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Riverine flux of dissolved phosphorus to the coastal sea may be overestimated, especially in estuaries of gated rivers: Implications of phosphorus adsorption/desorption on suspended sediments

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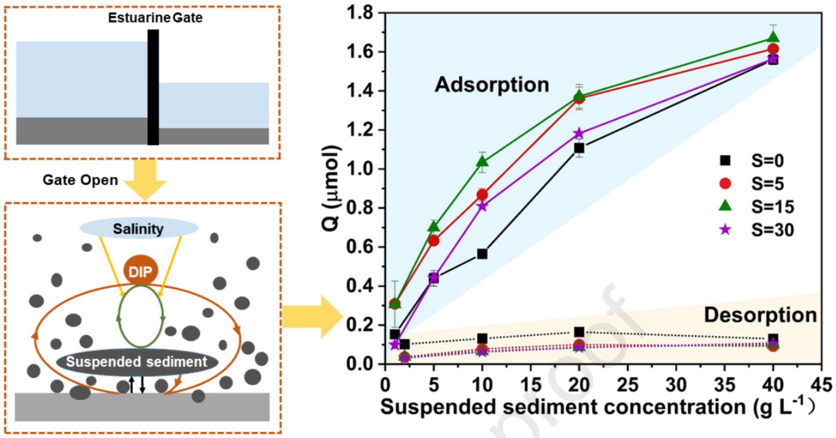
Estuarine Gate
Gate Open
Salinity
DIP
Suspended sediment
Adsorption
Desorption
S=0
S=5
S=15
S=30
Q (μmol)
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
0
5
10
15
20
25
30
35
40
45
Suspended sediment concentration (g L^{-1})

Abstract

The flux of terrestrial dissolved inorganic phosphorous (DIP, i.e. PO_4^{3-}) via rivers into coastal seas is usually calculated by simply multiplying its concentration with the corresponding water flow at the river mouth. Subsequent adsorption/desorption of DIP onto suspended sediment and the influence of salinity in the estuary are often overlooked. A series of DIP adsorption/desorption experiments under different salinities (0, 5, 15, 30) and suspended sediment concentrations (1-40 g L^{-1}) were conducted in order to assess the potential influence of these factors on the overall DIP loading to the coastal zone. The effect of different sea-salt ions on DIP adsorption/desorption was also assessed by comparing different experimental solutions (NaCl solution, artificial seawater and real seawater). In estuaries, the adsorption of DIP to suspended sediments was greater than desorption, and the net adsorption increased with increasing concentration of suspended sediments and salinity. This enhanced DIP adsorption onto suspended sediment reduces the riverine discharge of DIP to coastal ecosystems. Disregarding this process, especially for the gated estuaries with high sediment resuspension, potentially leads to an overestimation of the terrestrial DIP input to the coastal region.

Key words: seaward dissolved inorganic P flux; adsorption/desorption; suspended sediment; salinity

1. Introduction

Anthropogenic pressure on estuarine and coastal environments is increasing, due to the growing population along the coast (CIESIN, 2018). Coastal eutrophication is one of the most severe and wide-spread environmental problems, primarily caused by intensified influx of riverine nutrients (phosphorus and nitrogen) (Ballagh et al., 2019). Hence, quantification of nutrient inputs from land to sea is important for understanding and managing coastal eutrophication.

Typically, the nutrient flux to the sea is calculated by multiplying nutrient concentrations at the river mouth with corresponding water flow. However, this approach neglects the important physicochemical processes taking place in the estuarine systems, such as adsorption and desorption of nutrients on suspended sediment. These processes are highly affected by changing physicochemical conditions along the estuarine-coastal gradient. The calculated nutrient flux may therefore not represent an “effective” input to the coastal zone. These processes must therefore be taken into account when estimating the riverine loading of a pollutant, especially regarding particle-reactive chemical species such as phosphate (PO_4^{3-}). This is particularly the case for estuaries with river inflow controlled by gates, signified by pulsed flow patterns and high sediment resuspension. The complex chemical and hydrodynamic environment in estuaries fed by gated rivers affects the P cycling processes and thereby the DIP flux to the sea (Wang et al., 2010; Zhang et al., 2019a; Meng et al., 2014a). Although the total influx from gated rivers is lower than that from rivers without gates or dams, the increased hydrodynamic turbulence during discharge pulses enhances sediment resuspension, especially during the flood season (Duan et al., 2008; Lehner et al., 2011; Zarfl et al., 2015; Eiriksdottir et al., 2017; Ding et al., 2019). According to field investigation and other studies, the highest measured sediment concentration in gated rivers or the maximum estuarine turbidity zones is around 40 g L-1 (Uncles et al., 2006; Snedden et al., 2007; He et al., 2010; Yao et al., 2016; Palinkas et al., 2019).

Phosphorus (P) is an essential element for phytoplankton growth and DIP typically limits primary production during spring in estuaries receiving high inputs of nitrogen (Ruttenberg, 1992; Ballagh et al., 2019). Rivers are the dominant source of P to coastal seas (Benitez-Nelson, 2000). Accurate estimates of the riverine flux of bioavailable P (i.e., mainly dissolved inorganic phosphate, DIP) is a prerequisite for selecting appropriate abatement strategies to ensure a sustainable management of the coastal environment (Ding et al., 2019; Palinkas et al., 2019; Liu et al., 2020).

The transport and transformation of P through an estuary is governed by a complex set of processes. In addition to a large number of environmental factors in the water column affecting DIP concentrations, processes at the sediment-water interface also influence P transformations and thereby the P flux (Wang et al., 2012; Kang et al., 2018; Rapin et al., 2019). For example, oscillations in organic matter content and redox enhance P release from sediment (Wang et al., 2012; Kang et al., 2018), while submerged macrophytes will enhance DIP uptake (Wang et al., 2012). In estuaries, the interaction between DIP in water and suspended sediments is a key process controlling bioavailable P. Suspended sediments can efficiently scavenge DIP from the water column through adsorption, thereby decreasing the DIP flux to the sea (Millero et al., 1986; Millero et al., 2001; Liu et al., 2002; Egger et al., 2015). On the other hand, DIP released from the sediment via desorption may subsequently increase the DIP flux to the sea. For gated estuaries, it remains unclear whether enhanced sediment resuspension actually increases or decreases the net seaward DIP flux.

Adsorption of DIP onto suspended sediment is influenced by salinity, temperature and pH, as well as the characteristics of the sediments such as texture and organic matter content (Jones et al., 1993; Sundareshwar et al., 1999; Cao et al., 2011; Zhang et al., 2011; Zhou et al., 2018). Among these factors, salinity is the most prevalent factor affecting the adsorption process in estuaries, due to the large gradients in salinity from fresh to seawater. The estuarine gradients in temperature, organic matter and pH are generally smaller and to a large extent controlled by salinity (Jun et al., 2013; Flower et al., 2017; Paudel et al., 2019). The fact that salinity enhances phosphate adsorption implies that the phosphate oxyanion is bound to active sites via inner-sphere (ligand exchange) complexation. Electrostatic outer-sphere interactions between phosphate and suspended sediments are weakened by increased salinity due to competition for active sites by electrolyte anions, such as chloride (Bai et al., 2017; Huang et al., 2015). Moreover, the inner-sphere complexation is facilitated by the high sodium ion (Na^+) concentration weakening the electrostatic outer-sphere interactions through lowering of particle net negative surface charge (Zhang et al., 2019b). On the other hand, increased salinity also promotes particle flocculation (Asmala et al., 2014; Bai et al., 2017), which decreases the total surface area of the suspended particles and thereby their adsorption capacity.

Solutions of sodium chloride (NaCl) are commonly used instead of real seawater to analyze salinity effects on DIP adsorption (Wang et al., 2006; Wu et al., 2012; Bai et al., 2017; Zhang et al., 2019b; also see Fig. S1). In addition to sodium (Na^+) and chloride (Cl^-), seawater contains significant concentrations of calcium (Ca^{2+}), magnesium (Mg^{2+}), sulfate (SO_4^{2-}), and bicarbonate (HCO_3^-) (Riebesell et al., 2010). The influence of these additional ions on the DIP adsorption differs from that of Na^+ and Cl^- (Flower et al., 2017). Therefore, it is of special importance to assess the specific ionic effect on the estimates of the DIP seaward flux. This is especially a concern in estuaries where the load of suspended sediments is high, such as from rivers with gates.

This paper aims to evaluate the influence of suspended sediments on the seaward DIP flux, based on experiments for assessing the effect of salinity, ionic composition and sediment concentration on DIP adsorption and desorption processes, especially the scavenging effect caused by the elevated sediment resuspension in gated rivers.

2. Methods

2.1. Sampling

An estuarine surface sediment (ca. 0-5 cm) bulk sample was collected using a plastic shovel in November 2019 from the DuLiuJianHe River, Tianjin, China, above a tidal gate (38°45’58.25” N; 117°33’57.76” E) close to the river mouth. The sediment sample was air-dried at room temperature (~ 27 ℃) to a constant weight, then softly ground to crush aggregates and sieved through 30 mesh.

A real seawater sample, with salinity of 30, was collected with a plastic bucket in January 2020 from Dongjiang Port (39°1’8.38” N; 117°49’5.80” E), Tianjin, China. In the laboratory, the seawater was filtered through 0.45 μm filter to remove suspended particles, and stored in clean polyethylene bottles at 4 ℃ for later experiments.

Basic physical and chemical characteristics of the collected sediment and seawater samples are presented in Table S1.

2.2. Experiments

A solution with the suspended sediment sample in real seawater and NaCl solution were used to conduct the P adsorption and desorption experiments. To simulate the change in salinity from freshwater to seawater, solutions with a salinity gradient of 0, 5, 15 and 30 were prepared by diluting the real seawater, NaCl solution and artificial seawater with deionized water. For simplicity, the aqueous solutions made from real seawater, NaCl solution and artificial seawater are referred to as seawater, NaCl solution and artificial seawater, respectively. Four laboratory experiments were conducted: 1) Adsorption; 2) Desorption; 3) Kinetics; and 4) Salt-specific effects. All mixtures were shaken at 130 rpm for 48 h in a constant temperature-water shaker at 27 ℃, to achieve adsorption equilibrium. The suspensions were centrifuged at 6000 rpm for 10 min, and the supernatants were filtered through 0.45 μm filter. The DIP content in the filtrates were measured spectrophotometrically using the ammonium-molybdate method (Hansen and Koroleff, 2007; Murphy and Riley, 1962).

1) Adsorption experiments

For the phosphate adsorption on suspended sediment, 39 mL solutions with different salinities were added 1 mL 2 mM KH_2PO_4 solution, generating a P concentration of 50 μM. A maximum sediment concentration of 40 g L^{-1} was used in this study. A series of adsorption experiments were conducted using solutions (50 μM P) with sediment levels of 1, 5, 10, 20 and 40 g L^{-1} under different salinities (0, 5, 15, 30). Each treatment was measured in duplicate.

2) Desorption experiments

No extra P was added to the solutions. Besides from this, the DIP desorption experiments were conducted in a similar manner as for the adsorption experiments with sediment concentrations at 2, 10, 20, and 40 g L^{-1} and salinities at 0, 5, 15 and 30.

3) Kinetic experiments

The DIP concentration was reduced to 20 μM in the kinetic experiments in order to study the DIP adsorption efficiency on sediment devoid of any adsorption capacity limitations. Salinities and concentrations of suspended sediment tested in the kinetic experiments were 0, 5, 30 and 10, 20, 40 g L^{-1}, respectively. After the experiment started, sub-samples were collected at 0.5, 1.0, 2.0, 4.0, 6.0, 10.0, 16.0, 24.0, 34.0, 48.0 h. Other experimental conditions were the same as described for the adsorption experiment.

4) Salt specific experiments

In order to assess the specific effects of different sea salt ion compositions on DIP adsorption, $CaCl_2$, $MgCl_2$, Na_2SO_4, and $NaHCO_3$ solids were added, respectively, to NaCl solution with 15 salinity to form a final concentration of 0.01 M for the added salts. This led to a slight increase in the salinity by between 0.84 to 1.42. Then, the P adsorption experiments were conducted with a sediment concentration of 5 g L^{-1}. In order to compare influence of multiple ions on DIP adsorption, an adsorption experiment with salinities at 5, 15, 30 and sediment concentration at 10 g L^{-1} was conducted in seawater, NaCl solution and artificial seawater.

2.3. Statistical analysis

All the data of the amount of adsorbed and desorbed P was subjected to the ANOVA using MIXED procedure (SAS Institute 2010) to evaluate the effects of salinity, sediment concentration, and solution types with their interaction. The effects were deemed significant when $P < 0.05$; differences among least square means for all of the treatment pairs were identified using LSMEANS (/diff) statement (t-test). The results are listed in Tables S2 and S3.

3. Results and discussion

3.1. Scavenging effect of suspended sediments

Field-based studies reported decreasing DIP concentrations with increased loading of suspended sediments during flood season (Li et al., 2017; Ding et al., 2019). Our laboratory experiments explain these results by showing that with an increase in suspended sediment loading (Sc, g L^{-1}), the total amount of adsorbed P (Q_{ad}, μmol) onto the suspended sediment generally increased (**Fig. 1a**), i.e. removing more than 80% of the DIP in water. The relative amount of absorbed P normalized to the amount of sediment (q_{ad} = μmol Q_{ad} / g sediment) decreased with increased suspended sediment concentration (**Fig. 1a**), indicating a limitation of DIP on the adsorption equilibrium.

The total amount of desorbed P (Q_{de}, μmol) from the suspended sediment increased almost linearly with the suspended sediment concentration for concentrations below 20 g L^{-1} (**Fig. 1b**). Above this level, the Q_{de} decreased slightly with increased suspended sediment (**Fig. 1b**) due to re-adsorption of desorbed P to the suspended sediments. This may indicate a DIP limitation to how much P can be desorbed from the suspended sediments due to opposing adsorption/desorption processes. Regardless, the absolute values for DIP adsorption were always higher than that for P desorption. This reflects that P tends to adsorb onto suspended sediment, rather than desorb from suspended sediment.

The normalized values of adsorbed and desorbed P (q_{ad} and q_{de}) are linearly correlated with the suspended sediment concentration after logarithmic transformation (**Fig. 1c**). The regression slopes were negative albeit larger than -1 (as would be expected if there was no effect of sediment concentration on adsorption/desorption) for all the adsorption and desorption experiments **(Table 1)**. This is partly due to the more effective contact between dissolved phosphorus and the suspended sediment surface under the conditions of relatively low sediment loading (Cao et al., 2011).

The slopes of the adsorption regressions generally increased with increased salinity. This indicates that the influence of suspended sediment concentration for DIP adsorption is less important under low salinity (**Fig. 1c**, **Table 1**). On the other hand, in the desorption experiments no significant difference in the slopes under different salinities were observed, except for deionized water (Salinity=0) (**Fig. 1c**, **Table 1**). This implies that for saline water the suspended sediment concentration primarily controls the desorption of P. The exception for zero salinity suggests that the desorption of P from the sediment suspended in freshwater is stronger than that in saline water.

DIP absorption typically reached equilibrium within 10 hours and 80% of the equilibrium within an hour (**Fig. 2**). The DIP removal percentage from aqueous phase was strongly correlated ($r = 0.96 - 0.99$) with the concentration of suspended sediment, reaching between 60% and 85% of DIP in water being scavenged by suspended sediment when the suspended sediment concentration was 40 g L^{-1} (**Fig. 2**). Increasing the salinity from 0 to 5 increased the DIP removal at equilibrium (Q_{ad}) by up to 80% in the saltwater solutions (**Fig. 3**). This strong increase in removal efficiency, with only slight increase in salinity, illustrates the important effect increased salinity has on the sediments' scavenging of DIP in brackish estuarine waters.

These adsorption/desorption laboratory experiments demonstrate that suspended sediments in the estuarine zone have a strong capacity to efficiently and rapidly remove DIP from estuarine water. This is especially important in estuaries receiving discharge from gated rivers. The strong hydrodynamic force when the gate opens re-suspends bed sediments, thereby episodically increases the concentration of suspended sediments in the gated estuary. Increasing salinity in estuaries enhances the DIP scavenging by the suspended particles. Salinity increase also causes the suspended particles with the adsorbed P to flocculate into larger particles that settle faster in the deeper estuarine channels (Snedden et al., 2007; Figueroa et al., 2020a, 2020b). The fate of the DIP absorbed to settling particles remains unknown, but it is possible that the actual seaward flux of DIP, and hence total P, is overestimated by disregarding the scavenging effect by suspended sediment, especially for the estuaries of gated rivers.

3.2. Salinity effect

Due to the predominant inner-sphere specific chemical bonding of PO_4^{3-} to net negatively charged particles, ions such as Ca^{2+}, Mg^{2+}, SO_4^{2-} and HCO_3^- actually determine the overall P adsorption and desorption in estuarine water. The effect of increased salinity on DIP adsorption differed between the three experimental solutions, i.e. NaCl solution, seawater, and artificial seawater (**Fig. 3**). Generally, the P adsorption is stronger in the artificial seawater and lower in the NaCl solution. This indicates that the DIP adsorption is underestimated if NaCl solution is used and overestimated if the artificial seawater is used to simulate DIP sorption in estuarine waters.

Specifically, in the case of NaCl solution, the total adsorbed P onto the sediment (Q_{ad}) at low salinity (0, 5) was larger than that at high salinity (15, 30). It was reported that the structural destruction of Na^+ on particle and the competitive behavior of Cl^- for active point on sediment, could reduce the P adsorption on sediment (Zhang et al., 2011; Wu et al., 2012; Nguyen et al., 2019). For artificial seawater, there is a strong increase of Q_{ad} from 0 to 5 salinity, but no significant change was measured with further increase in salinity from 15 to 30. For seawater, the DIP adsorption under both low and high salinity conditions is lower than that under moderate salinity (i.e. 15)

(**Fig. 3**). The instable influence of salinity in seawater may be caused by coexistence of cation promotion and anion restraint effects on DIP adsorption (Meng et al., 2014b). Similarly, it seemed that the cation effect played a more important role with the increase of salinity for the artificial seawater. This entails that the underestimation of DIP adsorption for NaCl-solution-based experiments would increase with salinity, and the overestimation of DIP adsorption for artificial-seawater-based experiments would be larger for low and high salinity conditions.

The differences in DIP adsorption change with salinity among the three types of saltwater solutions may conceivably be related to their ionic compositions, as indicated in **Fig. 4**. Divalent cations, such as Mg^{2+} and Ca^{2+}, conceptually promote phosphate adsorption on sediments, by forming bridge binding between negatively charged surface functional groups and the phosphate anion, described above, enhancing the number of DIP adsorption sites on sediments surface (Millero et al., 2001; Spiteri et al., 2008). Concurrently, divalent cations promote coagulation and flocculation and thereby settling of the sediments (Liu et al., 2002). Although restraining effect of SO_4^{2-} on PO_4^{3-} adsorption in other studies (e.g., Flower et al., 2017), SO_4^{2-} was relative to Cl^- observed to enhance the DIP adsorption (**Fig. 4**). The calcite content is 2% in the sediment. It was reported that sulfate may increase calcite solubility, and thereby can promote Ca^{2+} release from the sediment (Liu et al., 2012). The cation Ca^{2+} can offer adsorption point for phosphate on sediment. Free Ca^{2+} will precipitate with SO_4^{2-} in advance because the concentration of sulfate is higher than phosphate (200:1). Calcium sulfate will then be converted into hydroxyapatite acting as a continuous calcium supply pool for phosphate removal (Liu et al., 2012). This pathway may explain the phenomenon, as the pH of the solution was 7.0-8.0 and the Ca^{2+} content of the sediment was 28.8 g kg^{-1} (Table S1). Compared to the pure NaCl solution and seawater, the HCO_{3-} inhibited DIP adsorption. This may be due to the competition between HCO_3^- and PO_4^{3-} for the positively charged sorption sites on sediment (Miller et al., 2001; Flower et al., 2017; Nguyen et al., 2019). Compared with seawater, artificial seawater has higher SO_4^{2-} (3.160 g L^{-1}) and lower HCO_3^- (0.139 g L^{-1}) contents (Table SI-1), and there are no other ions except Na^+ and Cl^- in the NaCl solution. This difference in sulfate could partly explain why the P adsorption was highest in artificial seawater, and lowest in NaCl solution (**Fig. 3**).

3.3. Combined effects on seaward P flux

As described above, both salinity and suspended sediment concentration have significant effect on DIP adsorption on sediment. Spatially, salinity increases and suspended sediment concentration decreases with the distance from the river mouth. In the estuarine environment of gated rivers, the temporal changes in salinity and suspended sediment concentration are governed by the strong hydrodynamic episodes related to the opening of the gates. This means that the effects of salinity and suspended sediment concentration on DIP adsorption and desorption are generally combined in space and time. The distribution ratio (DR) between P in suspended sediment and DIP in water varied with salinity and suspended sediment concentration (**Fig. 5**). In the adsorption experiment using seawater (30 salinity), the DR value increased with suspended sediment concentration (**Fig. 5a**). Within the same level of suspended sediment concentration, the highest DIP adsorption was achieved under medium salinity condition (15). However, the influence of suspended sediment concentration on DIP adsorption was larger than that of salinity (cf. **Fig. 1a**). This implies that during periods with open gates, the enhanced sediment resuspension is probably more important for the DIP adsorption than the antagonistic effect caused by the concurrent reduced salinity of the freshwater pulse.

In the desorption experiment using seawater, the DR values tended to increase with increasing suspended sediment concentration (**Fig. 5b**), indicating that desorption decreases with increasing suspended sediment concentration. On the other hand, the DR value showed a sharp decrease at low salinity (<5), implying that salinity may promote P desorption under such conditions. At higher salinity (>5), the DR values remained stable at low suspended sediment concentration (< 25 g L^{-1}).

However, the DR value increased with continuous increased salinity (>5) in the solutions with higher suspended sediment concentration (> 25 g L^{-1}), indicating that the combined effect of salinity (> 5) and higher sediment concentration (> 25 g L^{-1}) may reduce P release from sediment.

3.4 Implications and perspectives

Both suspended sediment concentration and salinity were found to affect P adsorption and desorption processes in estuaries, though the effect of suspended sediment concentration is larger than salinity. Although P desorption at high sediment concentration is highest around 10 salinity, under low salinity conditions the P desorption is outweighed by the role of suspended sediments. Therefore, the scavenging effect for DIP plays an important role, implying that the overall DIP concentration decreases through the estuarine zone.

DIP concentrations in the world's estuaries exhibit many different patterns across the salinity gradient (**Fig. 6**). These changes from land to sea are governed by many different processes from physical mixing between the riverine and oceanic end-member, inputs from point sources along the salinity gradient, uptake by plants and bacteria, remineralization processes and release of iron-bound phosphate. In addition to these well-studied processes, we submit that absorption/desorption processes should also be considered for assessing the potential impact of riverine DIP inputs on the coastal biogeochemistry, at least in estuaries with hydrological control of freshwater discharge.

The pulse-like water discharge during the open gate period promotes sediment resuspension, and this will enhance adsorption of DIP onto the suspended sediment, potentially reducing the DIP flux to the sea. Even though moderate (15) salinity initially accelerate the P desorption, higher salinity may reduce the P desorption (**Fig. 1b**). Certainly, this effect is limited by the DIP adsorption capacity of suspended sediments, and the P adsorbed on buried sediment may release back into the water column. However, for high sediment concentrations the P adsorption capacity could be sufficient to maintain P absorbed to particles, and the release of DIP from sediment to water column is generally slow and limited within the sediment surface (López-Hernández et al., 1980). Hence, burial of DIP absorbed to sediment particles could be an important sink for P, thereby reducing coastal eutrophication and the potential release of iron-bound phosphate.

Simply scaling the freshwater DIP concentration with the water flow will possibly provide an overestimate of the seaward flux of DIP, especially in estuaries fed by gated rivers. The discrepancy between the actual and estimated values of seaward DIP flux depends on the concentration of DIP and particles in the freshwater discharge. The latter is determined by the number of gates, the fluctuation in hydrodynamic strength, and the gate location, as well as the physical and chemical properties of estuarine sediment. In addition, particle sedimentation is probably enhanced when the gates shut down the upstream water flow. This suggests that alternating hydrological regimes could potentially promote sediment burial of P and reduce the seaward flux of DIP.

Therefore, it is necessary to introduce an imperial correction model to eliminate the erroneous estimates of DIP seaward flux. The imperial correction model might be estuary-specific, but could be derived from the calibration of lab experimental data and field monitoring data. Correspondingly, the monitoring of P in the estuary should cover the salinity range from freshwater to seawater and the whole process of gate open and close, also the above-mentioned influencing factors.

4. Conclusion

The seaward flux of DIP from the terrestrial environment is important for controlling coastal and marine eutrophication. However, this flux is possibly overestimated by not accounting for the physicochemical processes taking place in the gated estuaries where DIP scavenging by suspended sediment is enhanced. Increasing salinity generally enhances the adsorption, and in addition, polyhaline conditions may reduce P desorption. For estuaries fed by gated rivers, the seaward P flux may be limited during period with open sluices due to enhanced P scavenging by higher sediment resuspension, even though the salinity is lower.

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Table 1. The linear fitting parameters of $\ln q_{ad}$ and $\ln q_{de}$ with $\ln Sc$.

		Deionized water	Seawater			NaCl solution		
Salinity		0	5	15	30	5	15	30
Adsorption	slope	-0.37	-0.54	-0.53	-0.23	-0.50	-0.44	-0.12
	intercept	1.33	2.04	2.09	1.05	1.66	1.05	0.24
	R^2	0.97	0.99	0.99	0.78	0.99	0.76	0.65
	p	0.002	0.0002	0.0003	0.05	0.0006	0.05	0.1
Desorption	slope	-0.89	-0.67	-0.66	-0.59	-0.69	-0.70	-0.61
	intercept	0.92	-0.23	0.29	-0.51	0.59	0.49	0.37
	R^2	0.99	0.97	0.99	0.99	0.98	0.97	0.96
	p	0.007	0.02	0.001	0.0003	0.01	0.01	0.02

Figure Captions

Fig. 1. The total amount of adsorbed and desorbed phosphorus (Q, μmol g $^{-1}$) (a), the amount of adsorbed and desorbed phosphorus for unit sediment (q, μmol g $^{-1}$) (b), and the linear relationships between the natural log of adsorbed/desorbed P and natural log of suspended sediment concentration (Sc, g L $^{-1}$) (c) under different suspended sediment concentrations and salinities, using both seawater and NaCl solutions. The error bars mark one standard deviation of the duplicates. The solid and dashed lines denote adsorption and desorption, respectively

Fig. 2. DIP removal percentage over time from the kinetic experiment for different suspended sediment concentrations (Sc) at different salinities of seawater solution.

Fig. 3. Comparison of P adsorption capacity of 10 g L^{-1} suspended sediment for different salinities of NaCl solution, artificial seawater, and seawater. Q_{ad} (a), the total amount of adsorbed P.

Fig. 4. Influence of Ca^{2+}, Mg^{2+}, HCO_3^- and SO_4^{2-} on phosphorus adsorption in NaCl solution (salinity = 15, suspended sediment concentration = 5 g L^{-1}). The concentration of the added ions is 0.01 M. Pure seawater and NaCl solution without ion addition are set as the control.

Fig. 5. The combined effects of salinity and sediment concentration in seawater on P distribution ratio (DR) between P adsorbed to sediment surface and DIP in the water column. Note that the distribution ratio is not the same as equilibrium distribution coefficient (K_d). The data were interpolated by the Kriging method.

Fig. 6. DIP concentrations along the salinity gradient for different estuaries worldwide (De Jonge et al., 1989; Upchurch et al., 1974; Ballagh et al., 2019; Meng et al., 2015; Xu et al., 2015; Lin et al., 2012; Pamplona et al., 2013).

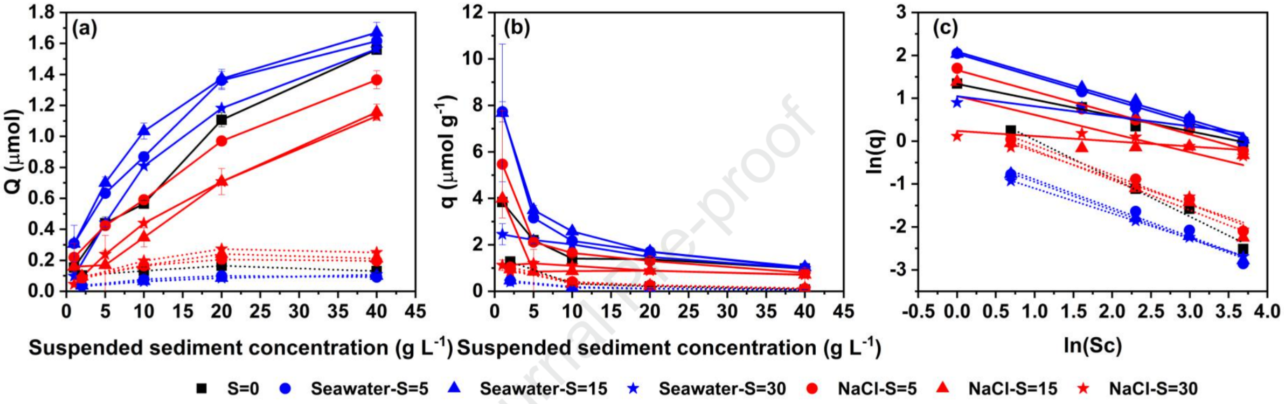
(a)
Q (μmol)
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
0
5
10
15
20
25
30
35
40
45
Suspended sediment concentration (g L^{-1})
(b)
q (μmol g^{-1})
12
10
8
6
4
2
0
Suspended sediment concentration (g L^{-1})
(c)
ln(q)
3
2
1
0
-1
-2
-3
-0.5
0.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
ln(Sc)
S=0
Seawater-S=5
Seawater-S=15
Seawater-S=30
NaCl-S=5
NaCl-S=15
NaCl-S=30

Fig. 1.

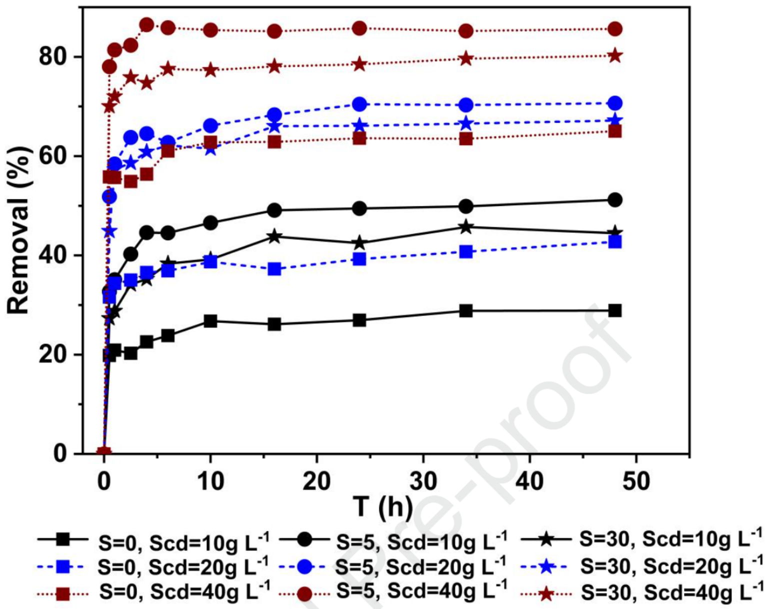
Removal (%)
0
20
40
60
80
T (h)
0
10
20
30
40
50
S=0, Scd=10g L^{-1}
S=5, Scd=10g L^{-1}
S=30, Scd=10g L^{-1}
S=0, Scd=20g L^{-1}
S=5, Scd=20g L^{-1}
S=30, Scd=20g L^{-1}
S=0, Scd=40g L^{-1}
S=5, Scd=40g L^{-1}
S=30, Scd=40g L^{-1}

Fig. 2.

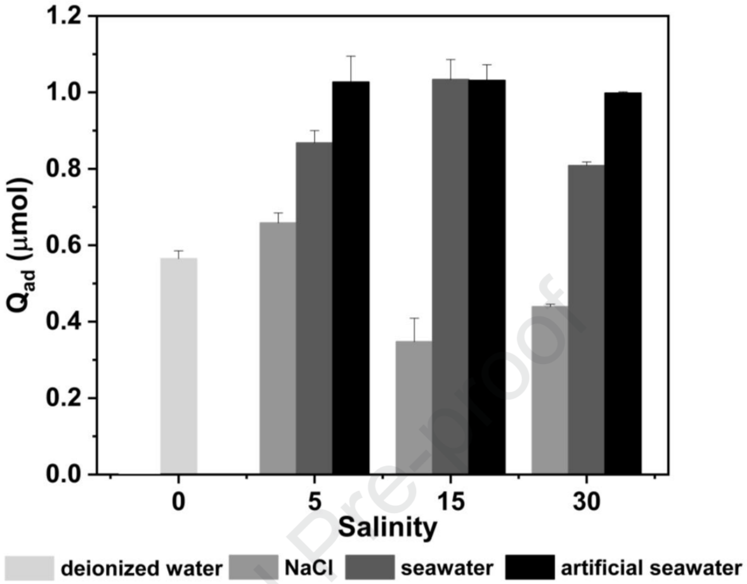
Q$_{ad}$ (μmol)
1.2
1.0
0.8
0.6
0.4
0.2
0.0
0
5
15
30
Salinity
deionized water
NaCl
seawater
artificial seawater

Fig. 3.

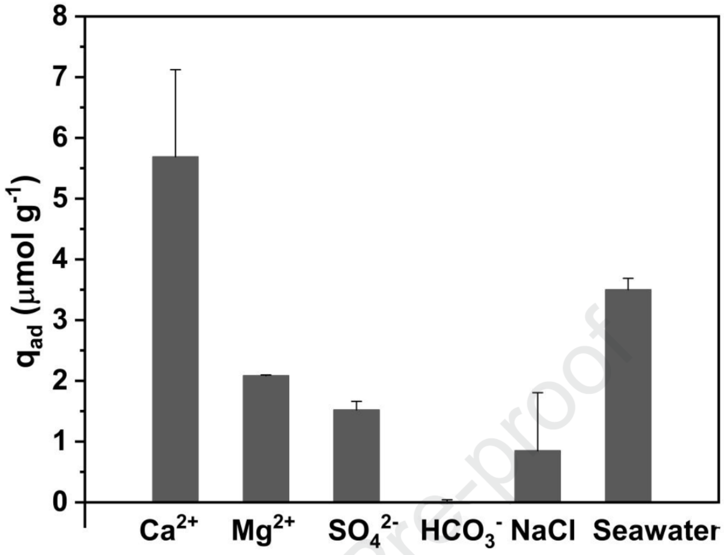

Fig. 4.

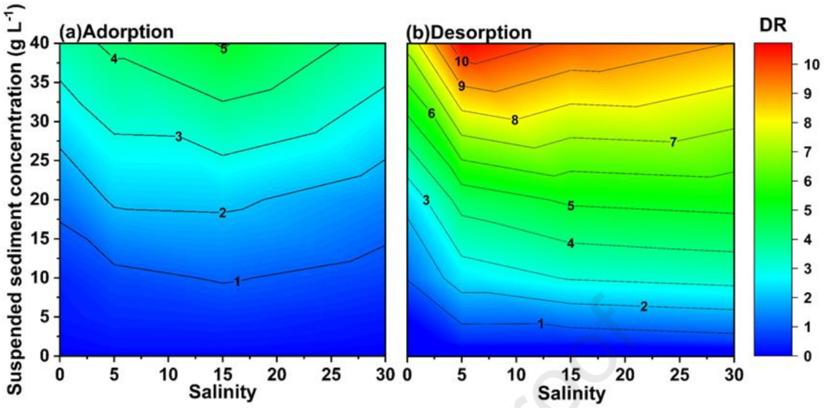
(a)Adorption
(b)Desorption
Suspended sediment concerntration (g L-1)
Salinity
Salinity
DR
0
5
10
15
20
25
30
35
40
1
2
3
4
5
6
7
8
9
10

Fig. 5.

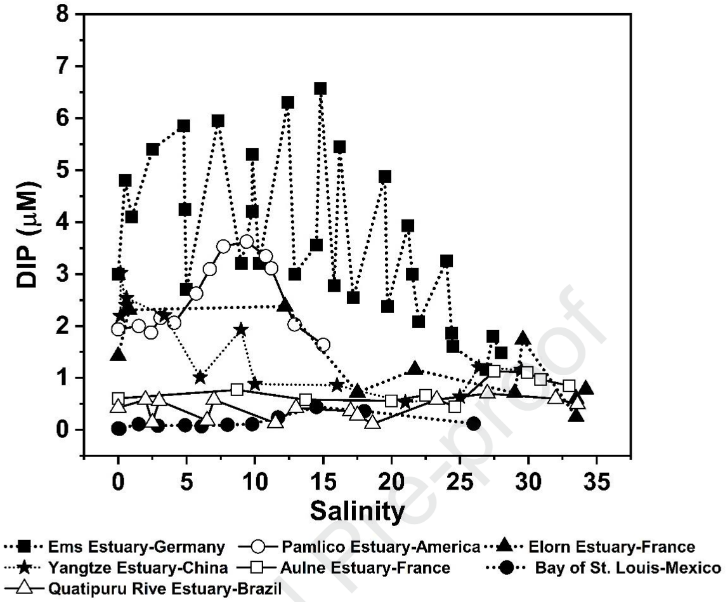
DIP (μM)
0
1
2
3
4
5
6
7
8
Salinity
0
5
10
15
20
25
30
35
Ems Estuary-Germany
Pamlico Estuary-America
Elorn Estuary-France
Yangtze Estuary-China
Aulne Estuary-France
Bay of St. Louis-Mexico
Quatipuru Rive Estuary-Brazil

Fig. 6.

Highlights

1. DIP scavenging by suspended sediment is prevalent for gated estuaries.
2. The increasing of salinity can enhance the DIP adsorption on sediments.
3. Riverine flux of dissolved phosphorus to the coastal sea may be overestimated.
4. Not-real-seawater-based experiments may mislead the DIP flux estimation.

Declaration of interests

☒ 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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: