



Enhancement of phosphorus storage capacity of sediments by coastal wetland restoration, Yellow River Delta, China

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ABSTRACT

Un-restored (R_0) and restored (in 2006 (R_{2006}) and in 2002, (R_{2002})) coastal wetlands were selected to study phosphorus (P) storage capacity and the risk of P loss in the Yellow River Delta (YRD). The results showed that the maximum P sorption capacity (Q_{max}) in the wetland sediments varied between 201.8–1168.6 mg kg⁻¹; the average value was 576.2 mg kg⁻¹. Q_{max} increased with increasing time since restoration ($R_{2002} > R_{2006} > R_0$). The eutrophication risk index (ERI) ranged between 0.27–2.07% and decreased with increasing time since restoration, but the relationship was not statistically significant ($P > 0.05$). A correlation analysis demonstrated that Fe_{ox} , Mg, and Al_{ox} are the main P sorption agents in wetland sediments, whereas TOC, pH, and clay particles have an important effect on P sorption and release. The analysis suggests that we should restore degraded coastal wetlands to help reduce the P load to offshore waters.

1. Introduction

Phosphorus (P) is an essential nutrient for ecosystems and a key factor controlling the eutrophication of water bodies (Sundareswar et al., 2003; Schindler et al., 2008). The rapid social-economic development of coastal environments has increased the input of P from many point and non-point sources to offshore areas, resulting in a multiple-fold increase in P content in offshore waters. For example, the input of labile P to China's offshore waters increased by 2–5 fold from 1970 to 2000, and it is expected that the amount of P transported by runoff to the coast will continue to increase by 1–3 fold from 2000 to 2050 (Strokal et al., 2014). Excessive P input has altered P cycling in the offshore ecosystem, and the water body is in a state of severe eutrophication. According to the Bulletin for the Environmental Quality of Coastal Waters in China (2013–2017), the water within all of the important bays in China, including Liaodong Bay, Bohai Bay, the Yangtze River Estuary, and the Pearl River Estuary are heavily polluted, particularly with regards to inorganic nitrogen and labile phosphate. The

increase in P content has triggered frequent occurrences of red and green tides in offshore waters, resulting in the deterioration of water quality, a sharp decline in fishery resources, and an imbalance in the ecosystem. When combined, these changes have seriously affected the sustainable development of the regional economy and its associated natural resources (Smith and Schindler, 2009).

Wetlands are known as the “kidneys” of the Earth, and are a natural purifier for wastewater. Coastal wetlands are located at the intersection of land and sea, and are the last barrier to keep terrestrial pollutants (e.g., P) from entering the ocean. Coastal wetlands have a strong ability to filter external P (Reddy et al., 1999, 2011). Coastal wetlands act as one of the world's most important “sources,” “sinks,” and “reactors” of P, and therefore play a critical role in the global P cycle (Reddy et al., 2011). Coastal wetlands in the Yellow River Delta (YRD) are one of the youngest and most extensive wetland areas in a warm temperate climate in China. However, in recent years, severe wetland degradation has occurred in the YRD because of reduced freshwater inputs, increased point and non-point source pollution, and the marine erosion of

Abbreviations: P, Phosphorus; YRD, Yellow River Delta; EPC0, equilibrium phosphorus concentration; NAP, native adsorbed phosphorus concentration; Q_{max} , maximum phosphorus sorption capacity; DPS, degree of phosphorus saturation; ERI, eutrophication risk index; TOC, total organic carbon; TN, total nitrogen; TP, total phosphorus; PSI, phosphorus sorption index; k, sorption constant; MBC, phosphorus maximum buffer capacity; K_p , solid-liquid partition coefficient; Olsen-P, available soil phosphorus

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wetland sediments (Wang et al., 2012; Bai et al., 2019a). In response to these threats, flow and sediment regulations were implemented in 2002 at the Xiaolangdi dam to increase water discharge and sediment loads to lower reaches of the Yellow River (Bai et al., 2019b). At the same time, artificial dikes and channels were constructed to deliver Yellow River water to the degraded wetlands. With implementation of these wetland restoration projects, the salinity of the wetlands was reduced, and plant coverage and growth increased, thereby promoting wetlands functioning and wildlife habitats in the Yellow River Delta (Cui et al., 2009; Lu et al., 2018). However, while P is an important nutrient in wetland productivity, little is known about the effects of wetland restoration on the retention capacity and potential loss of P from sediment within the YRD.

The present study hypothesized that the P storage capacity of wetlands decreases with increasing time since restoration because P sorption sites in the wetland will gradually become saturated. In contrast, the risk of P loss from the wetland sediments will increase with restoration time. The aims of this study were to (1) characterize P sorption capacity of, and risk of P release from, YRD wetlands restored at different times in the past; (2) identify the main factors controlling P sorption and the risk of P loss from the sediments; and (3) evaluate the policy of wetland restoration based on P sorption/release.

2. Materials and methods

2.1. Study sites and sample collection

The study sites (37°26′–38°09′N, 118°33′–119°18′E) are located in the Yellow River Delta (YRD) of China (Fig. 1), one of the most active deltas in terms of land-ocean interaction in the world. Large amounts of sediment are carried by the Yellow River and deposited at the river mouth to form the youngest wetland in China. The annual average temperature is 12.1 °C and the annual average rainfall is 552 mm. The major vegetation in this area includes *Phragmites australis*, *Tamarix chinensis*, and *Suaeda salsa*. The texture of the sediments ranges from loam to silty loam; their organic-matter content was low due to its short formation time and high salinity (Cui et al., 2009). During this study, the effect of freshwater restoration projects on P storage capacity was investigated in three typical wetlands covered by *Phragmites australis*.

These three wetlands included a wetland restored in 2002 (R₂₀₀₂), a wetland restored in 2006 (R₂₀₀₆), and a wetland (R₀) that possessed no restoration projects at the time of data collection.

Within each studied wetland, three sampling sites were selected. At each of these sampling sites, five randomly distributed sediment profile (cores) were collected. The sediment profile was then sectioned at depths of 0–5 cm, 5–10 cm, 10–20 cm, and 20–40 cm. The sediments from the same depth in the five profiles were combined to get a more representative sample. All of the samples were freeze-dried. Impurities in the samples were subsequently removed, and samples were ground, passed through a 0.149 mm sieve, and stored in a refrigerator at 4 °C for future use.

2.2. Experimental methods

2.2.1. Phosphorus sorption experiment

Duplicated sediments from the three studied wetlands were used for the P sorption experiment. Approximately 1 g samples were placed into screw-cap centrifuge tubes (50 mL), along with a 20 mL phosphate standard solution (as KH₂PO₄). Concentrations of standards included 0.00, 0.50, 1.00, 2.50, 5.00, 10.00, 15.00, 30.00, 50.00, 75.00, 100.00, 120.00, 160.00, and 240.00 mg L⁻¹. After mixing, the solution was shaken at 25 °C for 24 h to reach equilibrium. After shaking, the sample was centrifuged for 10 min at 4000 rmin⁻¹ (Nair and Reddy, 2013). The supernatant was filtered through a 0.45 µm membrane and the P content was measured by a continuous flow technique. The sorbed P was calculated by the difference between the added amount of P and the measured amount of P in the equilibrium solution.

The sorption data were fitted using a modified Langmuir model, and the maximum P sorption capacity (Q_{max}) and sorption constant (k) of the sediment were obtained from the model. The modified Langmuir sorption isotherm parameters are expressed in equations (1)–(5) (Zhou et al., 2005).

$$Q_e = \frac{Q_{max} \times k \times C_e}{1 + k \times C_e} - NAP \quad (1)$$

$$MBC = Q_{max} \times k \quad (2)$$

$$NAP = \frac{Q_{max} \times k \times C_0}{1 + k \times C_0} + C_0 \frac{V}{W} \quad (3)$$

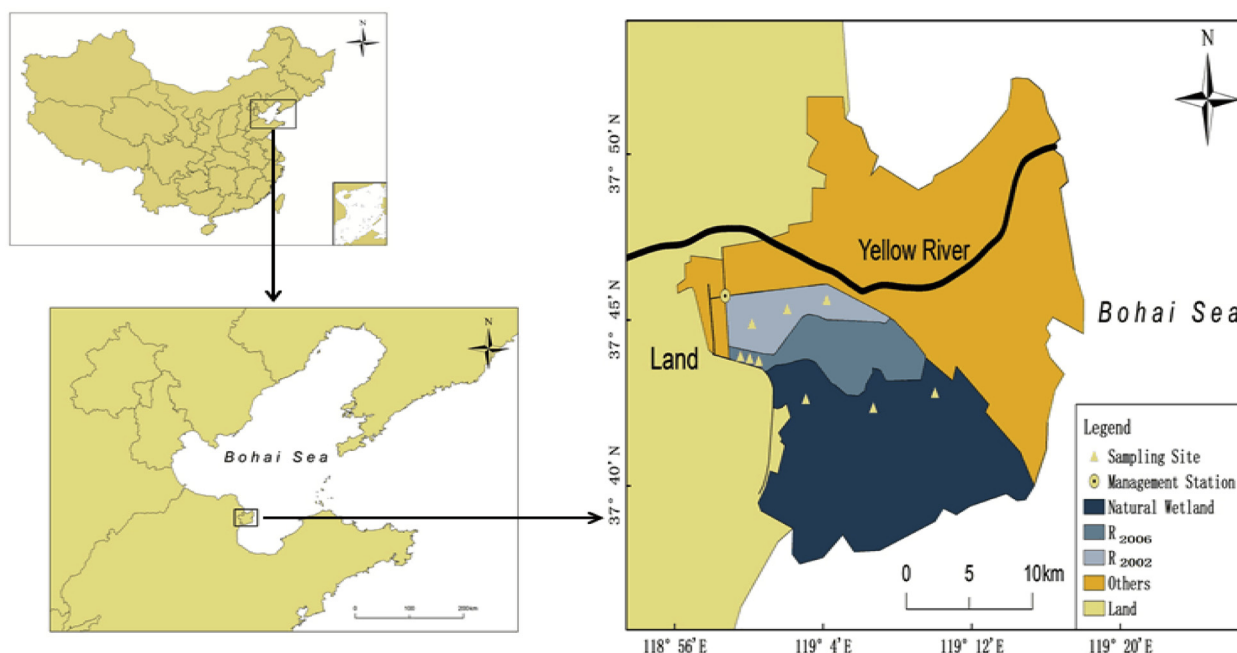


Fig. 1. Maps showing (A) the location of Bohai Sea in China; (B) the location of the study area in the Bohai Sea, and (C) the location of the sampled wetlands.

Table 1

The physicochemical properties of different sediments. Values reported in the table are the mean \pm standard deviation. The different lowercase letters indicate significant differences among data of different depths from the same column, and different uppercase letters indicate significant differences among data of different restoration time from the same sediment depth.

Sampling site	Depth (cm)	pH	< 5 μ m Clay particle (%)	TOC (mg g ⁻¹)	TN (mg kg ⁻¹)	TP (mg kg ⁻¹)	Fe _{ox} (mg kg ⁻¹)	Al _{ox} (mg kg ⁻¹)	P _{ox} (mg kg ⁻¹)	Ca (mg kg ⁻¹)	Mg (mg kg ⁻¹)
R ₀	0–5	8.2 \pm 0.1bA	17.9 \pm 0.6aB	4.5 \pm 0.7aB	423.1 \pm 15.9aA	495.1 \pm 5.5aA	2244 \pm 259aB	464 \pm 310aA	156 \pm 36aA	12037 \pm 978aA	11550 \pm 1071aB
	5–10	8.4 \pm 0.0abA	15.8 \pm 6.0abB	2.0 \pm 0.5bB	278.6 \pm 129.6abC	473.8 \pm 15.0abA	1402 \pm 161bA	260 \pm 99aA	156 \pm 13aB	13155 \pm 783aA	10217 \pm 125abB
	10–20	8.4 \pm 0.1aA	11.2 \pm 2.5abA	2.0 \pm 0.7bA	250.3 \pm 43.7abA	494.6 \pm 3.2aA	504 \pm 25cB	397 \pm 136aA	158 \pm 78aA	11632 \pm 1589aA	8931 \pm 1269cbB
	20–40	8.4 \pm 0.0aA	7.0 \pm 1.7bA	1.2 \pm 0.2bA	141.0 \pm 40.4bB	458.9 \pm 6.9bA	432 \pm 42cB	224 \pm 8aA	82 \pm 6aB	11898 \pm 507aAB	7605 \pm 774cB
R ₂₀₀₆	0–5	8.0 \pm 0.1bA	17.7 \pm 6.2aB	14.7 \pm 9.0aA	1091.9 \pm 579.1aA	548.7 \pm 31.5aA	1828 \pm 549aB	767 \pm 166aA	258 \pm 84aA	8429 \pm 982bB	10015 \pm 1019bB
	5–10	8.2 \pm 0.1aA	17.7 \pm 5.6aB	6.8 \pm 2.2bA	616.7 \pm 146.7abB	518.8 \pm 19.0aA	1590 \pm 477abA	847 \pm 373aA	223 \pm 46abAB	11847 \pm 790aA	9479 \pm 573bB
	10–20	8.3 \pm 0.1aA	18.0 \pm 9.6aA	3.7 \pm 1.0bA	450.8 \pm 207.2bA	519.6 \pm 36.5aA	1329 \pm 308abA	553 \pm 192aA	150 \pm 26bA	12325 \pm 582aA	9693 \pm 1446bB
	20–40	8.2 \pm 0.1aB	14.2 \pm 8.5aA	2.5 \pm 0.8bA	265.3 \pm 52.4bA	513.6 \pm 37.1aA	989 \pm 215bA	420 \pm 125aA	130 \pm 17cA	12488 \pm 461aA	11701 \pm 619aA
R ₂₀₀₂	0–5	8.0 \pm 0.3aA	30.2 \pm 2.2aA	16.5 \pm 2.8aA	1184.8 \pm 243.8aA	505.5 \pm 14.3aA	3514 \pm 302aA	766 \pm 203abA	297 \pm 50aA	10814 \pm 98aA	14871 \pm 1013aA
	5–10	8.0 \pm 0.3aA	27.9 \pm 3.0aA	9.0 \pm 1.7bA	860.6 \pm 20.5abA	516.5 \pm 38.8aA	2247 \pm 672bA	913 \pm 201aA	289 \pm 50aA	8986 \pm 3218aA	14378 \pm 374aA
	10–20	8.2 \pm 0.3aA	19.2 \pm 9.9abA	6.2 \pm 5.2bA	620.6 \pm 371.3bcA	496.0 \pm 28.0aA	1435 \pm 506bcA	582 \pm 24abA	157 \pm 12bA	9315 \pm 3153aA	12996 \pm 1307abA
	20–40	8.2 \pm 0.1aAB	11.6 \pm 3.1bA	2.0 \pm 0.7bA	211.1 \pm 62.6cAB	502.2 \pm 7.1aA	832 \pm 176cAB	714 \pm 180bA	148 \pm 5bA	11121 \pm 523aB	11312 \pm 1254bA

$$\text{NAP} = \frac{Q_{\max} \times k \times \text{EPC}_0}{1 + k \times \text{EPC}_0} \quad (4)$$

$$K_p = \frac{\text{NAP}}{\text{EPC}_0} \quad (5)$$

where Q_e (mg kg⁻¹) and Q_{\max} (mg kg⁻¹) are the equilibrium amount and the maximum amount of P sorbed onto sediment, respectively; C_0 (mg L⁻¹) is the equilibrium P concentration when the initial phosphorus in the solution was 0 mg L⁻¹; C_e (mg L⁻¹) is the equilibrium P concentration; k (L mg⁻¹) is the Langmuir equilibrium constant; MBC (L kg⁻¹) is the P maximum buffer capacity; NAP (mg kg⁻¹) is the native adsorbed P; EPC₀ (mg L⁻¹) is the zero equilibrium P concentration; and K_p is the distribution coefficient for a solid and liquid.

2.2.2. Assessing the risk of P release

The P sorption index (PSI) and degree of P saturation (DPS) are frequently used to characterize the P sorption capacity and potential risks of P release from sediments (Bache and Williams, 1971; Sekhon et al., 2014). The available soil P (Olsen-P) concentration in the sediments was extracted using a 0.5 mol L⁻¹ NaHCO₃ solution and subsequently determined colorimetrically. The DPS of the sediment was obtained according to the following equation (Jin et al., 2006):

$$\text{DPS} = \frac{\text{Olsen} - P}{\text{Olsen} - P + Q_{\max}} \times 100\% \quad (6)$$

2.3. The PSI was calculated as

$$\text{PSI} = \frac{X}{\lg C} \quad (7)$$

where PSI (L kg⁻¹) denotes P sorption index, X (mg kg⁻¹) is the amount of P sorbed and C (mg L⁻¹) is the equilibrium P concentration of filtrate after equilibration was reached between a 1 g sediment sample and 20 mL of a 75 mg P L⁻¹ solution in an end-over shaker for 24 h (Gao et al., 2018).

Huang et al. (2004) proposed a P eutrophication risk index (ERI), which is calculated on the basis of sediment PSI and DPS. ERI was then used to evaluate the risk posed by P in surface sediments. ERI was calculated using the method proposed by Huang et al. (2004), where

$$\text{ERI} = \frac{\text{DPS}}{\text{PSI}} \times 100\% \quad (8)$$

2.3.1. Sediment chemical analysis

The pH of the sediment was measured by a pH meter (soil to water ratio: 1:2.5); total P (TP) was determined using the molybdenum blue colorimetric method after digestion using a mixture of concentrated sulfuric acid-perchloric acid (Murphy and Riley, 1962). Total organic carbon (TOC) and total nitrogen (TN) were determined using an elemental analyzer (TOC-VCPH, Shimadzu). Amorphous iron (Fe_o) and aluminum (Al_o) oxides were extracted using ammonium oxalate, and their concentrations were determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES) (Bai et al., 2017). Calcium (Ca) and magnesium (Mg) were extracted using a 1 mol L⁻¹ hydrochloric acid solution; their concentrations were then determined using an atomic absorption spectrophotometer (TAS-990) (Pant and Reddy, 2001). The particle size distribution of the sediments was determined using a laser diffraction particle size analyzer (Mastersizer, 2000; Malvern, England). During the determinations of TOC, TN, and TP concentrations, a standard sample (GBW07427) was used for quality control. The measured error of the standard samples was less than 5%.

2.4. Statistical analysis

All data are reported as the mean and standard deviation of three

replicates. One-way ANOVA was carried out using the SPSS computer package (SPSS Inc. 1999; Chicago, USA), and significant differences between means were determined using an LSD test. Differences were considered statistically significant at the $P < 0.05$ level. A two-tailed Pearson correlation analysis was used to determine the correlations between P sorption parameters and physicochemical parameters in the sediment. The SigmaPlot 10.0 software package was used to fit isotherms to the phosphorus sorption data using modified Langmuir models.

3. Results

3.1. Physicochemical properties of Yellow River Delta sediments

The physicochemical properties of sediments collected from the three sampled wetlands within the YRD are shown in Table 1. The pH of the wetland sediments ranged from 7.6 to 8.5 and was slightly alkaline. Sediment pH did not change considerably with time since restoration and decreased with increasing depth. The TOC and TN contents in the sediments ranged between 1.1–22.8 mg g⁻¹ and 112.4–1643.7 mg kg⁻¹, respectively. Their concentrations gradually increased with increasing time since restoration ($R_{2002} > R_{2006} > R_0$), but decreased with increasing sediment depth. The clay content of the wetland sediments ranged from 5.8% to 32.7%. The amount of clay at sites R_{2002} and R_{2006} were higher than that in R_0 ; the clay content at all sites decreased with increasing depth. The TP content in the sediment ranged from 454.0 to 587.8 mg kg⁻¹. Significant changes in TP content were only observed between the surface (0–5 cm) and bottom layers (20–40 cm) of R_0 . The Fe_{ox} and Al_{ox} contents varied between 403–3845 mg kg⁻¹ and 190–1137 mg kg⁻¹, respectively. An increase in the Fe_{ox} concentration occurred in the surface layer (0–5 cm) at R_{2002} in comparison to R_{2006} or R_0 . Otherwise, Fe_{ox} contents did not vary significantly with restoration time. The Fe_{ox} content generally decreased with increasing sediment depth in each wetland, while Al_{ox} concentrations did not change significantly with depth. The Ca content (8429–13155 mg kg⁻¹) did not change significantly with the restoration time or with depth in the wetland sediments. The Mg content (7605–12996 mg kg⁻¹) in the sediment ranked in the order of $R_{2002} > R_{2006} \approx R_0$. The Mg content of the surface sediments (0–5 cm and 5–10 cm) at R_{2002} and R_0 were significantly higher than that of the bottom layers (20–40 cm) at the two plots.

3.2. Phosphorus sorption isotherm characteristics of wetland sediments

As is shown by the P sorption isotherm parameters ($R^2 > 0.94$) (Fig. 2, Table 2), the modified Langmuir model accurately described the sorption behavior of P in sediments of the YRD. The maximum P sorption (Q_{max}) values ranged between 201.8–1168.6 mg kg⁻¹ with an average of 576.2 mg kg⁻¹. Q_{max} values followed the order of $R_{2002} > R_{2006} > R_0$. The Q_{max} values gradually decreased with increasing sediment depth. The Q_{max} of the surface layer (0–5 cm) was significantly higher ($P < 0.05$) in comparison to those of the lower layers (10–20 cm and 20–40 cm). The k values varied between 0.040 and 0.290 L mg⁻¹, whereas the P maximum buffer capacity (MBC) ranged between 11.5 and 163.6 L kg⁻¹. The k and MBC values did not statistically change with time since restoration or with sediment depth.

The natural adsorbed P (NAP) in the sediments ranged between 0.009–0.375 mg kg⁻¹, whereas the EPC₀ ranged between 0.0006–0.0029 mg L⁻¹. Changes in NAP and EPC₀ values with restoration time were insignificant ($P > 0.05$). However, the NAP and EPC₀ values of the surface sediments (0–5 cm or 0–10 cm) were significantly higher ($P < 0.05$) than those of the lower (10–20 cm) and bottom sediments (20–40 cm).

3.3. Assessment of phosphorus sorption/release risk

The PSI of YRD sediments varied from 8.28–50.33 mg L/(100 g μmol) with an average of 26.65 mg L/(100 g μmol) (Fig. 3). Overall, the PSI of the surface sediments (0–5 cm and 5–10 cm) was larger than those of the lower (10–20 cm) and bottom sediments (20–40 cm). The PSI values varied in the order of $R_{2002} > R_{2006} > R_0$, which is consistent with the change in Q_{max} between sites. The DPS of the wetland sediments ranged from 0.27% to 2.07% and averaged 0.80%. DPS did not change significantly with time since restoration or depth (Fig. 3). The ERI value of the wetland sediments from YRD varied from 0.68% to 13.89% (Fig. 3). As the time since restoration increased, the ERI values generally followed the order of $R_{2002} < R_{2006} < R_0$ although differences among the three plots were not statistically significant ($P > 0.05$).

3.4. Correlation analysis between P sorption/release behavior and sediment characteristics

As shown in Table 3, there was a significant negative correlation between Q_{max} and pH ($P < 0.01$), and a significant positive correlation in the concentrations of Fe_{ox}, Al_{ox}, TOC, TN, and Mg, as well as the

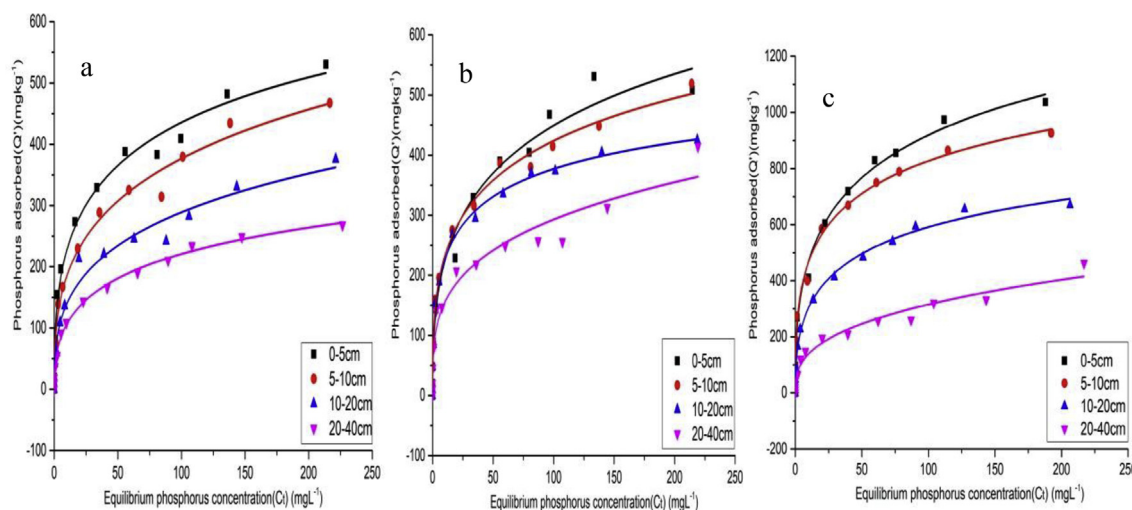


Fig. 2. Sorption isotherms of phosphorus in sediments from restored wetlands of differing age; a, b, and c refer to data from the control plot (R_0), wetlands restored since 2006 (R_{2006}), and wetlands restored since 2002 (R_{2002}), respectively.

Table 2

Characteristic values of isothermal sorption of the Langmuir model for different sediments. The data in the tables are the mean \pm standard deviation. The different lowercase letters indicate significant differences among data of different depths from the same column, and different uppercase letters indicate significant differences among data of different restoration times from the same sediment depth.

	Q_{\max} (mg·kg ⁻¹)	k (L mg ⁻¹)	MBC (L kg ⁻¹)	R ²	NAP (mg·kg ⁻¹)	EPC ₀ (mg·L ⁻¹)	K _p (L kg ⁻¹)
R ₀ (0–5)	494.1 \pm 8.6 aB	0.113 \pm 0.054 aA	55.8 \pm 27.9 aA	0.96	0.114 \pm 0.044 aA	0.0021 \pm 0.0003 aA	55.8 \pm 27.9 aA
R ₀ (5–10)	455.7 \pm 189.3 aB	0.086 \pm 0.048 aA	34.6 \pm 5.6 aB	0.96	0.041 \pm 0.007 bB	0.0012 \pm 0.0000 bA	34.6 \pm 5.6 aB
R ₀ (10–20)	333.4 \pm 121.4 aA	0.086 \pm 0.028 aA	26.9 \pm 1.2 aA	0.94	0.021 \pm 0.004 bA	0.0007 \pm 0.0001 bA	26.9 \pm 1.2 aA
R ₀ (20–40)	259.4 \pm 81.5 aA	0.071 \pm 0.012 aA	17.8 \pm 2.6 aA	0.96	0.015 \pm 0.005 bA	0.0008 \pm 0.0001 bA	17.8 \pm 2.6 aA
R ₂₀₀₆ (0–5)	589.2 \pm 220.5 aB	0.141 \pm 0.047 aA	88.1 \pm 55.6 aA	0.95	0.230 \pm 0.134 aA	0.0027 \pm 0.0002 aA	88.1 \pm 55.6 aA
R ₂₀₀₆ (5–10)	567.4 \pm 216.5 aB	0.162 \pm 0.044 aA	86.1 \pm 19.7 aA	0.94	0.138 \pm 0.061 aB	0.0016 \pm 0.0005 bA	86.1 \pm 19.7 aA
R ₂₀₀₆ (10–20)	548.5 \pm 314.5 aA	0.210 \pm 0.089 aA	98.6 \pm 24.3 aA	0.96	0.105 \pm 0.049 bA	0.0010 \pm 0.0003 cA	98.6 \pm 24.3 aA
R ₂₀₀₆ (20–40)	384.8 \pm 93.3 aA	0.146 \pm 0.097 aA	49.4 \pm 26.6 aA	0.95	0.038 \pm 0.020 bA	0.0008 \pm 0.0001 cA	49.4 \pm 26.6 aA
R ₂₀₀₂ (0–5)	998.0 \pm 198.1 aA	0.085 \pm 0.050 aA	78.0 \pm 30.5 aA	0.96	0.173 \pm 0.080 aA	0.0022 \pm 0.0004 aA	78.0 \pm 30.5 aA
R ₂₀₀₂ (5–10)	943.0 \pm 153.2 aA	0.110 \pm 0.021 aA	102.1 \pm 10.0 aA	0.95	0.157 \pm 0.010 aA	0.0015 \pm 0.0001 bA	102.1 \pm 10.0 aA
R ₂₀₀₂ (10–20)	669.7 \pm 237.0 aB	0.151 \pm 0.065 aA	103.7 \pm 58.9 aA	0.95	0.107 \pm 0.055 aB	0.0011 \pm 0.0003 bC	103.7 \pm 58.9 aA
R ₂₀₀₂ (20–40)	444.5 \pm 188.5 bA	0.084 \pm 0.039 aA	33.5 \pm 1.5 aA	0.94	0.031 \pm 0.005 bA	0.0009 \pm 0.0002 cA	33.5 \pm 1.5 aA

percentage of clay particles in the sample ($P < 0.01$). ERI exhibited the opposite pattern. NAP, EPC₀ and K_p exhibited a significant negative correlation with pH ($P < 0.01$), and a significant positive correlation in the contents of Fe_{ox}, Al_{ox}, TOC, TN, Mg and clay particles ($P < 0.01$).

4. Discussion

4.1. Factors affecting phosphorus sorption and release

Phosphorus sorption by wetland sediments is the predominant mechanism through which P is removed from water (Reddy et al., 1999). Previous studies have shown that P sorption by sediments is controlled by several parameters, including the type and content of clay particles, organic matter content, Fe and Al oxide concentrations, Ca and Mg concentrations, and sediment pH (Antoniadis et al., 2016). Generally, for sediments with a pH < 6.5 , amorphous Fe and Al oxides determine P sorption; however, for sediments with a pH > 7.5 , Ca and Mg species tend to dictate P sorption (DeLaune and Reddy, 2008). The pH of wetland sediments in the YRD was slightly alkaline (pH > 7.5) (Table 1). The correlation analysis suggested that Fe_{ox}, Mg, and Al_{ox} are the main regulators of P sorption, while the sorption capacity of P by Ca species was not pronounced. This is consistent with previous studies in which the Ca binding capacity for P was smaller than that for Fe/Al oxides in slightly alkaline sediments (pH < 9) (Reddy et al., 1999). The lower degree to which P was sorbed to Ca was likely due to the longer time needed for Ca to bind with P (Barrow, 1978; Bai et al., 2017). Both the calcium sorption of P and the precipitation of P increase with increasing pH in alkaline sediments (Ann et al., 1999). If the

binding of P to Ca dominates P sorption, a positive correlation between Q_{\max} and pH is expected. However, the significant negative correlation between Q_{\max} and pH ($P < 0.01$) observed herein indicated that the sorption of P to Ca in the YRD was negligible. There are studies that suggest that Ca species regulate P sorption in alkaline soil (Bertrand et al., 2003; Ige et al., 2005a,b). Ige et al. (2005a,b), for example, suggested that Ca and Mg concentrations, extracted using the Mehlich 3 solution, could be used to characterize the P sorption capacity of calcareous soils. In addition, more than 78% of the P in the wetland sediments of the YRD is Ca phosphate (Xu et al., 2012). Therefore, the long-term P retention by Ca species in the YRD is unclear and requires further study.

Organic matter is also an important factor affecting P sorption, but the sorption mechanism is relatively complicated. Organic matter can absorb P through metal bridging (DeLaune and Reddy, 2008). On the other hand, organic matter including low-molecular-weight carboxylic acids can reduce P sorption capacity by competing with P for binding sites. Organic matter may also inhibit the crystallization of Fe and Al oxides by complexation, decreasing the total sorption capacity for P (Guppy et al., 2005). The wetland restoration projects in the YRD significantly increased organic matter content thereby increasing P sorption through metal bridging (Cui et al., 2009). The P-fixation capacity of sediments is also related to clay particle content. A higher content of clay particles leads to a larger specific surface area of the sediment, increasing the P-fixation capacity (Stumm, 1992).

The DPS of the sediments is an indicator of the percentage of P adsorbed by sediment to the total P sorption capacity. It can therefore be used to evaluate the P sorption capacity of sediments and assess the risk of losing P by leaching and runoff (Sekhon et al., 2014). It is

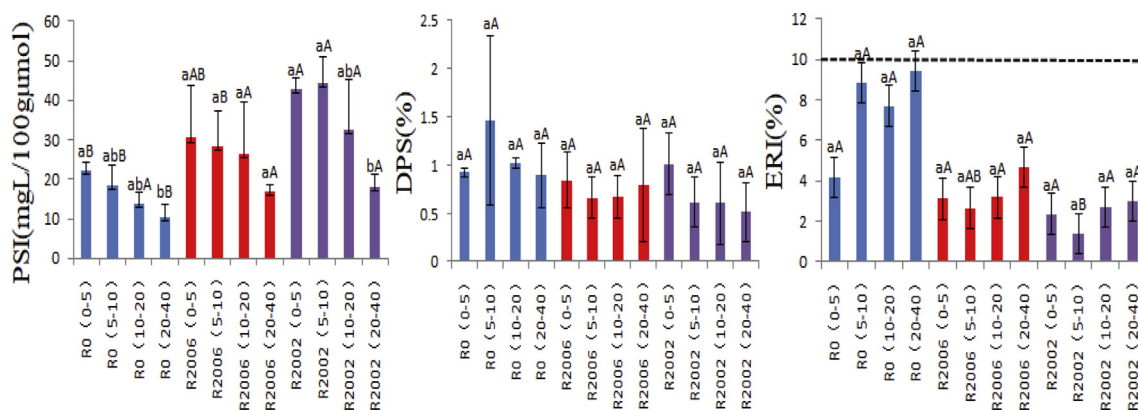


Fig. 3. Distributions of phosphorus PSI, DPS and ERI determined for different restored wetlands in the YRD. Data in the figure represent means of three replicates (\pm SD). Different uppercase letters on error bars indicate significant differences at $P < 0.05$ for different restoration time. Different lowercase letters on error bars indicate significant differences at $P < 0.05$ for different sediment depth. The dashed line indicates low risk (ERI < 10) values of P from sediments.

Table 3

Correlation analysis between phosphorus sorption/release and physicochemical parameters in sediment samples from the YRD.

	pH	Feox	Alox	TOC	TP	Clay particles	HCl-Ca	HCl-Mg
Q _{max}	−440 ^b	0.805 ^b	0.488 ^b	0.663 ^b	0.174	0.871 ^b	−0.280	0.702 ^b
DPS	0.158	0.029	−0.208	−0.013	−0.039	−0.102	0.164	−0.187
ERI	0.458 ^b	−0.466 ^b	−0.523 ^b	−0.447 ^b	−0.289	−0.523 ^b	0.258	−0.527 ^b
K	−0.122	−0.074	0.145	0.036	0.126	0.012	−0.080	−0.078
NAP	−0.640 ^b	0.622 ^b	0.571 ^b	0.876 ^b	0.394 ^a	0.549 ^b	−0.394 ^a	0.344 ^a
EPC ₀	−0.564 ^b	0.601 ^b	0.430 ^a	0.702 ^b	0.386 ^a	0.327	−0.369 ^a	0.158
K _p	−0.514 ^a	0.485 ^b	0.518 ^b	0.584 ^b	0.194	0.631 ^b	−0.360 ^a	0.464 ^b
MBC	−0.514 ^a	0.485 ^b	0.518 ^b	0.584 ^b	0.194	0.631 ^b	−0.360 ^a	0.464 ^b

^a Significantly correlated at the 0.05 level.^b Significantly correlated at the 0.01 level (two-tailed test).

generally believed that low DPS values indicate that P sorption sites in the sediment are not saturated, and the sediment will have a relatively high P sorption capacity (Nair, 2014). The DPS of the sediments in the YRD ranges between 0.27–2.07%, which is lower than that in the Okeechobee wetland of Florida, USA (5–18%) (Dunne et al., 2006), and the Minjiang River Estuary in China (7.3%) (Gao et al., 2018). It is also considerably lower than those of inland reservoirs, such as the Beijing urban reservoirs in China (7.97–50.5%) (Lu et al., 2011) and the Taihu reservoirs in China (3.8–15.3%) (Jin et al., 2006). The relatively low DPS values indicate that the wetlands of the YRD have a greater potential to adsorb or remove P from water.

The EPC₀ of sediments is an important indicator for determining the P adsorbed by, or released from, sediments. When the concentration of soluble P in the overlying water is > EPC₀, the sediment will adsorb P from the water body, and the sediment will act as a P sink. Otherwise, P will likely be released to the water body, and the sediment will serve as a P source. The EPC₀ in the present study ranged from 0.0006 to 0.0029 mg L^{−1}, which was lower than the P concentration of the overlying water (0.016–0.126 mg L^{−1}). Therefore, the wetland sediments within the YRD are likely to serve as a P sink and have a high potential to remove P from surface water. The relatively lower DPS and EPC₀ values maybe related to the relatively low P loads (see details in section 4.3) and to limited soil development due to young age of the sediments within the YRD (Bai et al., 2019a,b).

ERI can be used to subdivide the risk of eutrophication induced by P into four categories: very high risk (ERI > 25), high risk (20 < ERI < 25), moderate risk (10 < ERI < 20) and low risk (ERI < 10) (Huang et al., 2004). The ERI of the YRD varied between 0.68% and 13.89%. These values are similar to those of the wetlands in the Minjiang River Estuary in China (9.6–10.6%) (Gao et al., 2018), but are much lower than those of inland wetlands, such as the Beijing urban reservoirs in China (17.2–247.2%) (Lu et al., 2011) and the Taihu reservoirs in China (10–55%) (Jin et al., 2006). These ERI values indicate that the risk of P release from sediments in the YRD is generally low, which is consistent with the fact that the YRD has not experienced large-scale eutrophication (Bai et al., 2012).

4.2. Comparison of wetlands in the Yellow River Delta with other wetlands

The YRD wetlands exhibited TP, Q_{max}, and K_p values, and thus a P sorption potential, similar to other coastal wetlands (Table 4). However, k, NAP, and EPC₀ values were significantly lower than in other coastal wetlands. The parameter k is the half-saturation concentration and contrasts with the surface binding energy. The significantly lower k suggests that the wetland sediments in the YRD had a higher affinity for P than the sediments in other coastal wetlands (Zhou et al., 2005). The higher affinity for P maybe explained by the young age of the sediments within the YRD because the binding energy constant is controlled by strong sorption sites during the early stage of P sorption (Lin et al., 2009). NAP, which is also an indicator of sediment exchangeable P, tends to be in equilibrium with P in the water body and is therefore a

measure of the degree of P water pollution (Wang et al., 2005; Jin et al., 2006). The NAP value of the YRD wetlands (0.009–0.375 mg kg^{−1}) was substantially lower than that of the Hangzhou Bay tidal flats (1.853–4.777 mg kg^{−1}) (X. Shao et al., 2014) and wetlands within the Yangtze River Estuary (1.23–36.05 mg kg^{−1}) (Liu et al., 2002), indicating that only a small amount of P is being desorbed from sediments in YRD wetland. The EPC₀ of the YRD wetlands was 1–2 orders of magnitude lower than those of other coastal wetlands, suggesting that the YRD wetlands possess a larger potential to remove P from the water.

Compared to coastal wetlands, more than 66% of inland reservoirs exhibit eutrophic conditions and 22% of all water bodies are hyper-eutrophic in China (Zhang et al., 2008). The rank order of P dynamics in sediments was consistent with the trophic status of inland reservoirs. As a result, a comparison between different polluted inland and coastal wetlands may provide insights into P storage and release from coastal wetlands. Coastal wetlands exhibited similar TP, Q_{max}, k, and EPC₀ values to inland wetlands (Table 4). However, the NAP values of coastal wetlands were generally 1–2 orders of magnitude lower than the inland reservoirs (Table 4). The NAP value is positively correlated with the degree of eutrophication (Wang et al., 2005; Jin et al., 2006). For example, the sediment NAP values in eutrophic and moderately eutrophic areas of the Taihu were 44–380 mg kg^{−1} and 12–15 mg kg^{−1}, respectively (Zhou et al., 2005). The K_p value reflects the affinity of P for sediments (Zhou et al., 2005). The K_p value of the coastal wetlands tended to be lower than that of the inland reservoirs (Table 4). That is, P in coastal wetlands is more easily stored in the water than in sediment (Wang and Pant, 2010).

The YRD was formed slightly more than 100 year ago since the watercourse of the Yellow River changed in 1855. Compared to the long-term formation of other coastal wetlands (such as Yangtze River delta) (Saito et al., 2001), the YRD is the youngest coastal wetland in China, and is characterized by low levels of soil development (Yu et al., 2016; Bai et al., 2017). Progressive soil development has proved to increase P sorption capacity due to organic matter accumulation (Zou et al., 2011). In other words, the restored YRD sediments show great potential for P sorption capacity while low risk of P release to the adjacent ocean.

4.3. Significance of wetland restoration to the phosphorus cycle

Wetland restoration in the YRD has been conducted by introducing water into wetlands, which resulted in an increase in plant growth and coverage, an increase in biodiversity (e.g., benthic organisms and birds) and improvements in ecosystem health (Cui et al., 2009). This study showed that with increasing restoration time, sediment Q_{max} increased, indicating that wetland restoration can increase the P sorption capacity of wetland sediments (Fig. 4). Values of MBC, NAP, and EPC₀ did not significantly change with time since restoration, nor was there a significant increase in wetland DPS and ERI (Fig. 4). These results demonstrate that wetland restoration did not substantially increase the potential risk of P loss from the sediments. This conclusion is in contrast

Table 4
Comparison of phosphorus properties between coastal wetlands and inland reservoirs.

	Area	TP (mg·kg ⁻¹)	Q _{max} (mg·kg ⁻¹)	K (mg·L ⁻¹)	NAP (mg·kg ⁻¹)	EPC ₀ (mg·L ⁻¹)	K _p (L·kg ⁻¹)	Source/sink	Reference
Coastal wetland	YRD wetland	458.9–548.7	201.8–1168.6	0.040–0.290	0.009–0.375	0.0006–0.0029	11.5–163.6	Sink	This study
	Hangzhou Bay wetland	520–680	154.5–436.2	2.611–4.542	1.853–4.777	0.034–0.049	41.2–140.5	Sink	Shao et al. (2014)
	Yangtze River Estuary Wetland	656.8–1351	26.3–204.1	0.76–1.71	1.23–36.05	0.057–0.418	21.5–248.3	Sink	Liu et al. (2002)
	Indian River Lagoon system	58–942	31.2–500	0.15–6.43	–	0.01–0.19	15.2–346.6	–	Pant and Reddy (2001)
Inland reservoirs	Shihwa marsh	229.1–541.9	155–260	1.49–1.51	–	0.06–0.26	–	Sink	Yoo et al. (2006)
	West Lake	210–1480	610–1130	–	4–93	0.038–0.193	105.2–755.1	Source	Shao et al. (2014)
	Taihu Lake	420.2–809.2	307.7–798.3	0.59–1.986	18.34–46.28	0.033–0.143	323.23–625.12	Source	Wang et al. (2005)
	Dianchi Lake	1596.3–5558.5	535.7–41466.4	0.04–2.08	7.74–403.8	0.00–0.03	568.3–24917.8	Sink	Chen et al. (2014)
	Lake Okeechobee		65.4	49.9	16.18	4.37	3.69	Source	(Wang et al., 2010)

with conventionally constructed wetlands: the longer the wetland operation time, the higher the DPS of the wetland. Thus, the P sorption capacity of the wetland decreases with increasing wetland operation time (Craft, 1996). However, the restored YRD wetlands are natural wetlands that are significantly different from artificially constructed wetlands in three aspects. First, the external P load to the YRD wetlands is significantly lower than that of constructed wetlands. The external P load was calculated to be 8–63 mg m⁻² a⁻¹ on the basis of water inflows and P concentration of the water (Cui et al., 2009). In comparison, the external P load from an artificially constructed wetland ranged between 45–75 g m⁻² a⁻¹. (Vymazal, 2007; Pietro and Ivanoff, 2015). Second, vegetation started to colonize quickly after wetland restoration and the newly emerged macrophyte communities significantly absorbed P from the wetlands. The vegetation in the restored wetlands is collected every year, thus significantly decreasing P load to the wetlands (Cui et al., 2009). Finally, the wetland restoration increased plant growth and thereby enhanced organic matter content. In association with organic matter, microorganism will increase, along with soil development. The clay, Mg and surface Fe_{ox} contents also increase with soil development (Table 1). The combined effects of these processes is that the P retention capacity in the YRD significantly increased with the time since restoration, which is contrary to the hypothesis of decreasing storage of P with increasing restoration time (as found for constructed wetlands). However, it is also necessary to pay attention to the changes in P load and risk of P release after the long-term operation of restored natural wetlands to reduce the risk of water eutrophication.

As for coastal wetland restoration, we recommended bringing freshwater into degrade wetlands to improve plant growth. After initiating the plant communities, it is necessary to remove plants from the wetlands to obtain a higher efficiency of nutrient retention. In conjunction with the plant communities, soil development will enhance P storage.

5. Conclusions

This study showed that P storage capacity increased with increasing time since wetland restoration while the risk of losing (releasing) P from the sediments decreased with increasing time in the YRD. Therefore, coastal wetlands restoration is recommended for P reduction within the coastal zone.

The YRD is located at the intersection of land and sea. The upper reaches of the Yellow River have increased the P load to the wetlands, and ultimately to the Bohai Sea. The offshore water is affected by extra P load, leading to the occurrence of eutrophication of the Bohai Sea. In July 2018, the Ministry of Ecology and Environment of China launched the Action Plan for the Comprehensive Management of the Bohai Sea. The focus of the plan is to carry out the control of land-based pollution, and large rivers (such as the Yellow River) are key pathways through which terrestrial pollutants enter the sea. There are large areas of original and restored wetlands in the Yellow River estuary. We recommended that coastal wetlands be utilized to reduce upstream P pollution to the Bohai Sea to maintain a P balance in the land-wetland-offshore watershed.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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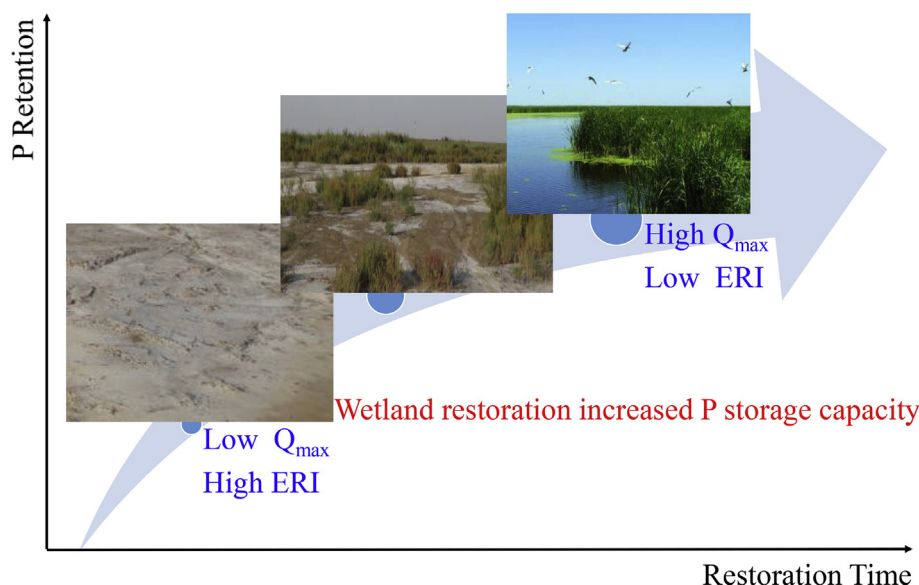


Fig. 4. Enhanced phosphorus storage capacity of sediments by coastal wetland restoration in Yellow River Delta, China. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

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