



Phosphorus release from coastal sediments: Impacts of the oxidation-reduction potential and sulfide



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ABSTRACT

The release of phosphorus (P) from benthic sediments can affect the P content, nutrient status and quality of overlying waters in coastal ecosystem. This study was carried out to investigate the influence of oxidation-reduction potential (ORP) and sulfide on P release from sediments in the coastal estuary of the Yuniao River, China. The results showed that ferric iron-bound P was the main P burial phase in the sediments. The P concentration in overlying water increased with ORP decrease and sulfide increase, displaying a significant linear correlation with the ORP and sulfide concentration. The results indicate that decreased ORP may elevate the zero equilibrium phosphorus concentration, enhancing the capability of P release. And increased sulfide may react or capture reactive iron in sediments, reducing the P adsorption capacity and accelerating P release. Therefore, the control of ORP and sulfide production is important in the sink/source conversion of P in coastal sediments.

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1. Introduction

Phosphorus (P) is an essential nutrient for biological productivity, and therefore plays an important role in oceans and continents. The total input of P into aquatic systems is determined by both external inputs as well as internal recycling from the benthic sediments to the water column (Pan et al., 2013; Kraal et al., 2015). Excessive input of P can lead to eutrophication, which adversely affects coastal zone ecosystems in many areas around the world (Yang et al., 2015). Phosphorus retention and release processes in estuarine and coastal sediments are also significant for the global P cycle because a great portion of P (~25% dissolved P) is trapped in such sediments and thus does not reach the open ocean (Ruttenberg, 1993; Paytan and McLaughlin, 2007).

Under some conditions, benthic sediment may act as an important P source for the overlying water (Sondergaard et al., 1999), and even create N-limited condition (Ferrera et al., 2016). P exchange processes between the sediment and overlying water are known to be affected by physico-chemical conditions and biological processes, which can also influence the existing forms of P (Qian et al., 2011; Pan et al., 2013). Anoxic marine sediments are generally thought to have diminished sedimentary P retention capacity (Ingall et al., 1993). Recent work has provided detailed insight into the relationship between reducing conditions and diminished sedimentary P sequestration (Kraal et al., 2015). Previous studies report that P flux between sediment and overlying

water may be strongly influenced by physico-chemical conditions, such as temperature, dissolved oxygen (DO) and pH (Smith et al., 2006; Lake et al., 2007; Berbel et al., 2015). In particular, oxidation-reduction potential (ORP) may be an especially crucial factor controlling the release of P to the overlying water from sediments. In this regard, P-sediment adsorption experiments may be useful in examining the ORP-dependent ability of sediment to either remove or release P to the overlying water (Lyons et al., 1998; Pan et al., 2002).

The P cycle in coastal ecosystem can be strongly coupled to sediment iron (Fe) and sulfur (S) transformations (Kraal et al., 2015). Understanding the interaction between Fe, P and S is key to predicting their biogeochemical fates and impact on water quality. In estuarine sediments rich in organic matter, oxygen is quickly depleted with microbial aerobic respiration below the sediment-water interface, thereby creating an anoxic environment. Under these conditions, both microbial iron reduction (Taillefert et al., 2000a) and sulfate reduction will occur (Burton et al., 2011). Iron reduction results in the dissolution of iron oxides to ferrous iron, whereas sulfate reduction results in the generation of sulfide. In the presence of sulfide, the Fe²⁺ ion can be quickly and effectively removed from pore waters by the formation of FeS and FeS₂, which also removes the toxic sulfide in the sediment-water system (Taillefert et al., 2000b). The biogeochemical reactions of Fe and S also affect both availability of sedimentary P to aquatic organisms and mobility of P within the sediments (Rozan et al., 2002).

In light of the global increase in the areal extent of coastal marine systems that experience oxygen depletion and alterations in benthic P cycling in response to eutrophication, it is of considerable interest to clarify the ORP-dependent P release processes (Diaz and Rosenberg,

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2008; Middelburg and Levin, 2009; Qian et al., 2011). Therefore, the goals of this study were 1) to investigate the influence of ORP on P release from sediments in a polluted coastal estuary using a combination of P adsorption experiments and selective extractions across an experimentally-controlled range of ORP conditions; and 2) to investigate the influence of sulfide addition on P release from sediments.

2. Methods and materials

2.1. Sampling and handling

The Yuniao River originates from the Muping City (Yantai, China) urban area (0.4 million population), and carries about 60% of the municipal sewage of the city. The river is located in the northeastern Shandong province in northern China on the Bohai Strait and the southern coast of the Northern Yellow Sea ($37^{\circ}23'35''\text{N}$, $121^{\circ}33'59''\text{E}$; Fig. 1). The river is a typical seasonal river, with domestic and industrial wastewater discharge accounting for most of the river water during the dry season (Sun et al., 2016). The tide in the river estuary is a regular semidiurnal tide, and therefore the water quality is variable (Li et al., 2016). Sediment samples were collected from the Yuniaohe River estuary using a Van-Veen grab sampler, and were stored in zip-lock plastic bags on ice in an adiabatic box. Before analysis, the samples were freeze-dried and sieved (80 mesh). All reagents used were analytical grade or above, and deionized water (Milli-Q) was used to prepare the overlying water. Milli-Q water was deoxygenated by purging with high-purity N_2 (>30 min).

2.2. Measurements and methods

Concentrations of P, Fe (digestion prior to determination) and chemical oxygen demand (COD_{Cr} , acid COD_{Mn} and alkaline COD_{Mn}) were measured with the corresponding method specified in the standard methods by APHA (2005). In brief, this involved the dichromate titration method for COD_{Cr} , acid and alkaline permanganate titration method for acid COD_{Mn} and alkaline COD_{Mn} respectively. Dissolved oxygen (DO) and ORP in overlying water were measured using Unisense meters and electrodes. Five solid-phase fractions of P (loosely sorbed P, E&L-P; ferric iron-bound P, Fe-P;

authigenic carbonate fluorapatite + biogenic apatite + CaCO_3 -associated P, ACA-P; detrital apatite P, De-P; and organic P, Or-P) in the sediment samples were determined by a sequential extraction method (SEDEX) (Ruttenberg, 1992). The concentrations of total nitrogen (TN), total sulfur (TS) and total organic carbon (TOC) in sediments were measured by an elemental analyzer (Vario MACRO cube, Elementar, Germany). The precision of the measurements was $\leq 0.5\%$.

2.3. ORP-dependent P release experiments

A group of experiments were set up to investigate the impact of the ORP on P release. In an anaerobic chamber, freeze-dried sediment was added to 6 beakers (named a, b, c, d, e, f) with anaerobic deionized water to achieve a sediment concentration of 10 g L^{-1} in each beaker. After allowing the sediment-water system to stabilize for 24 h, the DO and ORP were then measured. At the same time, a 5 mL sample of the supernatant water was collected from each beaker without disturbing the sediment-water interface to measure the concentration of P. Then, different dosages of NaBH_4 were added to achieve different ORP values in each beaker. 24 h later, DO, ORP and P were again measured.

2.4. Adsorption isotherm experiments

To determine the zero equilibrium phosphate concentration (EPC_0) for sediment samples under different ORP conditions, 0.20 g of freeze-dried sediment was placed in 48 polypropylene centrifuge tubes with 20 mL anaerobic deionized water. The 48 tubes were divided into 6 groups (A–F, with different ORP values), with each group containing 8 tubes. Each group was spiked with KH_2PO_4 to provide a range of initial phosphorus concentrations (0, 0.5, 1.0, 2.0, 6.0, 10.0, 15.0, 20.0 mg L^{-1}). The tubes were placed in a shaking bath (150 rpm, 25°C) for 24 h. After 24 h, each tube was centrifuged (3000 rpm), then aqueous P was measured after filtration to $<0.45\text{-}\mu\text{m}$. The difference in P between the start and the end of the sorption experiment, along with the sediment dosage were used to calculate the amount of adsorbed P. Sorption curves were obtained by plotting the aqueous equilibrium concentration of P against the amount of adsorbed P per kg sediment.

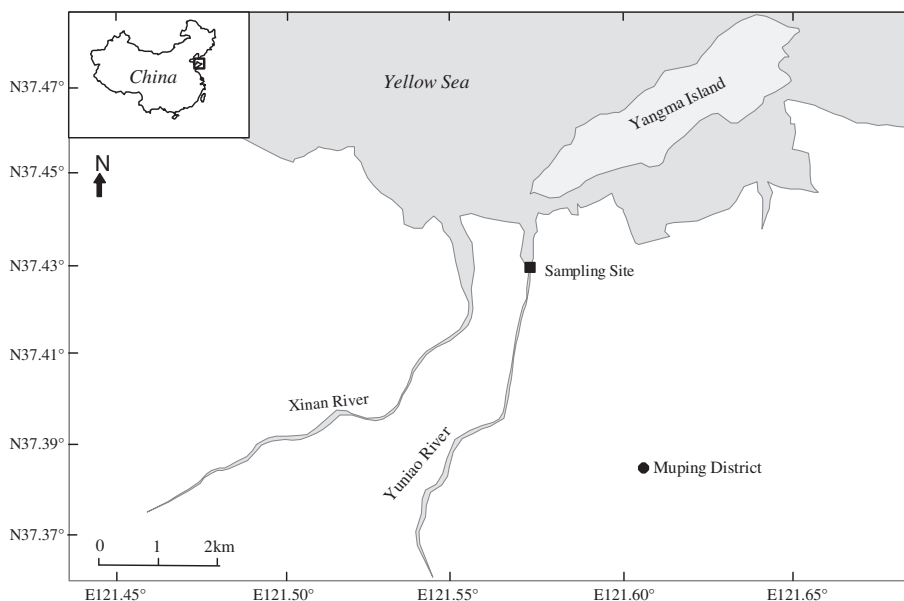


Fig. 1. Detailed location of the sampling site.

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