot reach the open ocean (Ruttenberg, 1993). The concentration of TP in suspended particulates of the rivers in the world varies from 18 to 64 μmol/g, with a mean of 37 μmol/g (Martin and Meybeck, 1979). While the total concentration of particulate P was 23 μmol/g in the Changjiang River and 18 μmol/g in the Huanghe River (Yellow River) (Qu et al., 1993; Meng et al., 2007). Marine sediments are mixtures of marine and terrestrial materials. The concentration of TP in surface sediment in the research area was 10.50 to 24.10 μmol/g, and it may be affected by sources of input materials, grain size, depositional environments, hydrological conditions and anthropogenic activities (Song et al., 2014; Song et al., 2006; Hou et al., 2001). These values obtained in the present study agree with those of the other marginal seas, such as the Iberian margin (van der Zee et al., 2002), the Gulf of Mexico (Filipek and Owen, 1981), the Amazon Shelf (Berner and Rao, 1994) and the North Sea (Slomp et al., 1998).

Exch-P represents the loosely absorbed P that comprises pore water P and leached P from decaying cells of bacterial biomass in deposited phytodetrital aggregates (Rydin, 2000; Pettersson, 2001). Exch-P is low, but it is easily available by algae. High values of Exch-P could be observed in the southern YS mud area, Zhejiang and Fujian mud area and outer shelf of the ECS. Although the material source for the muddy patches are different, the sediment is dominantly fine-grained clay (Fig. 1) and fine-grained clay has a large capability to adsorb P compounds (Salomons and Gerritse, 1981).

Fe-P is the indicator for the extent of environment pollution and tracer of material source (Hisashi, 1983). The Fe-P concentration in sediments of the Huanghe River Estuary was lower than that in river sediments. This may result from the input of marine sediments and the reduction of sulfate. The Fe-P concentration was more than 10 μmol/g in sediments in lower reach of the Huanghe River (Qu et al., 2014). While in the YS sediments, the contents of Fe-P were much lower than that of the Huanghe River sediments. The concentration of Fe-P in the ECS sediments decreased eastward from the Changjiang River Estuary. These observed values consistently agreed with the concentrations of phosphate (Chen, 2008) and particulate P (Wang et al., 2004) in seawater. The distribution of Fe-P may be affected by anthropogenic activities and can reflect the decreasing tendency for the extent of environment pollution from the river to the offshore sites.

High values of OP and ACa-P can also be observed in the northern part of the southern YS. The sediment here was thought to originate from the modern Huanghe River and transported by the Bohai Sea Coastal Current (Lan et al., 2005). Only fine-grained particles can be transported a long distance and reach this region due to hydrodynamic conditions (Fig. 1), and coarse-grained particles may sink and deposit in coastal area because of weak tidal currents (Dong et al., 2011). Weak tidal and water column stratification may be favorable for fine-grained sediments to be deposited in the sea bed (Hu, 1984; Dong et al., 2011). This region is a cold vortex mud area and the local sedimentary environment is benefit for preserving organic matter (Saito and Yang, 1995; Bian et al., 2013; Yuan et al., 2008; Wei and Wang, 2012). The concentrations of chlorophyll *a* are high in spring due to the onset of the phytoplankton bloom and there has been an increase of primary production in recent years (Fu et al., 2009; Wei and Wang, 2012). Nutrients supplied by vertical mixing of water column can promote the growth of phytoplankton that is the source of sinking debris rich in organic matter and ACa-P (Wei and Wang, 2012). ACa-P in the northern part of the southern YS could reflect both terrestrial input and authigenic input from calcium organisms. ACa-P was positively correlated with clay and silt content but negatively with sand content (Yu et al., 2013). The Huanghe River sediment is characterized by high contents of apatite and Ca compared with the Changjiang River sediment (Yang et al., 2004). The similarity in phosphorus concentration of river sediments and of soils from the Loess Plateau suggests that phosphorus transport of the Huanghe River is mainly influenced by the soil loss from the Loess Plateau (Yu et al., 2010). High contents of ACa-P and OP can also be observed in the outer shelf of the ECS which may be affected by the Kuroshio. The Kuroshio has the function for transporting nutrients from its origin area to the Kuroshio Extension (Guo et al., 2012). The nutrients carried by the Kuroshio may contribute to the primary production along the shelf break of the ECS and will have an impact in the waters in this region (Guo et al., 2012; Wen et al., 2012). The mean onshore nutrient transport from the Kuroshio to the continental shelf of the ECS was 9.4 kmol/s for nitrate and 0.7 kmol/s for phosphate, respectively (Zhao and Guo, 2011). ACa-P in the outer shelf of the ECS was impacted by authigenic input from calcium-bearing organisms under the influence of the Kuroshio (Lin et al., 2002). Meanwhile, high concentration of nutrient could maintain the growth of phytoplankton and zooplankton in spring and summer. After the blooms, sinking debris of plankton rich in organic matter and ACa-P would deposit in the sediment and resulted in high contents of ACa-P and OP.

The concentrations of different forms of P in the ECS sediments decreased eastward from the Changjiang River Estuary. These observed values consistently agreed with the concentrations of phosphate (Chen, 2008) and particulate P (Wang et al., 2004) in seawater, the horizontal distribution of chlorophyll *a* (Wang et al., 2009) and distribution of primary production (Lin et al., 2011). The concentrations of DAP in continent marginal sea are affected by the land-source sediment loads; the major sources of sediments in the ECS include terrigenous sediments from the Changjiang River, relict sediment from the middle shelf, biogenic carbonate from the outer shelf and sediments from the Yellow Sea (Fang et al., 2007). In the estuarine area and the Zhejiang and Fujian mud area, the sediment is characterized by fine-grained clay; while the sediment is characterized by coarse-grained quartz sand in the middle shelf region (Fang et al., 2007). The ECS is a transition area from an inner shelf covered by detrital sediments to outer shelf and slope dominated by biogenic sediments (Niino and Emery, 1961). DAP is the major fraction of sedimentary P in the ECS. Meanwhile, DAP was found to be the major components of IP whose content was more than 50% of TP in surface sediments of the Huanghe River (Ma et al., 2009). There is a negative correlation between DAP and clay or silt, but DAP is positively correlated with sand. DAP is easier to combine with coarse particles and exhibits coarse-grain enrichment (Qu et al., 2014). Fine-grained sediments that were transported a long distance are the main components in the northern part of the southern YS, and the concentrations of DAP were lower than that of the Changjiang River Estuary and its adjacent area.

### 4.2 *Bioavailable phosphorus*

Distinguishing P speciation is essential for understanding the potential bioavailability of P in surface sediment in the study region. Bioavailable P can contribute to local primary production when reaches the seawater due to sediment resuspension and/or bioturbation. Bioavailable P is defined as the amount of immediately available P and the P which can be transformed into the available form through natural processes (Sonzogni et al., 1982). Exchangeable P is the only one of the different forms of P that can directly be uptaked by phytoplankton, but the other forms of P may transform into exchangeable P when the sedimentary environment changes. ACa-P and DAP are almost insoluble in seawater (Liu et al., 2004). When Fe-P is reduced in anoxic environment the adsorbed phosphate is released into pore water; OP can become bioavailable by remineralization (Andrieux and Aminot, 1997). The sum of Exch-P, Fe-P and OP may be thought to be potentially bioavailable P in the research region. The percentage of bioavailable P in TP ranged from 13% to 61% (Fig. 3). The percentage of bioavailable P in TP in the YS was higher than that in the ECS. In the northern part of the south YS, the concentration of OP in sediment was quite high. The sediment here was thought to originate from the modern Huanghe River and transported by the Bohai Sea Coastal Current (Lan et al., 2005). While in the central part of the ECS, the sediment is characterized by coarse-grained sand or silt (Fig. 1), and DAP was the dominant form of sediment in this area, so the percentage of bioavailable P in TP was lower than that of the southern YS.

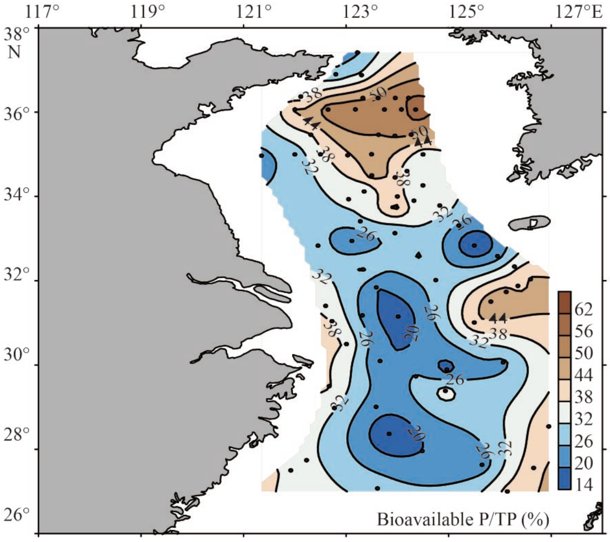

**Fig. 3.** The percentage of bioavailable P in TP.

### 4.3 *Bioavailable phosphorus burial flux*

Burial flux of biogenic elements may reflect the environmental changes and trends in a particular historical period and reveal biogeochemical cycles of biogenic elements (Song et al., 2014). P burial fluxes were calculated using the following relation: $F_b=c_i\omega=c_iS(1-\varphi)\rho_s$ (Ingall and Jahnke, 1994), where $c_i$ is the average sediment P concentration in μmol/g and $\omega$ is the sediment mass accumulation rate in g/(cm$^2$·a). For each sites where mass accumulation rate was unavailable, $\omega$ was calculated using the relation $\omega=S(1-\varphi)\rho_s$, where $S$ is the linear sedimentation rate in cm/a (Data derived from Li et al. (2012), Hu et al. (2011) and Li et al. (2002)), $\varphi$ is the average porosity and $\rho_s$ is the density of sediment solids in g/cm$^3$. A density of 2.1 g/cm$^3$ has been used for all sediments. In our previous study, TP burial flux was found to be in the range of 0.82–15.46 μmol/(cm$^2$·a) (Song et al., 2014). The present study investigated the content of bioavailable P ranged from 1.54–13.22 μmol/g in the YS and ECS. We calculated the bioavailable P burial flux to be in the range of 0.22 to 6.45 μmol/(cm$^2$·a) for the research region (Fig. 4). The distribution of bioavailable P burial flux agreed with that of TP burial flux (Fig. 4), and high values appeared in the Changjiang River Estuary and Zhejiang and Fujian mud areas, while low values could be observed in the south YS due to its low sedimentation rates and high porosity (Song et al., 2014; Li et al., 2012; Hu et al., 2011; Li et al., 2002). In the Changjiang River Estuary and its adjacent areas, a large number of terrigenous sediments transported by the Changjiang Diluted Water may sink and deposit and the sediment existing here contain huge amount of bioavailable P (Meng et al., 2014). So high values of bioavailable P burial flux in surface sediments in this region may result from high content of bioavailable P and high sedimentation rates.

### 4.4 *Potential impacts on ecosystem*

The nutrient enrichment effects can increase the phytoplankton productivity (Hodgkiss and Lu, 2004) and phytoplankton growth largely depends on the availability of inorganic nutrients. Sediment can provide a large amount of nutrient to the overlying seawater and is an internal source of nutrient in marginal seas world, including the YS and ECS (Slomp, 2011; Conley et al., 2002; Zhang et al., 2007b). Surface sediment will consistently resuspend and deposit in upwelling area or by other inducements (physical reworking or chemical drivers), and the biogenic elements in sediment may be regenerated and released to overlying water, increase the exchange of P between particles and water and enhance the primary productivity in seawater (Wang et al., 2005; Zhang et al., 2004). In our present study, the percentage of bioavailable P in TP ranged from 13% to 61%, which is a potential internal source of P in the YS and the ECS. Microbial breakdown of sedimentary organic matter and bioavailable P may result in the release of phosphate to the overlying water column and recycled where it is available for primary production and pose a threat of eutrophication to waters (Rozan et al., 2002), so regenerating dissolved P from sediments can play an important role in maintaining the low level of phosphate in the coastal area, and have an impact on phytoplankton production and community composition.

In spring, phytoplankton blooms is a common phenomenon in temperate marine systems (Fu et al., 2012), and phytoplankton can play an important role in ecosystem structure and function. The development of economic activities and increase in population are prone to promote eutrophication due to the large discharge of nutrients through several pathways (Anderson et al., 2002). Anthropogenic eutrophication and climatic changes may result in biomass increase and frequent harmful algal blooms (HABs) that may have an impact on the local ecosystem (Hays et al., 2005). High chlorophyll *a* and primary production could be observed in the southern YS and the Changjiang River Estuary due to the blooms of phytoplankton in spring (Zheng and Wei, 2010; Wen et al., 2012). Nutrients supplied by vertical mixing of water column can also promote the growth of phytoplankton in spring. After the bloom, phytodetrital aggregates would sink and supply the organic matter to the seafloor. The sinking debris would be the marine source of organic matter containing nutrient elements for the local sedimentary environment.

Riverine transportation, atmospheric deposition, the Kuroshio's incursion and the internal releasing from sediments are the sources of nutrient to the YS and the ECS; beside these sources, the nutrients transported through the Taiwan Strait are also important sources for the ECS (Zhang et al., 2007a). These nutrients are responsible for increases in algal biomass and HABs in recent years (Anderson et al., 2002), and hypoxia exacerbation in the estuaries (Zhang et al., 2010). P can be sequestered in sediments in the forms of organic P compounds and Fe-oxides in oxygenated bottom waters. In the Changjiang River Estuary, frequent resuspension and seasonal hypoxia can change the normal redox succession in sediments, making Fe and Mn experience repeated redox cycling and releasing phosphate because of the periodical reduction of Fe-P. Phosphate can also be gradually released during the process of remineralization of OP because of microbial degradation (Andrieux and Aminot, 1997). Abundant microbial diversity in the sediments of the YS and ECS can promote the decomposition of OM, and enhance the bioavailability of OP (Sutula et al., 2004). The high concentrations of phosphate in porewater and high fluxes of phosphate in sediment-water interface in the mud areas indicated the function of sediments in the release of bioavailable P (Meng et al., 2014). The primary production may be limited by P in a portion of the YS and ECS (Wen et al., 2012), while internal supply of P from sediment may supply a large amount of P to the water column and relax P-limitation primary production (Jin et al., 2013; Zhang et al., 2007a).

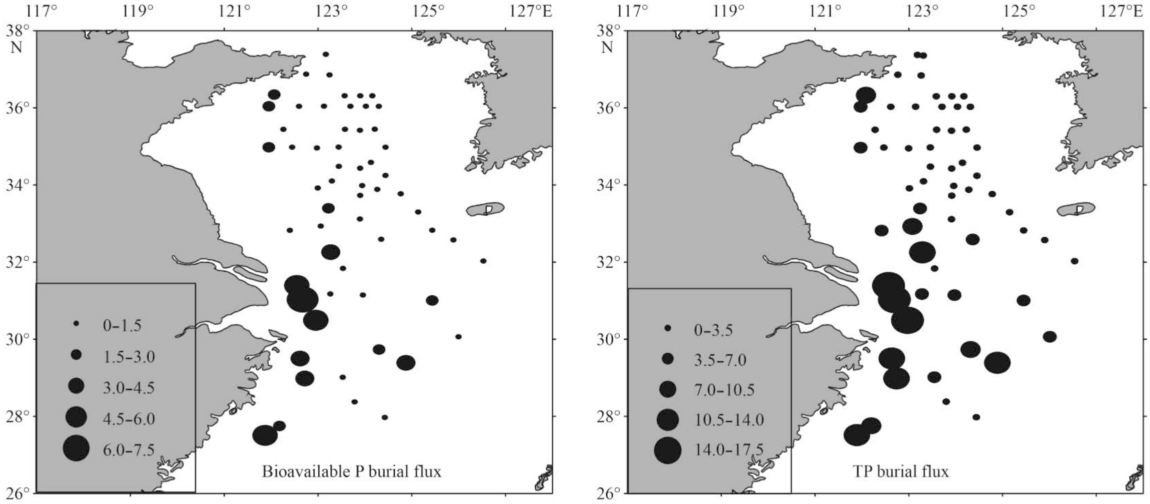

**Fig. 4.** Bioavailable P burial flux (μmol/(cm$^2$·a)) in surface sediments and TP burial flux is given for comparison.

## 5 Conclusions

In this study, the modified SEDEX procedure was employed to quantify the different forms of P in sediments of the YS and the ECS. Among the different forms of P, the P concentrations for Exch-P, Fe-P, ACa-P, DAP, and OP were 0.20–0.89 μmol/g, 0.37–2.86 μmol/g, 0.61–3.07 μmol/g, 6.39–13.73 μmol/g and 0.54–10.06 μmol/g, respectively; the average percentage of different forms of P in TP were 3%, 8%, 9%, 55% and 25%, respectively. IP was the major speciation accounting for 54%–95% in TP. The distribution of different forms of P in surface sediments may be affected by sources of input materials, grain size, depositional environments, hydrological conditions and anthropogenic activities. The percentage of bioavailable P in TP ranged from 13% to 61%. The distribution of bioavailable P burial flux was almost the same as that of TP burial flux affected by sedimentation rates, porosity and bioavailable P content. Bioavailable P which is a potential internal source of P in the YS and the ECS could be released and recycled under certain conditions and has potential impact and pressure on the marine ecosystem.

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### References

Ahlgren J, Reitzel K, Tranvik L, et al. 2006. Degradation of organic phosphorus compounds in anoxic Baltic Sea sediments: A $^{31}$P nuclear magnetic resonance study. Limnology and Oceanography, 51(5): 2341–2348

Anderson D M, Glibert P M, Burkholder J M. 2002. Harmful algal blooms and eutrophication: nutrient sources, composition, and consequences. Estuaries, 25(4): 704–726

Andrieux F, Aminot A. 1997. A two-year survey of phosphorus speciation in the sediments of the Bay of Seine (France). Continental Shelf Research, 17(10): 1229–1245

Aspila K I, Agemian H, Chau A S Y. 1976. A semi-automated method for the determination of inorganic, organic and total phosphate in sediments. Analyst, 101(1200): 187–197

Berner R A, Rao Jilong. 1994. Phosphorus in sediments of the Amazon River and estuary: implications for the global flux of phosphorus to the sea. Geochimica et Cosmochimica Acta, 58(10): 2333–2339

Bian Changwei, Jiang Wensheng, Quan Qi, et al. 2013. Distributions of suspended sediment concentration in the Yellow Sea and the East China Sea based on field surveys during the four seasons of 2011. Journal of Marine Systems, 121–122: 24–35

Broecker W S, Peng T H. 1982. Tracers in the Sea. Lamont-Doherty Geological Observatory. Palisades, New York: Columbia University

Cao Peikui, Hu Fangxi, Gu Guochuan, et al. 1989. Relationship between suspended sediments from the Changjiang Estuary and the evolution of the embayed muddy coast of Zhejiang Province. Acta Oceanologica Sinica, 8(2): 273–283

Cao Wenqing, Liu Sumei. 2010. A study on inorganic and organic phosphorus in core sediment of the Yellow and East China Seas. Periodical of Ocean University of China (in Chinese), 40(1): 69–74

Chai Chao, Yu Zhiming, Shen Zhiliang, et al. 2009. Nutrient characteristics in the Yangtze River Estuary and the adjacent East China Sea before and after impoundment of the Three Gorges Dam. Science of the Total Environment, 407(16): 4687–4695

Chen C T A. 2008. Distributions of nutrients in the East China Sea and the South China Sea connection. Journal of Oceanography, 64(5): 737–751

Conley D J, Humborg C, Rahm L, et al. 2002. Hypoxia in the Baltic Sea and basin-scale changes in phosphorus biogeochemistry. Environmental Science & Technology, 36(24): 5315–5320

Diaz R J, Rosenberg R. 1995. Marine benthic hypoxia: A review of its ecological effects and the behavioural responses of benthic macrofauna. Oceanography and Marine Biology. An Annual

Review, 33: 245–303

Dong Lixian, Guan Weibing, Chen Qi, et al. 2011. Sediment transport in the Yellow Sea and East China Sea. Estuarine, Coastal and Shelf Science, 93(3): 248–258

Dong Zhijun, Liu Dongyan, Keesing J K. 2010. Jellyfish blooms in China: Dominant species, causes and consequences. Marine Pollution Bulletin, 60(7): 954–963

Fang T H, Chen J L, Huh C A. 2007. Sedimentary phosphorus species and sedimentation flux in the East China Sea. Continental Shelf Research, 27(10–11): 1465–1476

Feng Qiang, Liu Sumei, Zhang Jing. 2001. Distribution of phosphorus in sediment of the Bohai and Yellow Sea. Marine Environmental Science (in Chinese), 20(2): 24–27, 41

Filipek L H, Owen R M. 1981. Diagenetic controls of phosphorus in outer continental shelf sediments from the Gulf of Mexico. Chemical Geology, 33: 181–204

Fu Mingzhu, Wang Zongling, Li Yan, et al. 2009. Phytoplankton biomass size structure and its regulation in the Southern Yellow Sea (China): Seasonal variability. Continental Shelf Research, 29(18): 2178–2194

Fu Mingzhu, Wang Zongling, Pu Xinming, et al. 2012. Changes of nutrient concentrations and N: P: Si ratios and their possible impacts on the Huanghai Sea ecosystem. Acta Oceanologica Sinica, 31(4): 101–112

Gao Lei, Li Daoji. 2009. Changes of nutrient concentrations in western areas of Yellow Sea and East China Sea in recent several decades. Marine Science (in Chinese), 33(5): 64–69

Guo Xinyu, Zhu Xiaohua, Wu Qingsong, et al. 2012. The Kuroshio nutrient stream and its temporal variation in the East China Sea. Journal of Geophysical Research, 117(C1): C01026

Hays G C, Richardson A J, Robinson C. 2005. Climate change and marine plankton. Trends in Ecology & Evolution, 20(6): 337–344

Hisashi J. 1983. Fractionation of phosphorus and releasable traction in sediment mud of Osaka Bay. Bulletin of the Japanese Society of Scientific Fisheries, 49(3): 447–454

Hodgkiss I J, Lu Songhui. 2004. The effects of nutrients and their ratios on phytoplankton abundance in Junk Bay, Hong Kong. Hydrobiologia, 512(1–3): 215–229

Hou Lijun, Liu Min, Xu Shiyuan, et al. 2001. Species of phosphorus in core sediments from the Changjiang Estuary and its environmental significance. Marine Environmental Science (in Chinese), 20(2): 7–12

Hu Dunxin. 1984. Upwelling and sedimentation dynamics: I. The role of upwelling in sedimentation in the Huanghai Sea and East China Sea-a description of general features. Chinese Journal of Oceanology and Limnology, 2(1): 12–19

Hu Bangqi, Li Guogang, Li Jun, et al. 2011. Spatial variability of the $^{210}$Pb sedimentation rates in the Bohai and Huanghai Seas and its influencing factors. Haiyang Xuebao (in Chinese), 33(6): 125–133

Humborg C, Ittekkot V, Cociasu A V, et al. 1997. Effect of Danube River dam on Black Sea biogeochemistry and ecosystem structure. Nature, 386(6623): 385–388

Ingall E, Jahnke R. 1994. Evidence for enhanced phosphorus regeneration from marine sediments overlain by oxygen depleted waters. Geochimica et Cosmochimica Acta, 58(11): 2571–2575

Jin Jie, Liu Sumei, Ren Jingling, et al. 2013. Nutrient dynamics and coupling with phytoplankton species composition during the spring blooms in the Yellow Sea. Deep-Sea Research Part II: Topical Studies in Oceanography, 97: 16–32

Kawasaki N, Ogata F, Tominaga H. 2010. Selective adsorption behavior of phosphate onto aluminum hydroxide gel. Journal of Hazardous Materials, 181(1–3): 574–579

Kraal P, Slomp C P, Forster A. et al. 2009. Pyrite oxidation during sample storage determines phosphorus fractionation in carbonate-poor anoxic sediments. Geochimica et Cosmochimica Acta, 73(11): 3277–3290

Lan Xianhong, Zhang Xunhua, Zhang Zhixun. 2005. Material sources and transportation of sediments in the southern Yellow Sea. Transactions of Oceanology and Limnology (in Chinese), (4): 53–60

Li Fengye, Gao Shu, Jia Jianjun, et al. 2002. Contemporary deposition rates of fine-grained sediment in the Bohai and Yellow Seas. Oceanologia et Limnologia Sinica (in Chinese), 33(4): 364–369

Li Jun, Hu Bangqi, Dou Yanguang, et al. 2012. Modern sedimentation rate, budget and supply of the muddy deposits in the East China Seas. Geological Review (in Chinese), 58(4): 745–756

Lin S, Hsieh I J, Huang K M, et al. 2002. Influence of the Yangtze River and grain size on the spatial variations of heavy metals and organic carbon in the East China Sea continental shelf sediments. Chemical Geology, 182(2–4): 377–394

Lin Chuanlan, Ning Xiuren, Su Jilan, et al. 2005. Environmental changes and the responses of the ecosystems of the Yellow Sea during 1976–2000. Journal of Marine Systems, 55(3–4): 223–234

Lin Zhiyu, Tong Jinlu, Chen Min. 2011. Distribution of primary production in the Yellow Sea and the East China Sea in summer. Journal of Isotopes (in Chinese), 24(Z1): 95–101

Liu Sumei, Zhang Jing, Li Daoji. 2004. Phosphorus cycling in sediments of the Bohai and Yellow Seas. Estuarine, Coastal and Shelf Science, 59(2): 209–218

Ma Qin, Li Beigang, Jiao Xiaobao. 2009. Distribution characteristics and bioavailability of phosphorus in surface sediments from the Yellow River. Journal of Agro-Environment Science (in Chinese), 28(11): 2379–2384

Martin J M, Meybeck M. 1979. Elemental mass-balance of material carried by major world rivers. Marine Chemistry, 7(3): 173–206

Meng Jia, Yao Peng, Yu Zhigang, et al. 2014. Speciation, bioavailability and preservation of phosphorus in surface sediments of the Changjiang Estuary and adjacent East China Sea inner shelf. Estuarine, Coastal and Shelf Science, 144: 27–38

Meng Wei, Yu Tao, Zheng Binghui, et al. 2007. Variation and influence factors of nitrogen and phosphorus transportation by the Yellow River. Acta Scientiae Circumstantiae (in Chinese), 27(12): 2046–2051

Milliman J D. 1991. Flux and fate of fluvial sediment and water in coastal seas. In: Mantoura R F C, Martin J M, Wollast R, eds. Ocean Margin Processes in Global Change. New York: Wiley, 69–89

Niino H, Emery J O. 1961. Sediments of shallow portions of East China Sea and South China Sea. Geological Society of America Bulletin, 72(5): 731–762

Paytan A, McLaughlin K. 2007. The oceanic phosphorus cycle. Chemical Reviews, 107(2): 563–576

Pettersson K. 2001. Phosphorus characteristics of settling and suspended particles in Lake Erken. Science of the Total Environment, 266(1–3): 79–86

Qu C H, Chen C Z, Yang J R, et al. 1993. Geochemistry of dissolved and particulate elements in the major rivers of China (The Huanghe, Changjiang and Zhunjiang Rivers). Estuaries, 16(3): 475–487

Qu Yingxuan, Sheng Yanqing, Ding Chaofeng, et al. 2014. Gochemistry characteristic of phosphorus in costal surface sediments. China Environmental Science, 34(1): 246–252

Rozan T F, Taillefert M, Trouwborst R E, et al. 2002. Iron-sulfur-phosphorus cycling in the sediments of a shallow coastal bay: Implications for sediment nutrient release and benthic macroalgal blooms. Limnology and Oceanography, 47(5): 1346–1354

Ruttenberg K C. 1992. Development of a sequential extraction method for different forms of phosphorus in marine sediments. Limnology and Oceanography, 37(7): 1460–1482

Ruttenberg K C. 1993. Reassessment of the oceanic residence time for phosphorus. Chemical Geology, 107(3–4): 405–409

Ruttenberg K C, Berner R A. 1993. Authigenic apatite formation and burial in sediments from non-upwelling, continental margin environments. Geochimica et Cosmochimica Acta, 57(5): 991–1007

Rydin E. 2000. Potentially mobile phosphorus in lake Erken sediment. Water Research, 34(7): 2037–2042

Saito Y, Yang Zuosheng. 1994. Historical change of the Huanghe (Yellow River) and its impact on the sediment budget of the East

China Sea. In: Tsunogai S, Iseki K, Koike I, et al., eds. Global Fluxes of Carbon and Its Related Substances in the Coastal Sea-Ocean-Atmosphere System. Sapporo: Hokkaido University, 7–12

Salomons W, Gerritse R G. 1981. Some observations on the occurrence of phosphorus in recent sediments from Western Europe. Science of the Total Environment, 17(1): 37–49

Schenau S J, Reichart G J, De Lange G J. 2005. Phosphorus burial as a function of paleoproductivity and redox conditions in Arabian Sea sediments. Geochimica et Cosmochimica Acta, 69(4): 919–931

She Jun. 1999. Pollution in the Yellow Sea large marine ecosystem: monitoring, research and ecological effects. In: Tang Qisheng, Sherman K, eds. Large Marine Ecosystems of the Pacific Rim: Assessment, Sustainability, and Management. Malden, US: Blackwell Science, 419–426

Shi Jinhui, Zhang Jing, Gao Huiwang, et al. 2013. Concentration, solubility and deposition flux of atmospheric particulate nutrients over the Yellow Sea. Deep-Sea Research Part II: Topical Studies in Oceanography, 97: 43–50

Slomp C P. 2011. Phosphorus cycling in the estuarine and coastal zones: sources, sinks, and transformations. In: Wolanski E, McLusky D S, eds. Treatise on Estuarine and Coastal Science, Vol. 5. Waltham: Academic Press, 201–229

Slomp C P, Epping E H G, Helder W, et al. 1996. A key role for iron-bound phosphorus in authigenic apatite formation in North Atlantic continental platform sediments. Journal of Marine Research, 54(6): 1179–1205

Slomp C P, Malschaert J F P, Van Raaphorst W. 1998. The role of adsorption in sediment-water exchange of phosphate in North Sea continental margin sediments. Limnology and Oceanography, 43(5): 832–846

Song Jinming, Li Xuegang, Shao Junbo, et al. 2006. Biogeochemical characteristics of nitrogen and phosphorus in the south Yellow Sea sediments. Oceanologia et Limnologia Sinica (in Chinese), 37(4): 370–376

Song Guodong, Liu Sumei, Zhang Guoling. 2014. Study on distribution of phosphorus in surface sediments of the Yellow Sea and the East China Sea. Environmental Science (in Chinese), 35(1): 157–162

Sonzogni W C, Chapra S C, Armstrong D E, et al. 1982. Bioavailability of phosphorus inputs to lakes. Journal of Environmental Quality, 11(4): 555–563

Sternberg R W, Larsen L H, Miao Y T. 1985. Tidally driven sediment transport on the East China Sea continental shelf. Continental Shelf Research, 4(1–2): 105–120

Sun Song, Wang Fan, Li Chaolun, et al. 2008. Emerging challenges: Massive green algae blooms in the Yellow Sea. Nature Precedings: hdl:10101/npre.2008.2266.1

Sutula M, Bianchi T S, McKee B A. 2004. Effect of seasonal sediment storage in the lower Mississippi River on the flux of reactive particulate phosphorus to the Gulf of Mexico. Limnology and Oceanography, 49(6): 2223–2235

Tang Qisheng, Zhang Jing, Su Jilan, et al. 2013. Spring blooms and the ecosystem processes: The case study of the Yellow Sea. Deep-Sea Research Part II, 97: 1–3

Van Cappellen P, Ingall E D. 1996. Redox stabilization of the atmosphere and oceans by phosphorus-limited marine productivity. Science, 271(5248): 493–496

Van der Zee C, Slomp C P, van Raaphorst W. 2002. Authigenic P formation and reactive P burial in sediments of the Nazaré canyon on the Iberian margin (NE Atlantic). Marine Geology, 185(3–4): 379–392

Wang Zuohua, Shi Xiaoyong, Zhang Chuansong, et al. 2009. Primary study on characteristics of distribution of chlorophyll a in the Yellow Sea and East China Sea. Progress in Fishery Sciences (in Chinese), 30(2): 95–101

Wang Pibo, Song Jinming, Guo Zhanyong, et al. 2005. The inducements of resuspention of sea surface sediment and their effects on recycle of biogenic elements. Marine Sciences (in Chinese), 29(10): 77–80

Wang Xiulin, Sun Xia, Han Xiurong, et al. 2004. Comparison in macronutrient distributions and composition for high frequency HAB occurrence areas in East China Sea between summer and spring 2002. Oceanologia et Limnologia Sinica (in Chinese), 35(4): 323–331

Wang Baodong, Wang Xiulin, Zhan Run. 2003. Nutrient conditions in the Yellow Sea and the East China Sea. Estuarine, Coastal and Shelf Science, 58(1): 127–136

Wei Qinsheng, Wang Baodong. 2012. Distributions of carbon, nitrogen and phosphorus in the surface sediments and their ecological implications in the areas of Cold Water Mass and off the western coast of the Southern Yellow Sea. Acta Scientiae Circumstantiae (in Chinese), 32(7): 1697–1707

Wen Fei, Sun Xiaoxia, Zheng Shan, et al. 2012. Spatial and seasonal variations of chlorophyll a and primary productivity in spring and summer in the Yellow Sea and East China Sea. Oceanologia et Limnologia Sinica (in Chinese), 43(3): 438–444

Yang Shouye, Jung H S, Li Congxian. 2004. Two unique weathering regimes in the Changjiang and Huanghe drainage basins: geochemical evidence from river sediments. Sedimentary Geology, 164(1–2): 19–34

Yu Tao, Meng Wei, Ongley E, et al. 2010. Long-term variations and causal factors in nitrogen and phosphorus transport in the Yellow River, China. Estuarine, Coastal and Shelf Science, 86(3): 345–351

Yu Yu, Song Jinming, Li Xuegang, et al. 2013. Environmental significance of biogenic elements in surface sediments of the Changjiang Estuary and its adjacent areas. Journal of Environmental Sciences, 25(11): 2185–2195

Yuan Dongliang, Zhu Jianrong, Li Chunyan, et al. 2008. Cross-shelf circulation in the Yellow and East China Seas indicated by MODIS satellite observations. Journal of Marine Systems, 70(1–2): 134–149

Zhang Jiazhong, Fischer C J, Ortner P B. 2004. Potential availability of sedimentary phosphorus to sediment resuspension in Florida Bay. Global Biogeochemical Cycles, 18(4): GB4008

Zhang Jing, Gilbert D, Gooday A J, et al. 2010. Natural and human-induced hypoxia and consequences for coastal areas: synthesis and future development. Biogeosciences, 7: 1443–1467

Zhang Jing, Liu Sumei, Ren Jingling, et al. 2007a. Nutrient gradients from the eutrophic Changjiang (Yangtze River) Estuary to the oligotrophic Kuroshio waters and re-evaluation of budgets for the East China Sea Shelf. Progress in Oceanography, 74(4): 449–478

Zhang Guosen, Zhang Jing, Liu Sumei. 2007b. Characterization of nutrients in the atmospheric wet and dry deposition observed at the two monitoring sites over Yellow Sea and East China Sea. Journal of Atmospheric Chemistry, 57(1): 41–57

Zhao Liang, Guo Xinyu. 2011. Influence of cross-shelf water transport on nutrients and phytoplankton in the East China Sea: a model study. Ocean Science, 7: 27–43

Zheng Xiaoshen, Wei Hao. 2010. Analysis of chlorophyll concentration during the phytoplankton spring bloom in the Yellow Sea based on the MODIS data. In: Li Kang, Jia Li, Sun Xin, eds. Life System Modeling and Intelligent Computing. v 6330. Berlin Heidelberg: Springer, 254–261

Zheng Libo, Ye Ying, Zhou Huaiyang, et al. 2003. Distribution of different forms of phosphorus in seabed sediments from East China Sea and its environmental significance. Oceanologia et Limnologia Sinica (in Chinese), 34(3): 274–282