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Effects of algae and hydrophytes on the inorganic phosphorus pool of shallow hypereutrophic Lake Taihu

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ABSTRACT

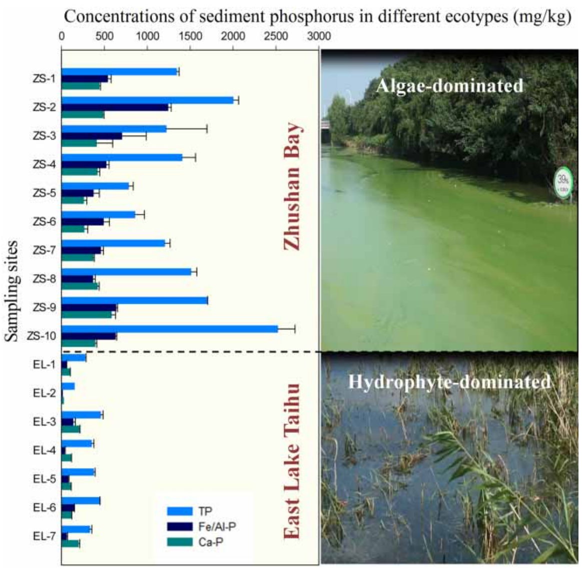
Sedimentary phosphorus is a crucial potential source of phosphorus in the eutrophic lake ecosystem. Different ecological types are supposed to affect the presence and variation of sediment phosphorus. On the basis of field investigations, the total sediment phosphorus load in Zhushan Bay was 1,457.48 mg/kg, nearly four times that of the hydrophyte-dominated area. Thereinto, 41.1% was in the form of iron and aluminum-bound phosphorus, which explicitly indicated the phosphorus contamination there. Analytical methods such as Pearson correlation, contamination assessment and principal component analysis were conducted to find out that 'contamination' was not equivalent to 'release risk'. The contamination classification of East Lake Taihu was 'clean' in general. However, 63.3% of the total phosphorus could be mobilized under certain conditions. Therefore, light phosphorus loading does not equate to less release risk. In the long run, the implicit phosphorus release by the activation of organic phosphorus in hydrophytic areas needs close attention. This study provides a reference to understand the influence of hydrophytes and algae on the phosphorus cycle of sediment.

Key words: algae, hydrophyte, Lake Taihu, phosphorus activation, sediment

HIGHLIGHTS

- Light phosphorus loading does not equate to less release risk.
- Under anaerobic conditions, more than 50% of organic phosphorus could be activated.
- In the long run, the phosphorus release potential of the hydrophytic bay needs close attention.
- The negative effects of algae on sediment phosphorus mainly start since their decay.

GRAPHICAL ABSTRACT



INTRODUCTION

Phosphorus (P) is one of the major elements that determine the trophic status of lakes (Rahutomo *et al.* 2018). Generally, lacustrine phosphorus has two origins, external and internal (Petkuviene *et al.* 2016). In shallow freshwater lakes, almost 70% to 80% of sediment pollution is from external inputs (Zeng & Wu 2009). Under the action of lake currents and molecular diffusion, terrestrial sourced nutrients are mixed and redistributed between water and sediments (Eyre & Balls 1999). However, the effect of external inputs is therefore instantaneous and time limited. After long-term deposition and decomposition, the incompletely decomposed organic matter is deposited and buried in the deeper layer of the sediments, which might be converted to a potential inner nutrient pool (Søndergaard *et al.* 2003), which offers 80% of the nutrient supply for the productivity of shallow lakes (Grenz *et al.* 2000). Therefore, the long-term dynamic of autochthonous phosphorus plays important roles in the lacustrine phosphorus budget (Søndergaard *et al.* 2003).

Generally, the phosphorus release and deposition between the sediment and the overlying water could realize the dynamic equilibrium of lacustrine ecosystems (Zhu & Yang 2018). Previous studies found that dissolved oxygen (Grenz *et al.* 2000), pH (Zhou *et al.* 2016), the presence of nitrogen (Grüneberg *et al.* 2015) and sediment biogeochemistry (Wang *et al.* 2013) were all factors that influence the dynamic of sediment phosphorus.

The existence of aquatic organisms is also of great significance in the phosphorus cycling (Petkuviene *et al.* 2016). Algae and hydrophytes are two of the primary producers in lake ecosystems. With the ecological succession of water areas, the habitats of algae or hydrophytes have developed into a stable environment of different ecotypes (Li *et al.* 2009). In fact, algal and hydrophytic lakes have no significant difference in their DO and pH values. It is different aquatic organisms that could control phosphorus dynamics by changing the DO and pH conditions (Wang *et al.* 2013). It is the same that both algae and hydrophytes could fulfill the photosynthesis and mineralization processes, which could create an oxygen-rich or oxygen-poor environment to control the phosphorus dynamics. What differentiates the hydrophyte from algae is the mechanism of roots on the releasing of sediment nutrients, i.e. roots affect both transmedia migration and transformation by the rhizosphere secretion of oxygen and other substances such as alkaline phosphatase, but algae could not do that (James *et al.* 2004). In addition, to predict the potential of phosphorus release, relying only on total phosphorus (TP) concentration results in inaccurate predictions (Kisand 2005). Not all phosphorus fractions are bioavailable (Li *et al.* 2021) and not all fractions could be activated (Yang *et al.* 2020). Therefore, characterizing specific phosphorus fractions is significant in predicting the possibility of sediment phosphorus release. With the influence of ecotypes (algae and hydrophytes), the transmedia migration and transformation process of specific phosphorus fractions are complex (Petkuviene *et al.* 2016). However, reports on the influencing mechanism and key factors controlling transmedia phosphorus cycling in the presence of vegetation or algae are not clear, which calls for further research.

Given the significant implications of ecotypes on nutrient loading in lake ecosystems, this study selected the algal Zhushan Bay (ZS) and hydrophytic East Lake Taihu (EL) of Lake Taihu (China) as the research subjects, and a sample from Lake Center (C) as blank control. With the dominance of inorganic phosphorus (IP) (60%–80%) to TP in sediments of most lakes, an analysis of inorganic phosphorus could provide substantial information on phosphorus dynamics (Meng *et al.* 2014). Based on SMT, Fe/Al-P and Ca-P are two of the major inorganic phosphorus forms that could be extracted; Fe/Al-P and Ca-P are indicators of autochthonous release potential and exogenous pollution, respectively (Ding 2010). The distribution trends of TP and major inorganic phosphorus forms were measured to predict the release potential of sediment phosphorus. Methods of Pearson correlation and principal component analysis (PCA) were applied to find the possible causes and limiting factors of inorganic phosphorus loading among different ecotypes, which might provide scientific support for the comprehensive management of eutrophic freshwater lakes.

METHODS

Study sites and sediment sampling

Sediment samples were collected from eutrophic Lake Taihu (30°55′40″N ~ 31°32′58″N, 119°52′32″E ~ 120°36′10″N) in August 2015. Lake Taihu, the third largest shallow freshwater lake in China with a surface of 2,338 km^2, is situated south of the Yangtze River Delta. The water depth is about 1.9 m and wave processes occur frequently in Lake Taihu due to its shallowness and special shape. It is a typical 'bi-stable' lake with algal and hydrophytic bays that maintain long-term stability (Zhang 2006). Zhushan Bay, located northwest of Lake Taihu, where the breakout of algae blooms happens frequently, is a typical algal bay. East Lake Taihu, located east of Lake Taihu with hydrophyte coverage reaching 72.4%, is a typical hydrophytic bay.

A total of eighteen intact sediment cores were collected with a gravity corer from Zhushan Bay (ZS), East Lake Taihu (EL) and the lake center (C) of Lake Taihu (Figure 1, Table 1). The sediment cores were sectioned horizontally into slices with a

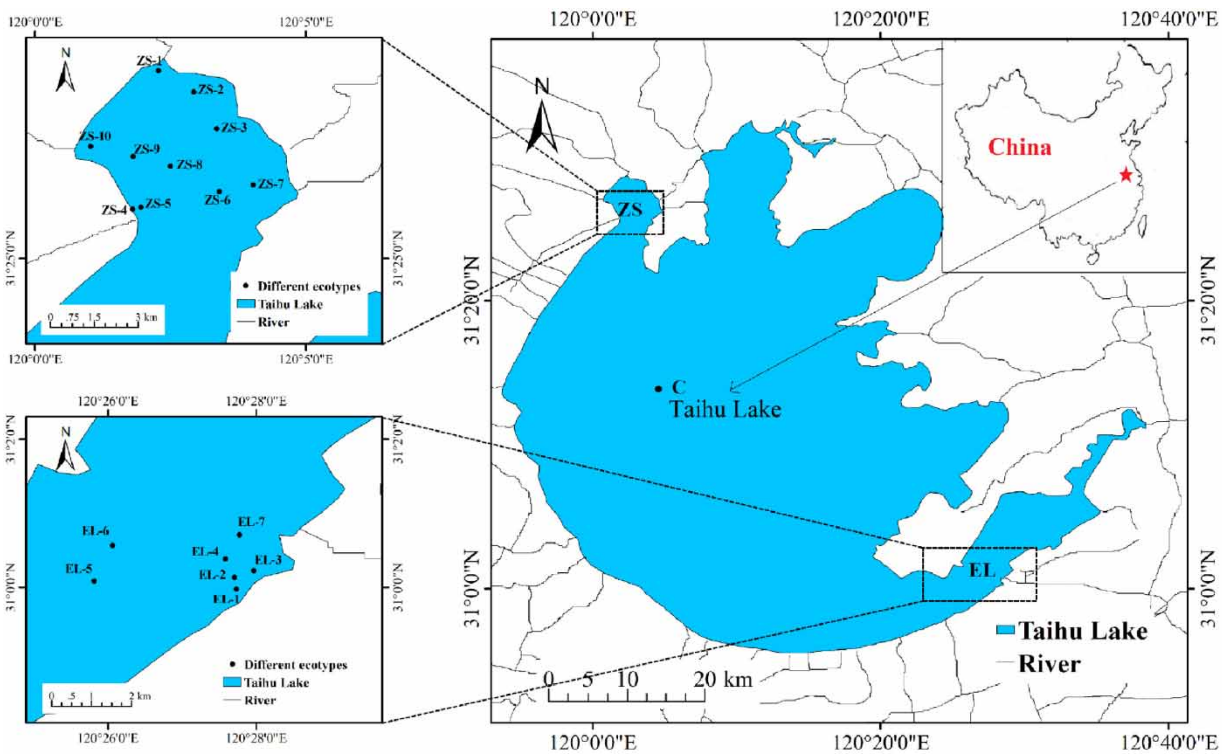


Figure 1 | Location of sampling sites in algae-dominated Zhushan Bay (ZS-1 to ZS-10) and hydrophyte-dominated East Lake Taihu (EL-1 to EL-7) of Lake Taihu (China) with the lake center (C) as the blank control.

Table 1 | Environmental parameters (water temperature (T), water depth (h), dissolved oxygen (DO), reduction/oxidation potential (ORP) and pH) in near-bottom water, sediment water content (WC), loss-on-ignition (LOI) and organic phosphorus (OP) concentrations in the surface layer of sediments

	Station	h (m)	T (°C)	DO ($mg \cdot L^{-1}$)	ORP (mV)	pH	WC (%)	LOI (%)	OP (mg/kg)	Basic conditions
Blank control (C)	Center	3.4	26.3	10.7	30.30	9.07	44.63	5.03	201.13	–
Zhushan Bay (ZS)	ZS-1	1.3	28.8	2.5	131.8	7.19	34.00	3.68	359.97	Reeds distributed along the coastline
	ZS-2	2.1	28.7	5	127	7.40	42.31	5.66	272.09	Hard and odorous, outside the fish farm
	ZS-3	2.3	28.4	6.0	118.3	7.46	39.09	5.47	105.56	Hard
	ZS-4	2.7	30.4	18.7	66.9	9.13	30.40	4.65	462.72	Hard
	ZS-5	2.3	29.7	15.0	62.3	8.76	36.95	3.75	147.71	Black with algae particles on the surface,
	ZS-6	2.5	29.1	9.1	120	7.99	37.80	5.35	100.74	Hard
	ZS-7	2.1	29.4	11.7	112.9	8.20	41.34	5.64	370.49	Soft
	ZS-8	3.1	25.1	6.3	−4.2	7.72	43.74	6.50	719.97	Location of ecological dredging in 2010
	ZS-9	2.4	24.6	6.0	68.4	7.78	58.45	6.50	477.60	–
	ZS-10	1.6	25.2	3.1	−13.5	7.38	40.88	5.12	1,495.16	–
East Lake Taihu (EL)	EL-1	0.7	25.4	0.48	226	7.07	40.03	6.77	119.37	Lotus communities
	EL-2	0.7	26.2	7.24	161	8.16	33.00	3.90	110.16	Reeds communities
	EL-3	0.6	25.6	2.61	184	7.86	47.33	3.77	110.47	Reeds communities
	EL-4	1.7	26.0	7.69	142	8.45	34.85	2.80	203.79	No flora
	EL-5	1.7	25.5	5.23	182	7.82	47.71	4.75	189.10	*Nymphoides peltatum* and *T. bicornis*
	EL-6	0.8	25.7	6.61	174	7.95	58.64	6.70	174.80	*Vallisneria spiralis*
	EL-7	0.5	26.0	7.74	136	8.53	44.47	6.18	69.49	*Zizania aquatica*

thickness of 3 cm (0–30 cm of the sediment cores). The sediment samples were kept at 4 °C and delivered immediately to the laboratory in polyethylene bags and freeze-dried at −45 °C to constant weight. All dried samples were ground with an agate mortar, well homogenized and dry sieved with a nylon mesh (<150 μm) before analysis.

Laboratory analysis

Basic physicochemical parameters, including dissolved oxygen (DO), pH, and reduction/oxidation potential (ORP) of the overlying water at the sampling sites were measured *in situ* using a Hach multi-parameter water quality analyzer (HQ30d, HACH, USA). The water content (WC) and loss on ignition (LOI) of sediment samples were measured before they were freeze-dried. Sediment WC was based on weight losses after drying the samples at 105 °C for 24 h to constant weight (Shi *et al.* 1990). LOI, referred to the organic matter content, was based on weight losses after combustion at 550 °C (Qian *et al.* 2011).

Total phosphorus (TP) and inorganic phosphorus (IP) fractions, including iron and aluminum-bound phosphorus (Fe/Al-P) and calcium-bound phosphorus (Ca-P) were leached following the procedure-SMT (the Standards, Measurements and Testing Programme of the European Commission) (Williams *et al.* 1980). Except TP (200 mg of sediment sample in a crucible to be calcinated at 450 °C for 3 h; add 20 mL of 3.5 M HCl, shake for 16 h and collect the extract), the pair of inorganic phosphorus fractions should be leached subsequently. After the leach of Fe/Al- P (add 20 mL of 1 M NaOH to 200 mg of sediment sample, shake for 16 h; add 4 mL of 3.5 M HCl to 10 mL of the collected extract, let the well-mixed solution stand for 16 h and collect the extract for the analysis of Fe/Al-P); the residue was used for the measurement of Ca- P (after twice washing residues with 12 mL of 1 M NaCl, add 20 mL of 1 M HCl, shake for 16 h and then collect the extract). The measurement of OP (dry the residue at 80 °C; calcinate at 450 °C for 3 h; add 20 mL of 1 M HCl, shake for 16 h and collect the extract) was also based on the residue of IP (add 20 mL of 1 M HCl to 200 mg of sediment sample, shake for 16 h and collect the extract) extraction. All the extracted supernatant collected from the above two steps was measured using the molybdenum blue method.

Each parameter was measured in triplicate for a single subsample (slice) of sediment. For quality control, reagent blanks and standard reference materials GBW07365 (GSD22, Institute of Geophysical and Geochemical Exploration, Chinese

Academy of Geological Sciences) were used to assess the accuracy and precision of the analysis. The analytical precision was within 5% variability. The recovery rates for TC, TN and TP were 98%, 102% and 95%, respectively.

Statistical analysis

Principal component analysis (PCA) is commonly used to find out possible sources of sediment nutrients (Łukawska-Matuszewska *et al.* 2014), under the condition that only factors with eigenvalues greater than 1 (Kaiser Criterion) have explanatory power. Pearson correlation method is always applied to determine the connections between different variables. Based on *in-situ* investigation and laboratory analysis, Pearson correlation and principal component analysis (PCA) were applied to find the relationship between sediment phosphorus loading and other physiochemical parameters, and then figure out possible sources and controlling factors of sediment phosphorus. All experimental data were treated with SPSS 18.0, which incorporates functions of data management, statistical analysis, chart analysis and output management. The analytical result is clear and intuitive. SPSS, together with SAS and BMDP, is known as the world's most influential statistical software, which has been promoted to a variety of operating systems on the computer.

Assessment methodology

At present, there is not an agreed criterion for the evaluation of the health of shallow lakes (Ding 2010). In this study, the Single Index Method and Comprehensive Index Method (Fang 1998) were used to make a scientific evaluation of the health condition of Zhushan Bay and East Lake Taihu.

Since 1970s, with the flourishing development of industry and agriculture, the aggravation of lake eutrophication was regarded as the immediate source of nitrogen and phosphorus increase (Fan & Zhang 2009). Therefore, we assumed that in 1960, Lake Taihu was free of pollution input; the nitrogen and phosphorus concentrations in unpolluted areas could be taken as background values. According to Wang & Dou (1998), the reference values of sediment nitrogen and phosphorus in 1960 were 670 mg/kg and 440 mg/kg, respectively. In this study, the single index and comprehensive index for sediment contamination were defined by the following equation:

$$S = \frac{C_i}{C} \tag{1}$$

$$\mathrm{FF} = \sqrt{\frac{F^2 + F_{\max}^2}{2}} \tag{2}$$

where S was the single evaluation index of TN or TP; C_i was the measured value; C was the background values of TN or TP. F and $F_{\max}$ represented the average and largest values of S_{TN} and S_{TP}; FF was the comprehensive index of each site in the study.

In general, that the relevant index is greater than 1 means that the content exceeds the evaluation standards, and sediment is polluted to some extent. Based on Equations (1) and (2), the sediment contamination was graded into four levels according to the evaluation criteria set according to Chinese national standards (Table 2).

RESULTS AND DISCUSSION

Physiochemical characteristics of algae- and hydrophyte-dominated areas

The physiochemical properties varied greatly across the 18 sediment samples due to the location and ecotypes (Table 1). Overall, water temperature and the pH values ranged from 25.2 to 30.4 °C and 7.07 to 9.13, respectively, with small spatial deviation. The DO concentration was between 0.48 and 18.74 mg/L, with a large spatial difference. The highest and lowest

Table 2 | Standard and level of contamination in sediments of lakes (Fang 1998)

S_{TN}	S_{TP}	FF	Degree
$S_{TN} < 1.0$	$S_{TP} < 0.5$	$\mathrm{FF} < 1.0$	Clean
$1.0 \le S_{TN} \le 1.5$	$0.5 \le S_{TP} \le 1.0$	$1.0 \le \mathrm{FF} \le 1.5$	Slight
$1.5 < S_{TN} \le 2.0$	$1.0 < S_{TP} \le 1.5$	$1.5 < \mathrm{FF} \le 2.0$	Moderate
$S_{TN} > 2.0$	$S_{TP} > 1.5$	$\mathrm{FF} > 2.0$	Hazardous

DO concentrations were found in the Zhushan Bay (ZS-4) and East Lake Taihu (El-1), respectively. The reduction/oxidation potential value ranged from −13.5 to 226 mV. And the relatively lower reduction/oxidation potential in Zhushan Bay than in East Lake Taihu (Table 1), indicated a relatively stronger reducibility in the algal bay. The breakout of algae blooms and the deposition of algae debris create an anaerobic environment, which favors the process of reducing (Wang *et al.* 2013). However, in hydrophytic East Lake Taihu, with dense hydrophytes distributed, the oxygen produced by the assimilation of roots changes the reduction/oxidation potential of the water-sediment interface, which increases the oxidizing ability.

The sediment water content (WC) is the indication of sediment compaction conditions, which determines the re-suspension of sediment nutrients (Li *et al.* 2020). The re-distribution of nutrients in lake ecosystems is largely determined by the re-suspension process. Higher water content represents a strong possibility of nutrient releasing to the overlying water (Li *et al.* 2020). The WC ranged from 30.40% to 58.64% in the two areas, and the average WC value in East Lake Taihu was larger than Zhushan Bay (Table 1). In hydrophytic areas, the silt-promotion effect of aquatic plants to the suspended particles contributes to the refinement of surficial sediment, which helps create a relatively higher water binding capacity than coarse particles (Li *et al.* 2009). While in Zhushan Bay, the average WC value was 40.50%, with the lowest water content in ZS-1 and ZS-4, areas near the estuary. The scouring action from the upper reach, together with the strong wind disturbance from the lake, prevented the deposition of fine particles and biodetritus, which maintained the terrestrial properties (low water content) of sediments there.

The LOI is widely used to represent the loading of organic matter (OM) in sediments (Qian *et al.* 2011). The LOI concentration ranged from 2.80% to 6.77% in Zhushan Bay and East Lake Taihu, with a mean value of 5.23% and 4.98%, respectively. Compared with the lake center, the LOI concentration ranged as Zhushan Bay> lake center > East Lake Taihu (Table 1). The source and fate of the OM in the three areas differed greatly due to the differences in location, hydrodynamics and productivity. In this study, the highest OM loading was at the hydrophyte-dominated East Lake Taihu with lotus communities distributed (EL-1). The production of organic matters during the life circle of hydrophyte and the accumulation of phytodetritus were the main reason for the high concentration of OM in EL (Kleeberg *et al.* 2010).

Surficial sediment phosphorus variations in different ecotypes

An obvious regional difference of sediment phosphorus was found in the algal and hydrophytic bays. With the formation of stable ecotypes, algal and hydrophytic bays were characterized by unique nutrient structure (Wang *et al.* 2013). The average TP concentration in Zhushan Bay was 1,457.48 mg/kg, almost four times that of the lake center (421.40 mg/kg) and East Lake Taihu (343.30 mg/kg) in this study (Figure 2(a)). Zhushan Bay, located at the northwest of Lake Taihu, is a typical targeted area of many inflow rivers. The heavy P loading is largely attributed to the heavy phosphorus input. It is reported that the full release of Fe/Al-P in the sediments of Lake Taihu could result in the phosphorus concentration increase of the overlying water by 0.249 mg/L (Zhu *et al.* 2011). But the TP concentration of the upstream river is greater than 0.47 mg/L (Zhang 2006). Therefore, a steep diffusion gradient from water to sediments favors the sediment adsorption of phosphate anions and formation of Fe/Al-P. With heavy labile phosphorus reserve, the release potential is enhanced accordingly. In addition, the breakout of algae blooms and the following heavy deposition of algal detritus are expected to cause anaerobic conditions, which might favor the release of Fe/Al-P from the phosphorus pool. While, in East Lake Taihu, the relatively lower phosphorus loading could be attributed to the presence of hydrophytes capable of mobilizing and releasing P despite the heavy nutrient input from the adjacent western lake and from the aquaculture facilities within the region (Zhang 2006). Large hydrophyte communities which occupy 72.4% of the East Lake Taihu area can fulfill the phosphorus mobilization through root absorption and translocation into shoots (Kleeberg *et al.* 2010). In addition, due to the decay of hydrophytes, the large amounts of phytodetritus create an anaerobic environment that favors the release of labile Fe/Al-P and the degradation of OP to phosphorus forms that could be taken up by algae and aquatic plants (Wang *et al.* 2013). Therefore, the dual loss of sediment phosphorus through direct uptake and indirect transformation reduced the phosphorus loading of the hydrophytic East Lake Taihu (Figure 2(a)).

In lakes, the phosphorus migration, transformation and deposition are in different forms (Noll *et al.* 2009), the presence of which have a close relation with the environment (Zhu *et al.* 2011). The percentages of inorganic Fe/Al-P and Ca-P, and organic phosphorus (OP) were 41.1%, 27.9% and 31.0% in Zhushan Bay and 22.6%, 36.7% and 40.7% in East Lake Taihu, respectively (Figure 2(d)). While, in the lake center (C), with less pollution input and organic detritus from algae or hydrophytes, only the wave and lake current could initiate the phosphorus release and transformation. After long term

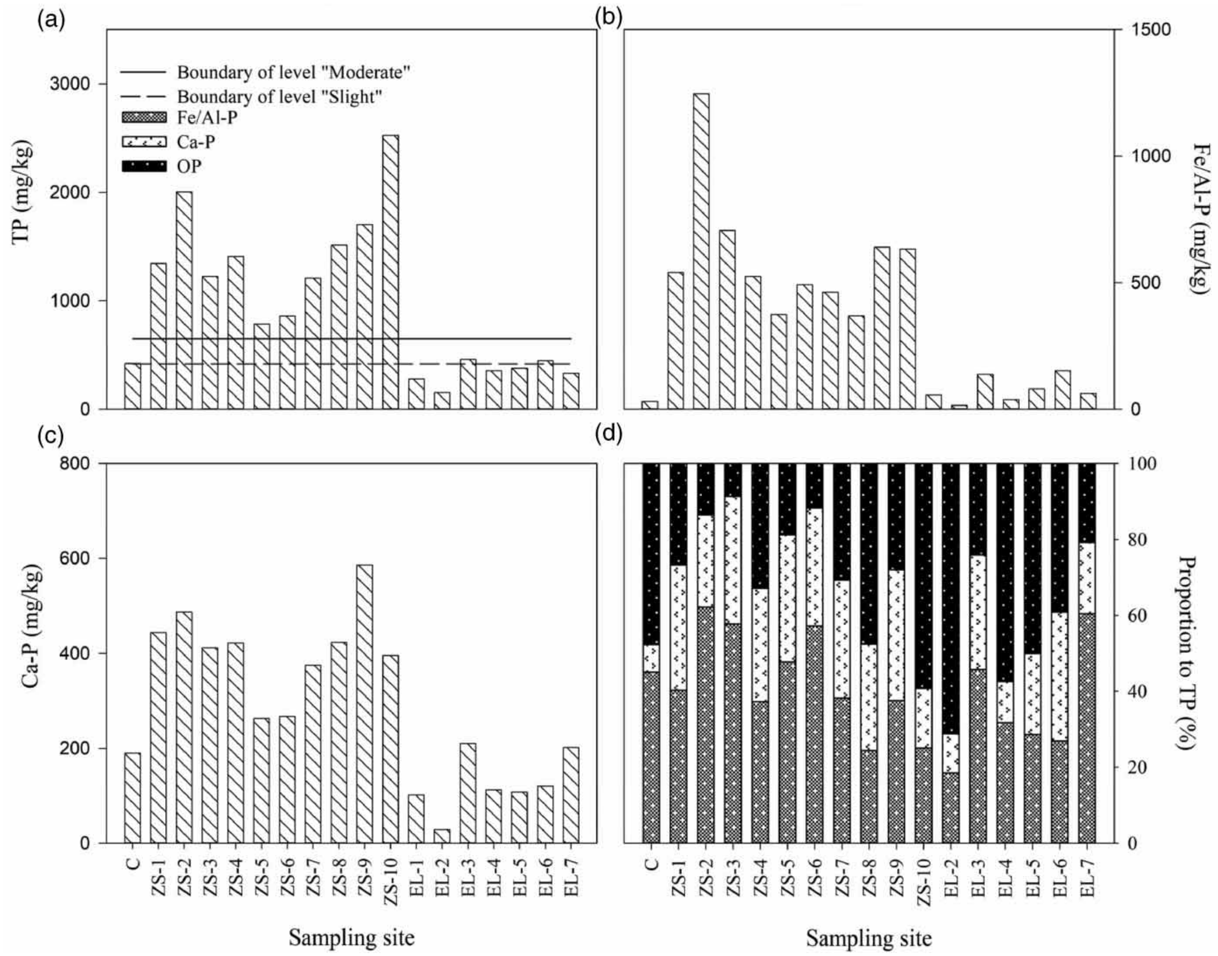


Figure 2 | Distributions of (a) TP, (b) Fe/Al-P, (c) Ca-P, and (d) proportions of Fe/Al-P, Ca-P and OP to TP in the sample groups of Lake Taihu (Black dash line is the boundary of level 'Slight'; Black solid line was the boundary of level 'Moderate', which means values above the line belong to the level of 'severely' polluted).

succession, the labile and available Fe/Al-P was almost depleted, leaving other phosphorus fractions accounting for more than 90% of the TP concentration.

Fe/Al-P has been reported as one of the bioavailable phosphorus forms (Ruban *et al.* 2001), particularly under anaerobic conditions. The concentrations of Fe/Al-P in Zhushan Bay and East Lake Taihu ranged from 19.24 to 1,246.36 mg/kg, with an average of 599.00 mg/kg and 77.75 mg/kg, respectively (Figure 2(b)). The absolute Fe/Al-P content of algal Zhushan Bay was seven times and twenty times greater than that of hydrophytic East Lake Taihu and the Lake Center, respectively. The correlation between sediment Fe/Al-P and parameters of the overlying water was stronger in Zhushan Bay than in other two areas (Table 3). In Zhushan Bay, allochthonous input dominates the phosphorus source, with Fe/Al-P positively correlated with TP ($r = 0.412$, $p < 0.01$) and TN ($r = 0.372$, $p < 0.01$). The Fe/Al-P concentration was also positively correlated with the ORP ($r = 0.493$, $p < 0.01$) (Table 3), which indicated the sensitivity of Fe/Al-P to the change of redox conditions (ORP). Except for the heavy input and source conversion, the quick metabolism of algae and the quick decomposition of its debris were other catalyzers of phosphorus mobilizing. In this sense, large sums of nutrient releasing during the decomposition could be in terms of dissolved fractions kept in overlying water or pore water in the short run. And in areas with few or less hydrophytes distributed, the dissolved nutrients could be assimilated by algae again and then be caught in a vicious circle. Therefore, more Fe/Al-P loading indicated larger phosphorus release potential in algal bays. However, in East Lake Taihu, Fe/Al-P had significant positive correlation with TN ($r = 0.687$, $p < 0.01$) and LOI ($r = 0.325$, $p < 0.01$), and the correlationship was much stronger than Zhushan Bay and Lake Center (Table 3). This phenomenon should be ascribed to the complicated influence from hydrophytes. In fact, the influence that hydrophytes exert on the Fe/Al-P mobility is bidirectional,

Table 3 | The correlation matrix of DO, ORP, WC, LOI, TP, OP, TN and N/P with phosphorus and phosphorus fractions based on the nutrients load of 18 sediment cores

	Zhushan Bay			East Lake Taihu			Lake Center		
Element	TP	Fe/Al-P	Ca-P	TP	Fe/Al-P	Ca-P	TP	Fe/Al-P	Ca-P
DO	−0.256*	−0.246*	−0.335*	−0.112	0.018	0.108	–	–	–
ORP	0.171	0.473**	0.114	0.056	0.067	−0.249*	–	–	–
WC	0.305**	0.256*	0.276**	0.353**	0.656**	−0.114	−0.301	0.411	0.777**
LOI	0.212*	0.147	0.202	0.044	0.354**	−0.214	−0.410	0.336	0.522*
TP	1	0.412**	0.802**	1	0.685**	0.382**	1	−0.012	−0.100
OP	0.592**	0.291	0.480*	0.879**	0.541**	−0.047	0.827**	−0.460	− 0.635**
TN	0.297**	0.372**	0.158	0.415**	0.687**	−0.004	−0.154	0.367	0.840**
N/P	− 0.636**	0.074	− 0.589**	−0.003	0.397**	−0.077	−0.430	−0.124	0.127

*Correlation is significant at 0.05 level (2-tailed).
**Correlation is significant at 0.01 level (2-tailed).

facilitating or inhibiting (James *et al.* 2004). Similar to algal lakes, the anaerobic condition created by the deposition of phytodetritus favors the release of labile Fe/Al-P. However, with the respiration and oxygen secretion of roots, the surface sediments were in a dynamic alternation between being aerobiotic and anaerobic. The transformation capacity of Fe/Al-P was reduced. In addition, the distribution of hydrophytes alleviated hydrodynamic influences, which might to some extent inhibit the phosphorus release. Therefore, the Fe/Al-P loading at vegetated area (EL-3 with reeds and EL-6 with *Vallisneria spiralis* densely distributed) was heavier than areas without flora distributed (EL-4) (Figure 2(b)).

The concentration of Ca-P ranged from 28.57 to 586.18 mg/kg, with an average of 407.28 mg/kg in Zhushan Bay and 125.95 mg/kg in East Lake Taihu (Figure 2(c)). The significant positive correlations of Ca-P and TP ($r = 0.802$, $p < 0.01$) indicated the homologous source of Ca-P and TP. Zhushan Bay is the typical target of exogenous inputs from the upper reaches of Taige River, Caoqiao River and Yincun Port. Ca-P, generally slightly soluble and rarely participating in phosphorus migration and transformation processes, is characterized by burying deposition, whether the original source is terrestrial input (Kaiserli *et al.* 2002) or autochthonous organic skeletons and detritus (Wang *et al.* 2012). Therefore, though with a smaller proportion (27.94%) than that of East Lake Taihu (38.68%) to TP, the absolute concentration of Ca-P in Zhushan Bay (407.27 mg/kg) was three times greater than that of East Lake Taihu. While in the lake center (C), Ca-P had a positive correlation with TN ($r = 0.840$, $p < 0.01$) and negative correlation with OP ($r = -0.635$, $p < 0.01$).

Vertical phosphorus variations in different ecotypes

Vertically, the distribution of phosphorus and different fractions in the three studied areas were incredibly different. The average vertical TP in the lake center, Zhushan Bay and East Lake Taihu were 383.82, 600.23 and 422.31 mg/kg, respectively (Figure 3). They were common in that sediment TP generally showed a downward trend with increasing depth. The vertical variation of TP in Zhushan Bay was the largest with an obvious enrichment in the surface layer (Figure 3). Fan & Zhang (2009) found the cumulative depth of the received phosphorus-rich input could be over 20 cm in Zhushan Bay. As to the specific inorganic phosphorus fractions, the transmedia migration and transformation was complex, which in turn made the vertical variation irregular. In general, with an increase in sediment depth, the enhanced anaerobic environment favors the release of labile Fe/Al-P from sediments (Noll *et al.* 2009). Therefore, the sediment Fe/Al-P decreased with the increase of sediment depth (lake center sample excepted) (Figure 3). The vertical variation of Ca-P in the lake center and East Lake Taihu showed a bottom enrichment trend (Figure 3). In Zhushan Bay, the constant supplement from terrestrial input and autochthonous biodetritus added the Ca-P concentration in the surface (Figure 3).

The difference in the vertical variation between algae- and hydrophyte- dominated environments was largely attributed to the participation of algae and hydrophytes in the transmedia circulation. At the sediment-water interface, the reoxygenation by frequent disturbance from biological factors and hydrodynamics resulted in the surficial enrichment of phosphorus. Vertically, if there is no exogenous input, the enhanced anaerobic environment with an increase in sediment depth always favors

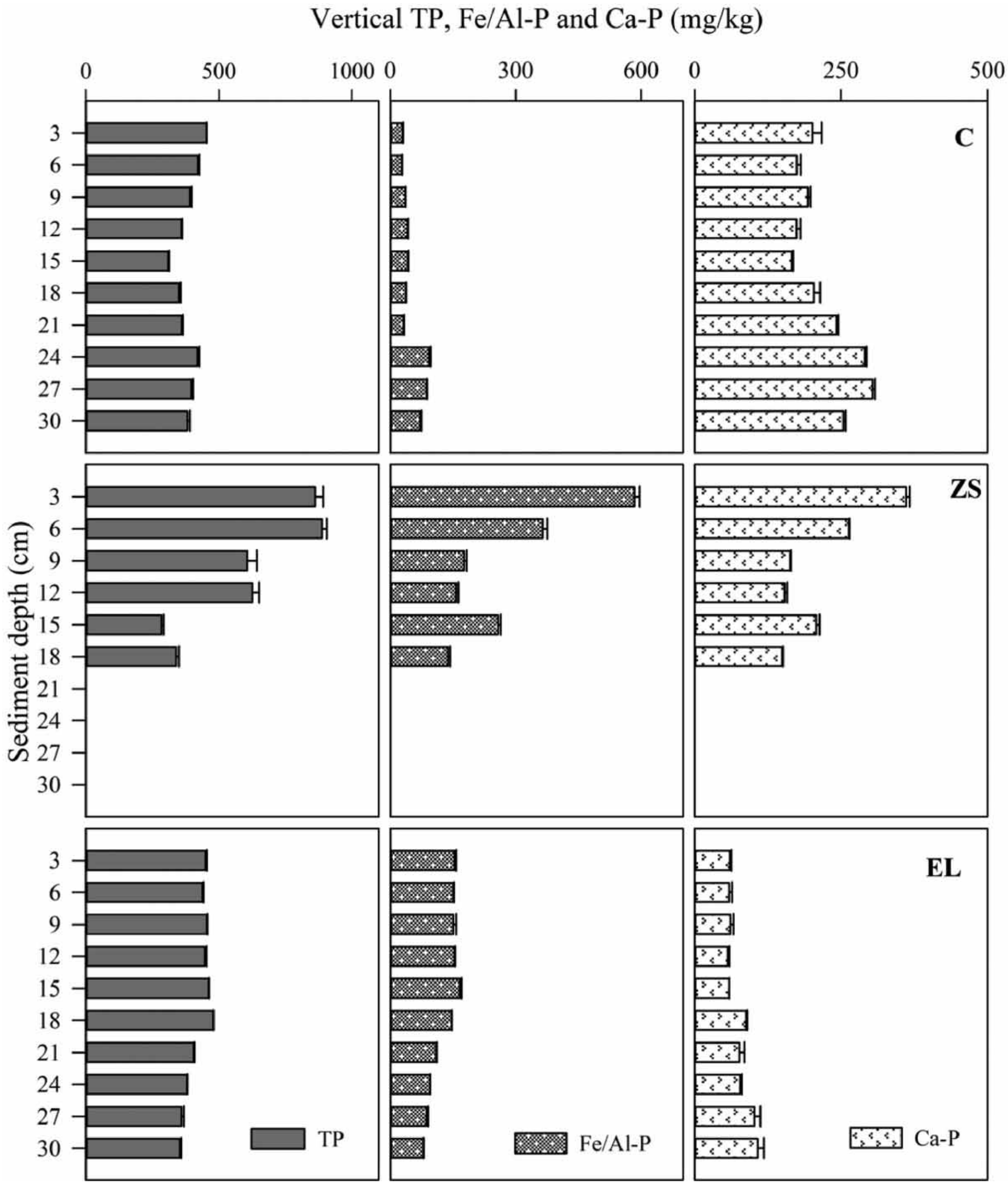


Figure 3 | Vertical profiles of TP, Fe/Al-P and Ca-P in 0–30 cm sediment cores of the lake center (C), Zhushan Bay (ZS) and East Lake Taihu (EL).

the release of labile phosphorus forms from sediments (Noll *et al.* 2009). Therefore, the TP concentration in the lake center decreased as the depth increased, with a relatively stable variation (Figure 3).

The TP variation is accomplished by a series of physical and chemical actions by different phosphorus fractions. Among them, Fe/Al-P was positively correlated with the ORP in algal Zhushan Bay ($r = 0.473$, $p < 0.01$) (Table 3). With the vertical depth increased, the decreased ORP favored the transformation from Fe^{3+} to Fe^{2+}, suggesting that the adsorbed $Fe(OH)_3$ might be released, and the adsorbed $FePO_4$ changed into dissolved form to result in the release of sediment phosphorus. However, the environmental factors such as DO, pH and ORP varied drastically from the water-sediment interface to 15 cm below (Qin *et al.* 2008). Therefore, deeper than 15 cm, due to the amount of phosphorus input surpassed the mineralization and transformation capacity, the proportion of inorganic phosphorus was larger than the surface (Figure 4). On the contrary, vertical Fe/Al-P in hydrophytic East Lake Taihu had a weak relationship with ORP, and strong relationship with sediment OP, WC and TN concentration (Table 3). As shown in Figure 3, the Fe/Al-P had small but stable vertical variation. From 5 to

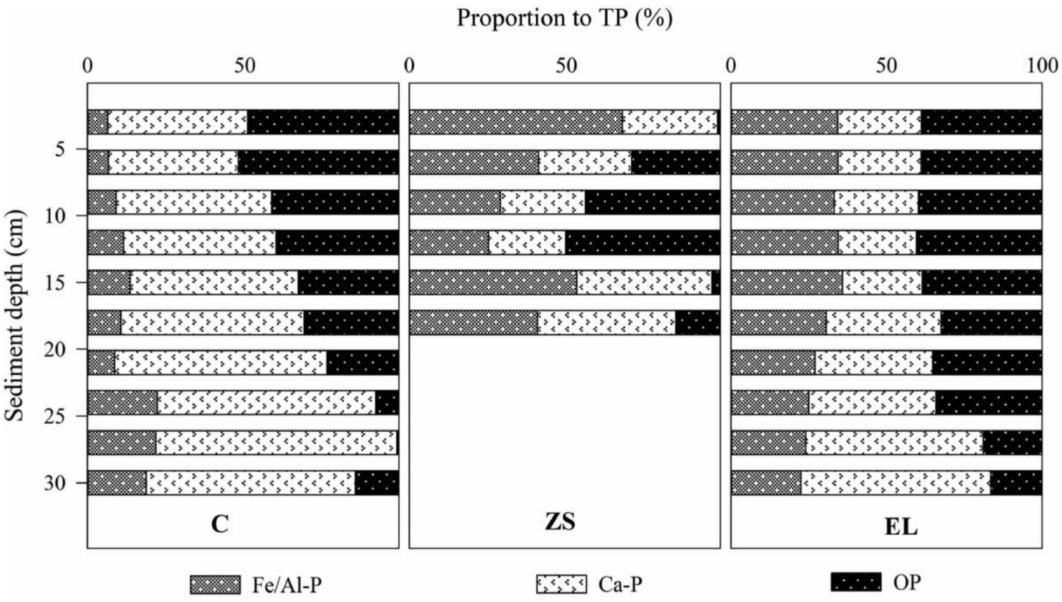


Figure 4 | Vertical proportions of P fractions to TP in Zhushan Bay (ZS), East Lake Taihu (EL) and lake center (C).

15 cm below the sediment surface, roots play the role of adjusting the equilibrium of the oxygen concentration (Wang *et al.* 2012). With the respiration and oxygen secretion of roots, the sediment DO condition was altered. The transformation capacity of Fe/Al-P was reduced. However, in the sediment below 15 cm, with few influences from the roots, the vertical variation of Fe/Al-P was small and nearly stable, which was in line with the lake center.

The vertical Ca-P proportion was in an increasing trend in the three areas (Figure 4). In general, the contributors to Ca-P are slightly soluble calcium phosphate minerals (Kaiserli *et al.* 2002). The high stability of these minerals in sediments makes them the final products of the early diagenesis (Rydin 2000). The constant burying and lower consumption characteristics determined the vertical increasing trend of the East Lake Taihu and the Lake Center (Figure 3). However, in the algae-dominated Zhushan Bay, algae breakout and decay are major sources of Ca-P (Zhang 2006). The accumulation of fresh algae and residues resulted in the surficial enrichment of absolute Ca-P concentration (Figure 3).

Contamination assessment and source assignment of phosphorus

In general, Zhushan Bay and East Lake Taihu were classified as hazardous and clean respectively by Comprehensive Index Method (Table 4). Compared with the contamination status of 2010 (Ding 2010), the comprehensive nutrient contamination of Zhushan Bay deteriorated due to the phosphorus pollution, while East Lake Taihu was better due to the abatement of nitrogen pollution.

The comprehensive index (FF) of Zhushan Bay ranged from 1.60 to 3.59 (Figure 5), with the mean of 2.89. Almost all sites at Zhushan Bay were at hazardously polluted level except for ZS-5 and ZS-6, which was attributed to the slight and moderate nitrogen pollution. But assessment based on the Single Phosphorus Index, all the sampling sites in the Zhushan Bay were at hazardously polluted level. The contamination level in the lake center was 'Slight' by Single Phosphorus Index method and 'Clean' by Comprehensive Index method (Table 4). Obviously, ZS-6 and C were common in that there were no hydrophytes or algae influence. The main difference between the two sites was the input from the dense inflow-river net and the trapped

Table 4 | Contamination assessment for the surface sediments Zhushan Bay and East Lake Taihu based on all sediment samples in this study

Station	S_{TN}	Degree	S_{TP}	Degree	FF	Degree
C	0.75	Clean	0.96	Slight	0.85	Clean
ZS	1.85	Moderate	3.31	Hazardous	2.98	Hazardous
EL	1.62	Moderate	0.78	Slight	0.91	Clean

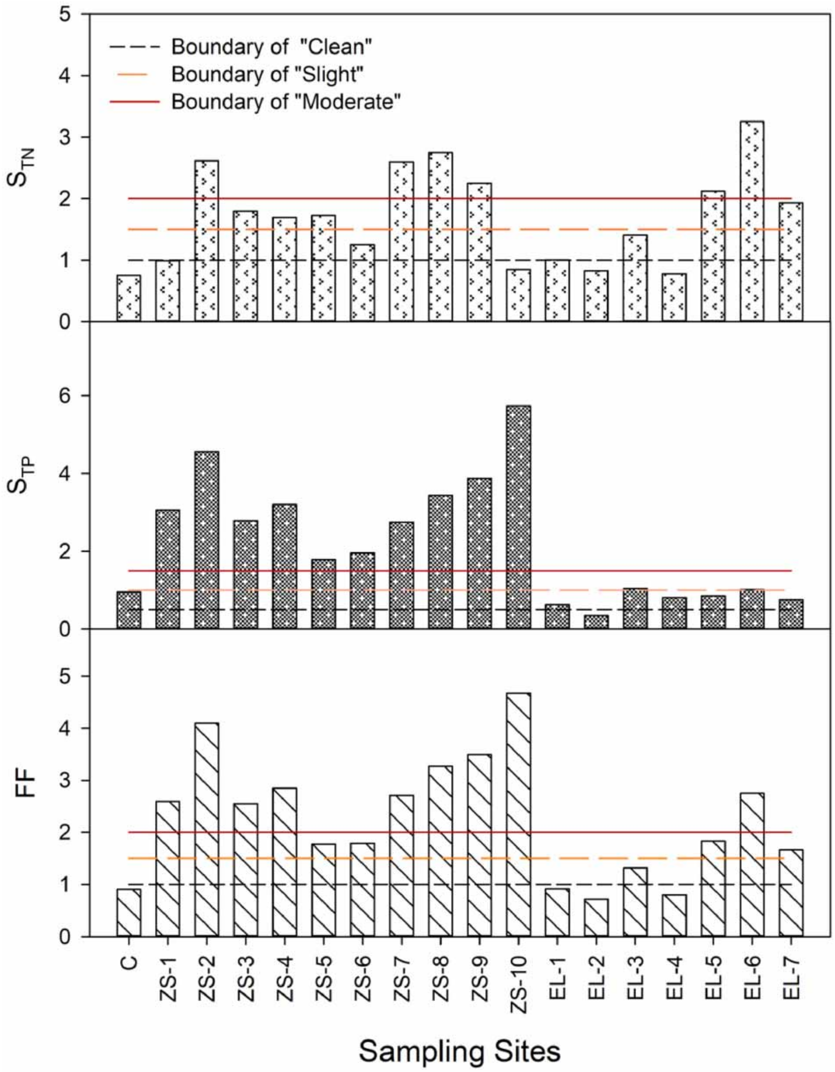


Figure 5 | Contamination assessment by Single Nitrogen Index (S_{TN}), Single Phosphorus Index (S_{TP}) and Comprehensive Index (FF) of the sediment in Lake Taihu. (Black dashed line is the boundary of level 'Clean'; Orange dashed line was the boundary of level 'Slight'; Orange dashed line was the boundary of level 'Moderate', which means values above the line belong to the classification of 'Hazardous' level). The full colour version of this figure is available in the online version of this paper, at http://dx.doi.org/10.2166/ws.2021.276.

water flow that created the nutrients deposition and retention of Zhushan Bay. In East Lake Taihu, the comprehensive index ranged from 0.59 to 2.13, nearly all sampling sites were at level 'Clean' and 'Slight', except for EL-6, where the *Vallisneria spiralis* were in their declining phase and rich in phytodetritus. And the Single Phosphorus assessment value ranged from 0.35 to 1.04, with all sites below the 'Moderate' polluted level.

Comparing the contamination conditions of Zhushan Bay, East Lake Taihu and the lake center, Zhushan Bay was hazardously contaminated both by Single Phosphorus Index and Comprehensive Index methods, the other two were at slight or clean level. But the nitrogen contamination was at the same level in Zhushan Bay and East Lake Taihu (Table 4, Figure 5). It was obvious that the nitrogen loading might be a potential catalyzer of nutrient release in the long run. In hydrophyte-dominated bays in particular, the phytodetritus might deposit on the surface sediments, which directly increase the loading of TN and OM (Wu & Hua 2014). In addition, under anaerobic conditions, Fe/Al-P could be regarded as totally mobile, and organic phosphorus could be degraded to mobile phosphorus forms (Rydin 2000). While in the lake center, the frequent reoxygenation by prevailing wind inhibited the phosphorus activation. Only Zhushan Bay and East Lake Taihu met the anaerobic requirements of the activation of Fe/Al-P and OP. Therefore, potential mobile phosphorus accounted for 70.56% and 63.3% in Zhushan Bay and East Lake Taihu respectively.

Table 5 | Component matrix for active variables (TP, Fe/Al-P, Ca-P, OP, TN) and explanatory variables (near-bottom concentration of dissolved oxygen (DO), redox potential (ORP), sediment water content (WC), loss on ignition and molar ratios of TN:TP

	Zhushan Bay			East Lake Taihu			Lake Center		
Element	1	2	3	1	2	3	1	2	3
TP	0.852	0.428	0.167	0.134	0.985	0.027	0.882	−0.361	0.136
Fe/Al-P	0.771	0.484	0.197	0.594	0.655	−0.006	0.202	−0.079	0.926
Ca-P	0.772	0.502	0.068	−0.249	0.389	0.450	0.565	0.232	0.731
OP	0.602	−0.057	0.190	0.107	0.866	−0.204	0.298	−0.511	−0.764
TN	0.511	−0.095	0.813	0.892	0.289	0.176	0.774	0.610	0.054
TN/TP	−0.329	−0.392	0.783	0.881	−0.139	0.201	−0.034	0.975	−0.079
WC	0.792	−0.140	−0.395	0.827	0.268	−0.053	0.916	−0.078	0.025
LOI	0.835	−0.007	−0.353	0.817	−0.092	−0.248	−0.117	0.779	0.314
ORP	0.281	0.871	−0.294	0.061	0.071	− 0.945	–	–	–
DO	−0.203	− 0.931	0.083	0.255	−0.174	0.911	–	–	–
Eigenvalue	4.092	2.481	1.755	3.437	2.519	2.104	3.125	2.625	1.420
% of variance explained	40.9%	24.8%	17.5%	34.4%	25.2%	21.0%	39.1%	32.8%	17.8%
% of cumulative	40.9%	65.7%	83.2%	34.4%	59.6%	80.6%	39.1%	71.9%	89.7%

In addition, the depositional environment and possible sources of pollutants also have impacts on the nutrient loadings of certain lakes. With the PCA method, the possible causes and limiting factors of inorganic phosphorus loading among different ecotypes were investigated, and three principal components were extracted to explain 83.2%, 80.6% and 89.7% of all the relevant variables in Zhushan Bay, East Lake Taihu and the lake center, respectively (Table 5).

In Zhushan Bay, PC1, PC2 and PC3 explained 40.9%, 24.8% and 17.5% of all the variables, respectively (Table 5). In general, PC1 always includes the decisive factors in the data (Łukawska-Matuszewska *et al.* 2014). In this study, the TP, Fe/Al-P, Ca-P, OP and sediment WC and OM concentration had substantial loadings on the PC1 axis, reflecting the dominant position of phosphorus and sediment quality of Zhushan Bay. Environmental parameters of the near-bottom water (DO and ORP) had substantial loadings on PC2. Sediment Fe/Al-P in Zhushan Bay had significant correlation with ORP ($r = 0.473$, $p < 0.01$), which indicated the significance of oxidation-reduction environment in the bioavailability of Fe/Al-P (Meng *et al.* 2014). Therefore, PC2 represented the phosphorus bioavailability of the bay. PC3, with high loadings of TN and TN/TP ratio, indicated that nitrogen loading could be another controlling factor of phosphorus surplus.

In East Lake Taihu, PC1, PC2 and PC3 explained 34.4%, 25.2% and 20.6% of all the variables, respectively (Table 5). The high loadings of TN, TN/TP, WC and LOI on PC1 highlighted the significance of the depositional environment and nitrogen loading. In the sample from East Lake Taihu, all the sections of the whole sediment core were covered with organic debris from the decay of hydrophytes, which were reported as rich in carbon and nitrogen (Wang *et al.* 2013). WC and LOI were indicators of the migration and transformation ability of sediment nutrients. In this study, the positive correlation of TP and Fe/Al-P with TN (Table 3), together with high loading of WC and LOI on PC1 reflected the importance of the depositional environment for phosphorus loading in East Lake Taihu, rather than phosphorus itself. On PC2, the high positive loadings of TP, Fe/Al-P, and OP indicated the secondary position of phosphorus in East Lake Taihu. And the significant correlation among the three fractions just verified the transformation possibility from OP to bioavailable Fe/Al. On PC3, the negative ORP and positive DO loadings reflected the impact of environmental factors on phosphorus transformation.

While in the lake center, PC1, PC2 and PC3 explained 39.1%, 32.8% and 17.8% of all the variables, respectively (Table 5). TP, TN and WC had positive loadings on PC1. There was no dominance of P or N in the lake center. The nutrients exchange between water and sediments was restricted by less terrestrial input and autochthonous deposition, with only wave and lake current able to control the nutrients exchange. In the lake center, frequently affected by wave and current, resuspension always happened. And WC was regarded as a good indicator of sediment resuspension potential. LOI had positive loading on PC2, which was consistent with its positive correlation with Ca-P and OP (Table 3). After long-term evolution, the labile Fe/Al-P was released and migrated by frequent suspension, with only Ca-P and OP left to be adsorbed by the remaining organic matter. Three major phosphorus fractions had high loadings on PC3, which indicated the importance of transformation between different fractions. The lake center was areas lack of conditions favoring this transformation. Therefore, the phosphorus fractions were not that important in the lake center.

CONCLUSIONS

The cause and status quo of sediment phosphorus pollution differs greatly in different ecotype areas. Contamination is not exactly equal to nutrient release. Algal Zhushan Bay, suffering from heavy pollution input from upstream carrying, was hazardously polluted with heavy labile Fe/Al-P loading. Leaving out the heavy input from inflow rivers and other sources, the effect of algae itself (with short life cycle and quick decomposition) might not that important. On the contrary, the vegetation distribution in East Lake Taihu altered the environment for phosphorus cycling frequently. During the growing phase, roots could uptake sediment phosphorus or secret oxygen to change the phosphorus transformation. While, during the decay phase, partially active OP could be activated into labile and bioavailable forms under certain conditions, and the phosphorus return by the phytodetritus was larger than by algae residues. Therefore, the activation potential of sediment phosphorus in hydrophyte-dominated bays was also large. Although Fe/Al-P was recognized as an indicator of contamination, contamination was not exactly the same as release risk. In hydrophyte-dominated bays/lakes with a relatively lighter TP loading, in particular, the long-run impacts from the activation of partially active phosphorus fractions calls for closer attention. However, how much phosphorus is taken up by hydrophytes? And how much is returned during decaying? All these questions are worth answering.

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CONFLICTS OF INTEREST

All of the authors claim that they do not have any actual or potential conflict of interest, including any financial, personal or other relationships with other people or organizations.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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