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Phosphorus speciation, controlling factors, and ecological risk assessment in lagoon sediments: a case study of the Shamei Lagoon, Qionghai, Hainan

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The Shamei Lagoon in Qionghai, Hainan, is an ecologically significant coastal wetland system that has recently faced increasing ecological risks due to phosphorus (P) pollution. In this study, 22 sediment samples were systematically collected from the main lagoon body, river inlets, and the transitional zone to the open sea. A comprehensive approach incorporating standardized phosphorus fractionation, spatial interpolation, redundancy analysis, and ecological risk assessment was used to investigate P speciation, spatial distribution patterns, controlling factors, and ecological risks. The results showed that: (1) Total phosphorus (TP) concentrations ranged from 261.87 to 875.74 mg/kg, with a mean of 415.72 mg/kg, comparable to typical lagoons in China. Inorganic phosphorus (IP) dominated the P fractions (mean: 317.70 mg/kg), with Fe/Al-bound phosphorus (Fe/Al-P, mean: 183.52 mg/kg) markedly exceeding Ca-bound phosphorus (Ca-P, mean: 105.88 mg/kg). Organic phosphorus (OP) had the lowest contribution (mean: 98.02 mg/kg). (2) The primary sources of P pollution were agricultural runoff and aquaculture waste, industrial discharges and vessel leakage, and regional calcareous deposition. High TP and OP concentrations were concentrated in the southwest river inlet zone, directly linked to fertilizer runoff and tourism-related wastewater. IP levels were elevated in the southern region, driven by hydrodynamic processes. Fe/Al-P was enriched in the central lagoon, likely associated with oil degradation from vessels. Secondary peaks of Ca-P occurred in the southwest aquaculture zone and the northeastern mangrove area, attributed to feed residues and shell debris, respectively. (3) Cation exchange capacity (CEC) was identified as the primary controlling factor for spatial variation in P speciation (explained variance: 46.0%, $p < 0.01$), followed by organic matter (OM), while pH exhibited a marked negative regulatory effect. (4) At 45.5% of sampling sites, TP levels exceeded the ecological safety threshold (600 mg/kg), and 18 sites, mainly located near the river mouth and central lagoon, were classified as heavily polluted, posing a threat to aquatic ecological security. These findings indicate that sediment P pollution in the

Shamei Lagoon is driven by multiple anthropogenic activities. Mitigation strategies should prioritize control of agricultural non-point sources, aquaculture residues, and vessel oil pollution, along with sediment remediation focused on enhancing CEC.

KEYWORDS

lagoon, sediment, phosphorus speciation, spatial distribution, pollution

1 Introduction

Phosphorus (P), an essential nutrient for plant growth, is a key regulator of the carbon cycle. Excessive P input can lead to cascading environmental impacts, disrupting ecological balance. In terrestrial ecosystems, the artificially inputted phosphorus load has become one of the fastest-growing pollution fluxes in many watersheds, directly driving eutrophication in aquatic systems (Hu et al., 2024). In marine environments, P exists in both inorganic (e.g., orthophosphate) and organic forms (e.g., dissolved and particulate organic phosphorus) (Zhang, 2009; Tian, 2018). Coastal sediments serve as major sinks for phosphorus, acting as both filters for overlying waters and potential internal sources of nutrients under specific conditions (Wang et al., 2018). Hence, tracking the dynamic changes in sediment P is vital for both scientific understanding and environmental management.

As a biogeochemically active element (Zhou et al., 2023), phosphorus faces the dual challenges of resource depletion (Mayer et al., 2016) and environmental pollution. The intensification of watershed P pollution reflects its resource scarcity. Phosphorus speciation in watershed systems is highly complex: in the water column, it can be categorized into inorganic phosphorus (IP) and organic phosphorus (OP) based on chemical properties, or into particulate phosphorus (PP) and dissolved phosphorus (DP) based on physical states (Bai et al., 2009). In sediments, phosphorus exists in diverse forms, including exchangeable (Ex-P), Fe-bound (Fe-P), Al-bound (Al-P), Ca-bound (Ca-P), organic (OP), and residual phosphorus (Res-P) (Choi et al., 2008; Liu et al., 2015; Zhu et al., 2023). These different forms and their biogeochemical transformations directly affect water quality. Soluble IP readily fuels aquatic primary production, while OP is recycled through microbial mineralization. However, excessive P input often induces eutrophication, algal blooms, and ecological imbalance. Therefore, understanding the multi-speciation behavior of phosphorus and its environmental implications is crucial for effective water quality protection and pollution control.

Previous researchers have conducted a series of studies on the key physicochemical parameters that affect changes in phosphorus content in sediments. Xu et al. (2014); Li et al. (2019) and Li et al. (2022) pointed out that pH values can affect the transformation and re-release of phosphorus forms in sediments, and low pH values promote the adsorption of phosphorus by sediments. Zhao et al.

(2014) and Han et al. (2020) pointed out that organic matter (OM) can cause changes in the form of phosphorus in sediments, and its total amount, activity, and degree of decomposition control the adsorption and release capacity, rate, and intensity of phosphorus in sediments, respectively. Jin et al. (2006) and Huang et al. (2020) found that the total phosphorus and cation exchange capacity (CEC) of sediments showed a synchronous increase trend, indicating that CEC can be used as an evaluation index for phosphorus enrichment and retention capacity. Wang et al. (2004) and Gao (2011) confirmed that particle size is the key physical constraint for the occurrence and adsorption of phosphorus in sediments. As the particle size of Lagoon sediments becomes finer, the total phosphorus content increases, with the highest content in clay mineral particles. These conclusions point to a close correlation between physical and chemical factors such as pH, OM, CEC, and Clay content on the form and content of phosphorus in sediments.

Located in Bo'ao Town, Qionghai City, Hainan Province, the Shamei Lagoon is a critical integrated wetland ecosystem that provides essential habitats for a wide range of species. In recent years, growing environmental concerns have brought increased attention to the ecological health of the lagoon, underscoring the urgent need for targeted management strategies. As a vital component of coastal wetlands (Cederwall and Elmgren, 1990) the Shamei Lagoon supports rich biodiversity and performs essential ecosystem services, including water purification, coastal defense against storm surges, and sustaining fishery resources. Although lagoon ecosystems provide obvious ecological services, they are inherently fragile and vulnerable to external disturbances. The rapid development of agriculture, aquaculture, and tourism has imposed considerable phosphorus-related environmental pressure on the lagoon, accelerating sediment degradation and triggering a series of ecological disturbances. This study, conducted under the China Geological Survey's Qionghai–Wanning Coastal Zone Integrated Geological Survey Program, focuses on the Shamei Lagoon. By integrating spatial interpolation and redundancy analysis, we explore the spatial distribution patterns of various phosphorus fractions in lagoon sediments, identify key controlling factors, and trace potential sources. The results serve to enhance theoretical understanding of P cycling in lagoon ecosystems and provide practical guidance for nutrient management and pollution mitigation strategies in tropical coastal regions.

2 Material and methods

2.1 Study area and sampling design

The Shamei Lagoon is a shallow, elongated water body separated from the South China Sea by the Yudaitan sandbar. It features gentle slopes and lies adjacent to Wanning City. The Jiuju River and Longgun River discharge into the lagoon from the west and south, respectively. The lagoon is bounded to the north by Dongyu Island and connected to the mouth of the Wanquan River, which flows into the South China Sea. It borders Jinniuling to the south, Nangang to the east, and Beichao Village to the west. The total study area covers approximately 25.87 km².

To capture the heterogeneity of the Shamei Lagoon, a stratified sampling strategy was applied across three representative geomorphological units: (i) the lagoon body (from Donghai Village to Lingxia Village), (ii) the river inlets (e.g., Wanquan River), and (iii) the transitional zone to the open sea. In the lagoon body, a 500 m × 300 m grid-based sampling approach was adopted, focusing on areas influenced by human activities such as tourist docks and vegetable farming zones near Donghai Village. Nine longitudinal transects along a depth gradient (0.5–3.5 m) were established. In the river inlet area, radial sampling lines extended from the estuary center along the main flow direction at 50 m intervals. For the open sea transition zone, arc-shaped transects were placed along the 2 km isobath. Sampling was conducted in the summer of 2021 at a working scale of 1:50,000, resulting in the

collection of 22 sediment samples. The sampling locations are shown in Figure 1.

2.2 Sediment collection and phosphorus fractionation

Sediment cores were collected using a modified stainless-steel gravity corer. The grab sampler was pre-rinsed with local water prior to sampling. At each site, at least 2 kg of wet bottom sediment was collected. Visible debris such as plant matter, stones, shells, and plastics were removed on-site. The sediment was sieved through a 2 mm nylon mesh, retaining at least 1.5 kg of fine material. The sieved sediments were wrapped in 60-mesh nylon fabric, drained as much as possible, and sealed in double-layer ziplock bags. Air was expelled before sealing, and sample identifiers were written on both sides of the bag using an oil-based marker. Each sample was labeled as “SMW-1,” where “SM” denotes the sampling area, “W” indicates sediment, and the number represents the sampling point.

After each sample was collected, the grab sampler and sieve were thoroughly cleaned to prevent cross-contamination. All sediment samples were stored properly to avoid contamination or degradation. During transport, low temperatures and minimal vibrations were maintained to ensure sample integrity and representativeness. *In-situ* environmental parameters such as water temperature, salinity, and pH were also measured using portable instruments.

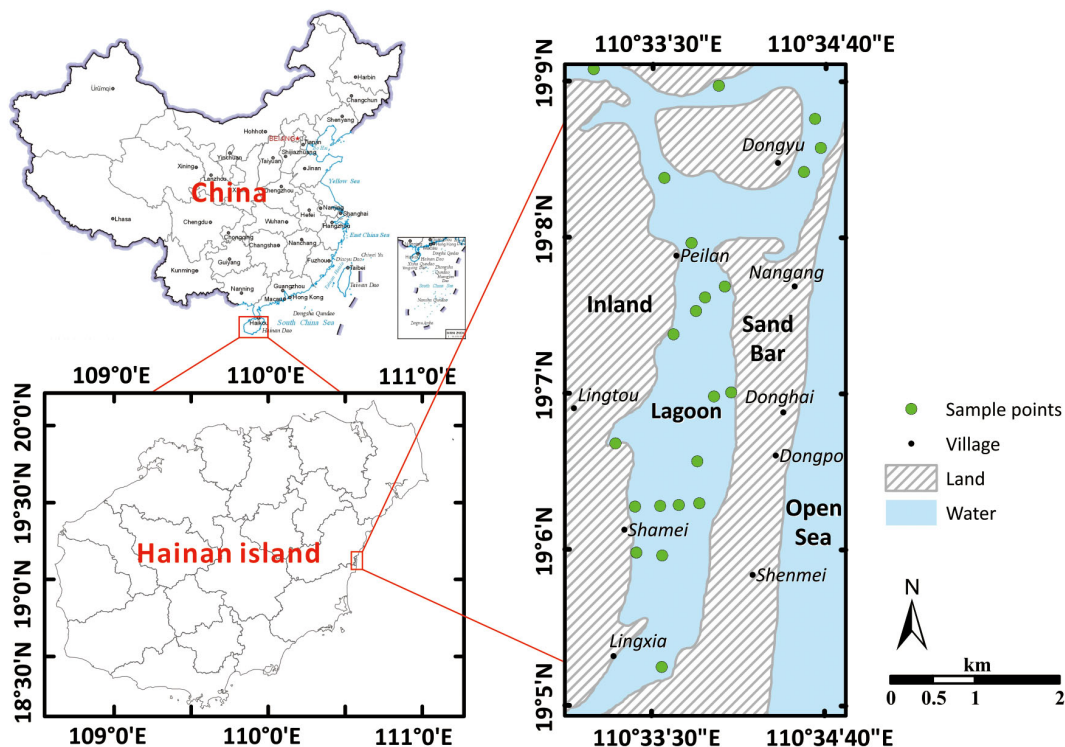


FIGURE 1
Distribution map of sample points in the study area.

Phosphorus speciation was analyzed using the standardized SMT sequential extraction protocol (González Medeiros et al., 2005). Specific procedures were as follows:

Fe/Al-bound Phosphorus (Fe/Al-P): Add 20 mL of 1 mol/L NaOH to 0.2 g of sediment and shake at constant temperature for 16 h. Centrifuge and collect the supernatant (Extract A). Use the remaining residue for Ca-P extraction. To 10 mL of Extract A, add 4 mL of 3.5 mol/L HCl, let it stand at room temperature for 16 h to obtain Extract B. Measure phosphorus concentration in Extract B using the molybdenum antimony blue method.

Ca-bound Phosphorus (Ca-P): Add 20 mL of 1 mol/L HCl to the residue from Fe/Al-P extraction and let stand at room temperature for 16 h. Measure P concentration in the resulting extract (Extract C) using the same method.

Inorganic Phosphorus (IP): Add 20 mL of 1 mol/L HCl to 0.2 g of fresh sediment and let stand at room temperature for 16 h. Measure the phosphorus concentration in the extract; the residue is reserved for OP analysis.

Organic Phosphorus (OP): Transfer the residue from the IP extraction to a porcelain crucible, dry it, and combust in a muffle furnace at 450 °C for 1 h. After cooling, add 20 mL of 1 mol/L HCl and let it stand for 16 h. Measure phosphorus concentration in the extract using the same colorimetric method.

2.3 Statistical analysis

Using SPSS 26.0 and the Kriging interpolation module in ArcGIS 10.8, we developed spatial distribution models for various phosphorus forms and key physicochemical parameters (e.g., pH, cation exchange capacity (CEC), clay content, and organic matter content). Spatial visualization and geostatistical analysis were used to characterize spatial heterogeneity and quantify gradients in sediment properties.

2.4 Environmental quality and pollution assessment

According to the Guidance Manual issued by the United States Environmental Protection Agency (Cieniawski et al., 2002), the potential ecological risk assessment criteria and pollution level grading criteria (Li, 1998; Zhou, 2018) in the Sediment Quality Guidelines developed by the Ontario Department of Environment

and Energy in 1992 were selected to evaluate the environmental quality and pollution level of sediments in the Shamei Lagoon.

The ecological toxicity effects that reflect environmental quality are classified based on TP content, including (1) Safety Level ($TP < 600 \text{ mg} \cdot \text{kg}^{-1}$), which has not yet caused harm to benthic organisms; (2) Lowest Effect Level ($600 \text{ mg} \cdot \text{kg}^{-1} \leq TP < 2000 \text{ mg} \cdot \text{kg}^{-1}$): sediments have been contaminated, but most benthic organisms can tolerate it; (3) Severe Level ($TP \geq 2000 \text{ mg} \cdot \text{kg}^{-1}$): indicates obvious damage to the benthic community. The pollution level is analyzed based on the single factor standard index method (Sun et al., 2021), and the standard index calculation formula is: $S_{TP} = C_{TP}/C_s$, where C_{TP} is the measured content of TP and C_s is the evaluation standard content (235 mg/kg); The evaluation criteria are divided into four levels, including (1) Level 1 ($S_{TP} < 0.5$): Unpolluted; (2) Level 2 ($0.5 \leq S_{TP} < 1.0$): Mild pollution; (3) Level 3 ($1.0 \leq S_{TP} < 1.5$): Moderate pollution; (4) Level 4 ($S_{TP} \geq 1.5$): Severe pollution. Through environmental quality and pollution assessment, the enrichment and pollution level of phosphorus in sediments can be revealed from multiple dimensions such as biological tolerance and chemical cycling, and pollution sources can be identified, providing data reference for improving the ecological environment quality of lagoons.

3 Results

3.1 Spatial distribution of different phosphorus fractions in lagoon sediments

This study systematically quantified the spatial distribution characteristics of phosphorus fractions in the sediments of the Shamei Lagoon. These findings provide essential data for elucidating the biogeochemical cycling of phosphorus and for early warning of potential ecological risks. According to Table 1, the average concentration of total phosphorus (TP) in the lagoon sediments was 415.71 mg/kg, with a maximum of 875.74 mg/kg and a minimum of 261.87 mg/kg. The coefficient of variation (CV) was 0.60. Compared with other regions (Table 2), the TP levels in the Shamei Lagoon are generally comparable to those reported in other domestic lagoons. The average content of inorganic phosphorus (IP) was 317.70 mg/kg, which was markedly higher than that of organic phosphorus (OP), with an average of 98.02 mg/kg. Among the inorganic forms, both Fe/Al-bound phosphorus (Fe/Al-P) and calcium-bound phosphorus (Ca-P) exhibited similar

TABLE 1 Statistical description of phosphorus forms in Shamei Lagoon sediments.

Indicators	TP (mg/kg)	Fe/Al-P (mg/kg)	Ca-P (mg/kg)	IP (mg/kg)	OP (mg/kg)
Maximum	875.74	502.99	246.50	679.03	196.71
Minimum	261.87	2.40	15.96	55.97	26.75
Mean	415.71	183.52	105.88	317.70	98.02
Standard deviation	254.16	158.90	61.00	218.51	56.21
Coefficient of variation	0.60	0.87	0.58	0.69	0.59

TABLE 2 Comparison of the range and average phosphorus content in Shamei Lagoon sediment with other domestic sea areas.

Study area	TP (mg/kg)	Sample time	References
Shamei Lagoon	261.87~875.74 (415.72)	2021	this paper
Qilihai Lagoon	250-470	2015/2019	Zhou (2018); Sun et al. (2021)
Erhai Lake	420-1750(900)	2011	Liu (2011); Huang (2012)
Dianchi Lake	300~5900(270)	2014	Meng et al. (2015); Li (2015)
Nan'ao mariculture areas	370-870(470)	2009-2010	Mao et al. (2012); Chen et al. (2011)
Western Guangdong coasts	240	2006-2007	Gu et al. (2010)
Yangtze Estuary	410	2015	Fan et al. (2017)
Jiaozhou bay	600-1500(1200)	2017	Lei et al. (2021)
Central Bohai Sea	100-400(300)	2015	Zhong et al. (2020)
Dongshan Bay of Fujian	500-740(630)	2016	Wang et al. (2019)
Daya Bay	330-470(430)	2006	Gu (2009)
Zhelin Bay	200-600(500)	2008	Feng (2011)
Guishan Island offshore	300-1900(800)	2009	Feng (2011)
South Yellow Sea	240	2007	Han et al. (2017)

distribution patterns, with average concentrations of 183.52 mg/kg and 105.88 mg/kg, respectively. The coefficients of variation for Fe/Al-P, Ca-P, IP, and OP were 0.87, 0.58, 0.69, and 0.59, respectively, indicating relatively high spatial variability in both the forms and concentrations of phosphorus across sampling sites.

Spatially, TP was primarily concentrated in the southwestern riverine inflow area (Figure 2A), gradually decreasing from south to north, forming a tiered distribution pattern. The spatial distribution of OP largely mirrored that of TP, with high-value zones overlapping (Figures 2A, B). IP showed a partially similar pattern to TP (Figure 2C), with its highest concentrations located in the southern river inflow zones and declining northward. Fe/Al-P exhibited high concentrations (256.98–265.43 mg/kg) mainly in the central part of the lagoon (Figure 2D), with decreasing levels toward both the southern and northern ends. Ca-P displayed apparent high-value areas in the southwestern river inflow region and along the lagoon's shoreline (173.92–213.86 mg/kg) (Figure 2E), with elevated values also observed near the northeastern estuarine outlet. Overall, Fe/Al-bound phosphorus was markedly more abundant than Ca-bound phosphorus, indicating that inorganic phosphorus in the sediments predominantly exists in the form of Fe/Al-P.

3.2 Coupling relationships between phosphorus fractions and environmental factors in lagoon sediments

In the sediments of Shamei Lagoon in Hainan, the spatial distribution of phosphorus fractions is closely associated with several physicochemical parameters, including pH, organic matter (OM) content, cation exchange capacity (CEC), and clay content. To better understand how these environmental variables influence phosphorus speciation in sediments, a correlation analysis was conducted, and the results are summarized in Table 3.

According to Table 3: (1) pH as a Regulatory Factor for Phosphorus. Overall, pH exhibited a negative correlation with total phosphorus (TP) in the lagoon sediments ($r = -0.41$), indicating that lower pH values are associated with higher TP concentrations. Among the different phosphorus forms, Fe/Al-bound phosphorus (Fe/Al-P) showed a significant negative correlation with pH.

(2) Bidirectional Role of Organic Matter (OM). At the whole-system level, organic matter content displayed a strong and significant positive correlation with total phosphorus, suggesting that OM generally promotes phosphorus accumulation in this lagoon environment. When examining specific phosphorus fractions, OM exhibited strong positive correlations with Fe/Al-P ($r = 0.72$), inorganic phosphorus (IP) ($r = 0.60$), and organic phosphorus (OP) ($r = 0.64$), all of which were highly significant.

(3) Phosphorus Retention Effect of CEC. CEC showed highly significant positive correlations with all phosphorus fractions. Notably, the correlation with total phosphorus exceeded 0.9. The spatial distribution patterns of CEC and OM were similar, and the correlation between them was also strong ($r = 0.9$), indicating their combined influence on phosphorus dynamics.

(4) Clay Content and Phosphorus Forms. Clay content in the sediments demonstrated moderate positive correlations with TP, Fe/Al-P, IP, and OP, while its correlation with Ca-bound phosphorus (Ca-P) was relatively weak but still positive.

As illustrated in Figure 3, total phosphorus exhibited significant correlations with all other phosphorus fractions, with correlation coefficients of 0.81 (OP), 0.84 (IP), and 0.80 (Ca-P), respectively. Organic phosphorus also showed strong positive correlations with Ca-P ($r = 0.71$) and IP ($r = 0.81$). In addition, Fe/Al-P was moderately positively correlated with both IP ($r = 0.65$) and Ca-P ($r = 0.55$).

3.3 Redundancy analysis of phosphorus fractions in lagoon sediments

To further elucidate the relationship between the spatial variation of phosphorus fractions in sediments and environmental factors (Ren et al., 2018; Wang et al., 2021), a redundancy analysis (RDA) was conducted. In this analysis, the

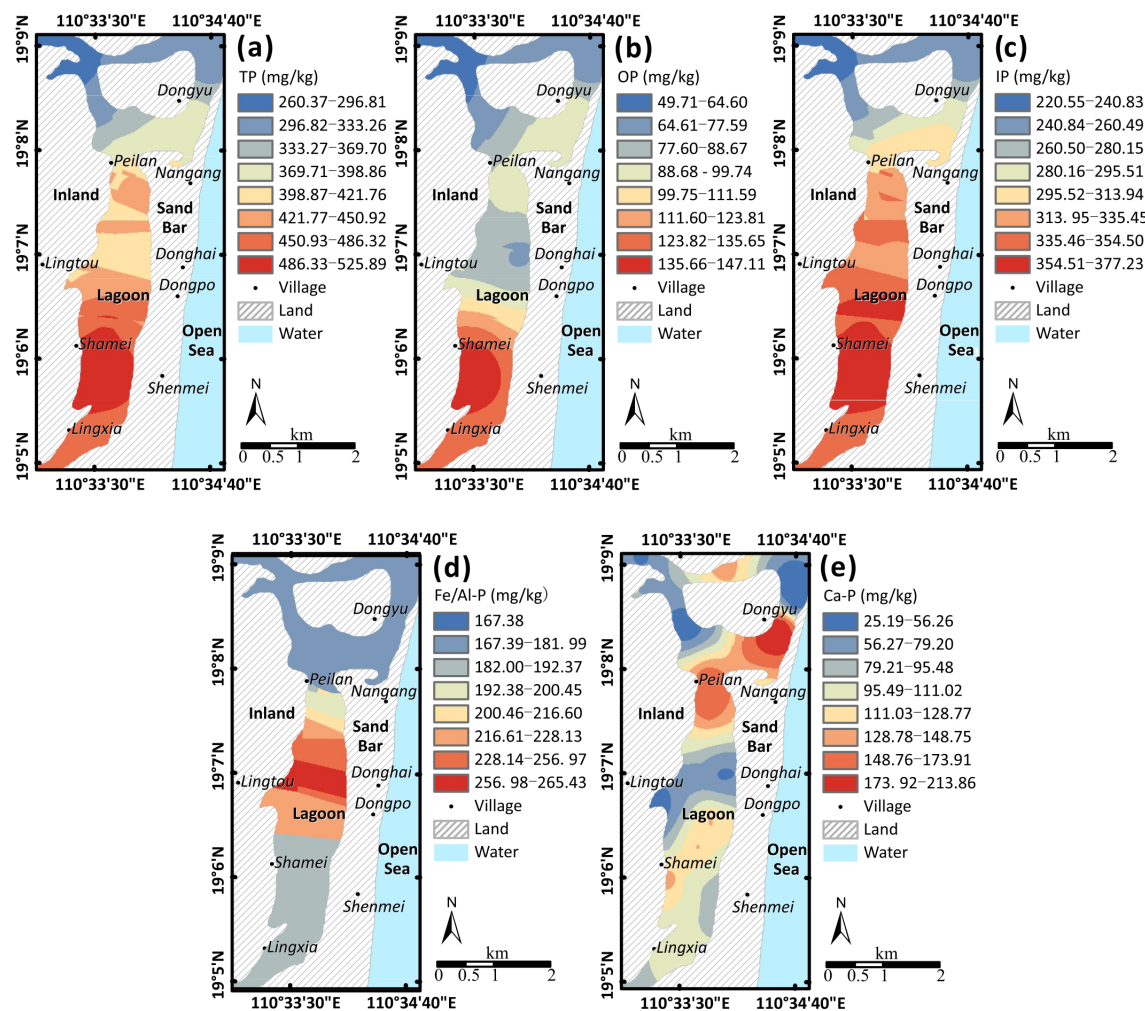


FIGURE 2 Spatial distribution of phosphorus forms. (A) Spatial distribution of total phosphorus (TP); (B) Spatial distribution of organic phosphorus (OP); (C) Spatial distribution of inorganic phosphorus (IP); (D) Spatial distribution of Fe/Al-bound phosphorus (Fe/Al-P); (E) Spatial distribution of Calcium-bound phosphorus (Ca-P).

contents of total phosphorus (TP), organic phosphorus (OP), inorganic phosphorus (IP), Fe/Al-bound phosphorus (Fe/Al-P), and calcium-bound phosphorus (Ca-P) were selected as response variables, while clay content, organic matter (OM), pH, and cation

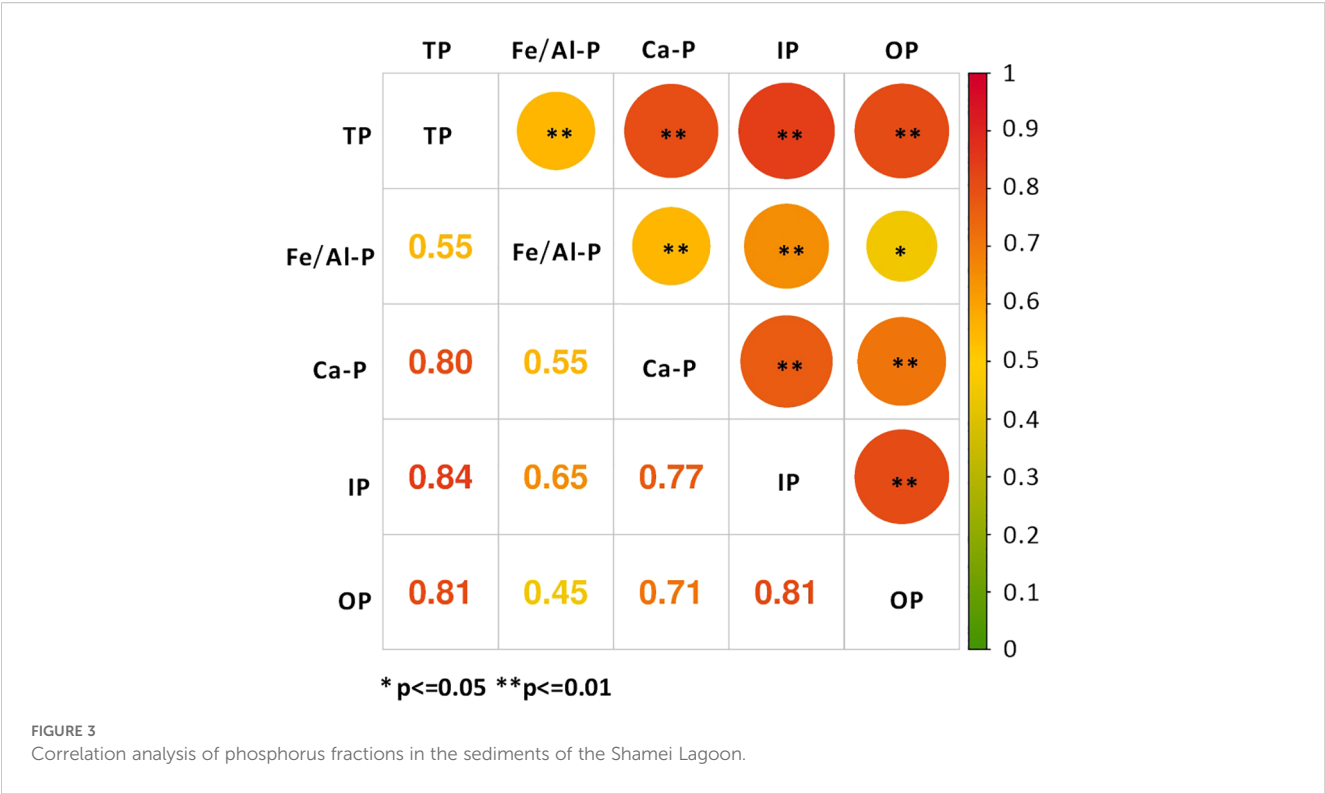
TABLE 3 Correlation between basic chemical properties and phosphorus fractions in lagoon sediments.

Phosphorus forms	pH	OM	CEC	Clay
Fe/Al-P	-0.53*	0.72**	0.68**	0.33
Ca-P	-0.19	0.30	0.54**	0.29
IP	-0.18	0.60**	0.70**	0.32
OP	-0.27	0.64**	0.72**	0.42
TP	-0.41	0.89**	0.90**	0.45*

*At the 0.05 level (two-tailed), the correlation is significant. **At the 0.01 level (two-tailed), the correlation is significant.

exchange capacity (CEC) were used as explanatory variables. Results from the Detrended Correspondence Analysis (DCA) showed that the gradient length of the first axis was 1.2, indicating a linear response model, thus supporting the use of RDA. The RDA results (Figure 4) revealed that the first two axes explained 47.18% and 5.68% of the total variance in phosphorus content, respectively. Collectively, 54.1% of the variation in phosphorus fractions in the sediments could be attributed to the selected environmental factors, suggesting that these factors exert an obvious influence on phosphorus distribution.

As shown in Table 4, among all the environmental factors, only cation exchange capacity (CEC) had a statistically significant effect ($P < 0.05$). According to Figure 4, Ca-P and IP showed relatively minor distributional differences. All phosphorus fractions were positively correlated with OM, CEC, and clay content, and negatively correlated with pH. In terms of explanatory power, the ranking of environmental variables was as follows: CEC (46.0%), OM (6.1%), pH (1.6%), and clay (0.4%). The corresponding



contribution rates were: CEC (85.0%), OM (11.3%), pH (2.9%), and clay (0.8%) (Table 4). “Safety Level” and the “Lowest Effect Level (LEL)”, indicating potential ecological risk.

3.4 Sediment environmental quality

According to the Sediment Quality Guidelines developed by Ontario Department of Environment and Energy, 1992, the ecological toxicity effect grading (Li, 1998; Zhou, 2018) indicates a threshold value for total phosphorus (TP) is 600 mg/kg. In the study area, TP concentrations in lagoon sediments ranged from 261.87 to 875.74 mg/kg. The lowest ecological toxicity effect level for TP falls within the range of 600~2000 mg/kg. As shown in Table 5, the TP concentrations in the sediments of Shamei Lagoon lie between the

3.5 Sediment pollution index

The pollution assessment index (S_{TP}) of phosphorus in the sediment of Shamei Lagoon was calculated based on the single factor pollution level evaluation model. According to the pollution level grading standards, <0.5 corresponds to level 1, 0.5~1.0 corresponds to level 2, 1.0~1.5 corresponds to level 3, and ≥ 1.5 corresponds to level 4, with pollution increasing in order. As shown in Table 6, the TP pollution index exceeded the standard threshold at all sampling sites, with values ranging from 1.11 to 4.70,

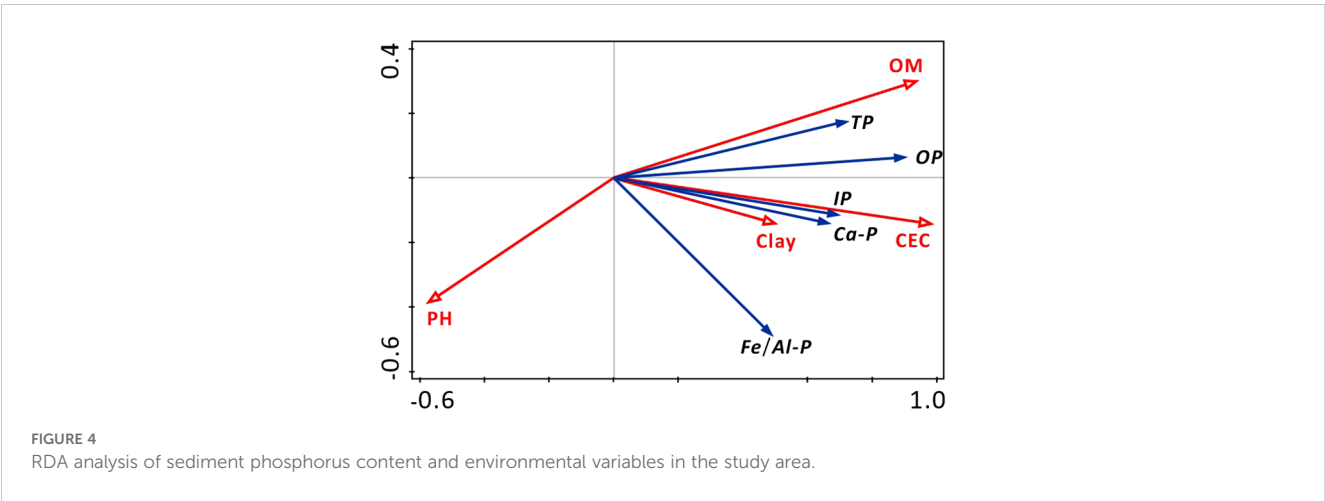


TABLE 4 Redundancy analysis (RDA) results between sediment phosphorus content and environmental variables in the study area.

Environmental factors	Explains (%)	Contribution (%)	Pseudo-F	P
CEC	46.0	85.0	17.0	0.004
OM	6.1	11.3	2.4	0.110
pH	1.6	2.9	0.6	0.530
Clay	0.4	0.8	0.2	0.920

indicating severe phosphorus contamination. Specifically, sampling sites SMW08, SMW11, SMW12, and SMW20 had S_{TP} values of 1.11, 1.46, 1.48, and 1.40, respectively, which classified as moderately polluted. The remaining 18 sites exhibited S_{TP} values exceeding 1.5, indicating heavy pollution.

4 Discussion

Phosphorus is a key factor regulating the growth and reproduction of aquatic organisms. Its speciation and spatial heterogeneity in lagoon sediments serve as core indicators for

evaluating ecosystem nutrient status and primary productivity. Monitoring data from the past decade indicate that, with the continuous increase in land-based nutrient inputs, surface sediments in many lagoons have exhibited obvious phosphorus enrichment.

4.1 Distribution and source analysis of phosphorus fractions

Statistical analysis (Table 1) reveals that phosphorus in the sediments of Shamei Lagoon is predominantly present in inorganic

TABLE 5 Standard index of phosphorus in lagoon sediments.

Sample point	TP	Sample point	TP	Sample point	TP	Sample point	TP
SMW08	Safety	SMW14	Safety	SMW20	Safety	SMW26	Lowest
SMW09	Lowest	SMW15	Lowest	SMW21	Lowest	SMW27	Lowest
SMW10	Lowest	SMW16	Lowest	SMW22	Lowest	SMW28	Lowest
SMW11	Safety	SMW17	Lowest	SMW23	Lowest	SMW31	Safety
SMW12	Safety	SMW18	Safety	SMW24	Lowest		
SMW13	Lowest	SMW19	Safety	SMW25	Lowest		

TABLE 6 Comprehensive pollution index of phosphorus in lagoon sediments.

Sample point	Evaluation index		Sample point	Evaluation index	
	S_{TP}	Level		S_{TP}	Level
SMW08	1.11	3	SMW19	2.30	4
SMW09	4.70	4	SMW20	1.40	3
SMW10	4.28	4	SMW21	4.01	4
SMW11	1.46	3	SMW22	3.15	4
SMW12	1.48	3	SMW23	4.50	4
SMW13	3.06	4	SMW24	4.64	4
SMW14	2.51	4	SMW25	2.87	4
SMW15	4.16	4	SMW26	4.23	4
SMW16	3.65	4	SMW27	4.41	4
SMW17	3.00	4	SMW28	4.50	4
SMW18	2.48	4	SMW31	1.79	4

forms, with substantial variation in both content and fraction among samples. This variation may be attributed to differences in sediment sources, depositional environments, and biological activity. Meanwhile, the dominance of the Fe/Al-P fraction within IP implies that a considerable portion of the sediment phosphorus is both potentially bioavailable and susceptible to release should redox conditions are reduced. Spatially (Figure 2), total phosphorus (TP) is concentrated near river inflow zones, which is directly associated with intensive agricultural activities in the Lingxia Village watershed. The migration of inorganic phosphorus (IP) is obviously influenced by hydrodynamics, consistent with its dissolved form and high mobility. The elevated levels of Fe/Al-bound phosphorus (Fe/Al-P) in the central lagoon correspond to frequent vessel activity, likely originating from the decomposition of organic phosphorus in ship-borne oil contamination. In contrast, calcium-bound phosphorus (Ca-P) is concentrated at river and estuary mouths and alongshore zones, potentially due to waterborne transport and aquaculture-related inputs.

By comparing the content and spatial distribution of various phosphorus fractions in the sediments and combining these with spatial patterns of anthropogenic activities, the sources of phosphorus pollution in the study area can be categorized into three groups:

(1)TP-OP-Ca-P Group: The co-enrichment of TP, organic phosphorus (OP), and Ca-P in the vicinities of Shamei and Lingxia Villages suggests strong correlations with agricultural (Lin et al., 2011) and aquacultural activities. The TP hotspots (southwestern inflow area) show strong spatial overlap with farmland distribution in Lingxia Village ($r = 0.89$), mainly due to runoff containing phosphate fertilizers (e.g., superphosphate). Ca-P accumulation (>150 mg/kg) is linked to the deposition of calcium carbonate-rich feed residues from aquaculture zones near Donghai Village. Additionally, OP enrichment near the Lagoon entrance (mean: 98.02 mg/kg) may be related to discharge of phospholipid-rich wastewater from tourism and food service operations.

(2)IP-Fe/Al-P Group: The co-accumulation of IP and Fe/Al-P in the lagoon center reflects the interaction of industrial activities and natural processes. The Fe/Al-P hotspot (central lagoon, mean: 156.15 mg/kg) overlaps with areas of diesel leakage from vessels. Here, sulfides from the oil combine with iron/aluminum oxides in the sediments to form short-lived complexes, setting the stage for subsequent reactions in reducing environments where sulfide (S^{2-}) reacts with iron to form iron sulfide (FeS) (Hao et al., 2019). This process dissolves iron oxides, thereby releasing the associated phosphorus (Fe/Al-P). The accumulation of IP in the central lagoon (>300 mg/kg) is related to the discharge of wastewater from upstream industrial areas. Shamei Lagoon is located at the mouth of Wanquan River in Hainan Island (Zhang et al., 2011), and belongs to a semi enclosed tidal channel lagoon. In recent years, it has gradually weakened under the influence of offshore tidal dynamics (Zhang, 2024). Upstream rubber factories use phospholipids during the processing, which, along with the phosphate ions carried by natural rubber itself, are discharged

into the surface runoff and migrate and diffuse to the lagoon. As the hydrodynamic conditions gradually weaken, they deposit in the center of the lagoon.

(3)Independent Ca-P Group: The isolated Ca-P hotspots in the northern lagoon suggest a regionally specific source. High concentrations of Ca-P at the northeastern estuarine outlet (173.90–213.86 mg/kg) may result from calcareous shell debris (Zong et al., 2025) associated with the surrounding mangrove zones, while lower concentrations near the Wanquan River mouth (<50 mg/kg) are likely due to the weaker adsorption capacity of sandy sediments.

4.2 Mechanisms of phosphorus fraction influences by environmental factors and other fractions

Correlation analysis between phosphorus fractions and environmental parameters (Table 3) suggests the following mechanisms:

1. Inverse Regulation by pH. In sediments overlain by low-pH water, hydroxide ions (OH^-) are more readily displaced by phosphate anions, increasing phosphorus concentrations in surface sediments. The most significant negative correlation was observed between pH and Fe/Al-P. Acidic conditions enhance the activity of iron and aluminum compounds, and displaced OH^- is quickly neutralized. Additionally, lower pH enhances the anion exchange and adsorption capacity of soils.
2. Bidirectional Effects of Organic Matter (OM). OM was significantly correlated with all phosphorus fractions except Ca-P, as OM plays a key role in phosphorus adsorption (Wang, 2017). Depending on conditions, OM can either enhance or inhibit phosphorus fixation. According to the study by Tang et al. (2023), the study area is a river-dominated brackish lagoon with a salinity of approximately 2.96‰. This salinity level is close to that of normal seawater. Under such conditions, the competitive effect of anions (e.g., SO_4^{2-}) and sulfate reduction weaken the adsorption and immobilization capacity of iron-bound phosphorus, thereby increasing its release risk. Additionally, sandbars developed at the estuary of the study area, primarily composed of grayish-yellow and yellow-white sandstone, along with dark gray organic-rich clay and silty sand. These materials can serve as mineral sources for lagoon sediments under the influence of river flow and wind. Within the dynamic redox environment of the lagoon, fresh and highly reactive iron oxides are continuously generated and combine with organic matter to form protected complexes. The pH in most parts of the study area ranges from slightly acidic to neutral, a range in which phosphorus predominantly exists as highly reactive $H_2PO_4^-$. Consequently, the bridges formed between

organic matter and Fe/Al in the study area are stable and efficient. Thus, pH and iron oxides in the sediments enhance the adsorption and immobilization of phosphorus by organic matter (OM), while salinity exerts an inhibitory effect.

3. The indirect effect of cation exchange capacity (CEC) on phosphorus. The correlation analysis results indicate a strong positive correlation between cation exchange capacity (CEC) and various phosphorus fractions, as well as total phosphorus (TP), with the correlation coefficient for TP exceeding 0.9 (Table 3). However, given that CEC primarily interacts with cations, while phosphorus predominantly exists in the environment as anions, this strong correlation may not reflect a direct causal relationship. Further analysis reveals that the spatial distribution of CEC closely aligns with that of organic matter (OM) ($r = 0.9$), suggesting that CEC in the study area is primarily derived from organic matter. Therefore, the co-variation between CEC and phosphorus is more likely jointly driven by organic matter: OM not only provides abundant negative charges that enhance CEC but also directly or indirectly participates in the immobilization and cycling of phosphorus through the formation of organic phosphorus, influence on microbial activity, and complexation with metal oxides.
4. Positive Feedback from Clay Content. Clay content partially determines the distribution of phosphorus fractions in sediments. The high clay content and water retention in Shamei Lagoon result in heavy-textured sediments, which reduce the diffusion efficiency of phosphate. Additionally, the large specific surface area of clay promotes phosphate-particle interactions during diffusion (Wang, 2017).

Phosphorus fractions are also influenced by one another (Figure 3). Correlation results indicate that OP, IP, and Ca-P collectively constitute the main forms of TP in the sediments. The distributions of OP, IP, and Ca-P are synergistic, likely due to shared sources or similar environmental conditions. Fe/Al P appears to participate in the transformation of IP and Ca-P, suggesting its pivotal role in the biogeochemical cycling of phosphorus. The strong inter-fractional relationships imply that phosphorus in lagoon sediments does not exist in isolation, but rather in a dynamic equilibrium regulated by mineral adsorption, OM complexation, and redox processes. Understanding these interactions deepens insight into phosphorus behavior and its environmental cycling mechanisms.

4.3 Dominant factors influencing the distribution of phosphorus fractions

Redundancy analysis results (Table 4, Figure 4) indicate that among the four explanatory variables: clay, OM, pH, and CEC, CEC

clearly plays a dominant role, which may be attributed to the multiple physicochemical properties of the sediments. As a parameter reflecting the ion exchange capacity of sediments, CEC is fundamentally influenced by the composition of the sediments themselves. Generally, soils with higher clay particle or OM content tend to exhibit higher CEC (Zhang et al., 2009).

Clay particles adsorb and immobilize phosphorus through mechanisms such as ligand exchange or electrostatic attraction. Although the clay particle content in the study area's sediments is not low, the RDA results suggest that their contribution to phosphorus adsorption is limited. This may be due to the type of clay minerals present, as different clay components exhibit varying capacities for phosphorus absorption and fixation. For instance, the theoretical saturated adsorption capacity of kaolinite for phosphorus is 544 mg/kg, while that of montmorillonite can reach as high as 1681 mg/kg (Yuan et al., 2004). Organic colloids formed during the decomposition of OM can enhance the phosphorus adsorption capacity of sediments. In this study, OM contributed to phosphorus adsorption; however, due to its relatively low content in the sediments, it could not serve as the primary controlling factor promoting phosphorus adsorption (Zhao et al., 2014). pH exerts a macro-regulatory effect on the entire system by influencing the charge state of adsorption surfaces and the chemical speciation of phosphorus. The pH of sediments in the study area predominantly ranges from slightly acidic to neutral. Under acidic conditions, iron and aluminum oxides exhibit stronger adsorption affinity for phosphorus. Moreover, the influence of pH on variable charge surfaces may already be reflected in the measured CEC values. While CEC itself is a pH-dependent indicator, when measured at a fixed pH, it reflects the intrinsic properties of the sediments.

In this study, the relatively low OM content limited its role in phosphorus adsorption and fixation. Although the clay content was high, variations in its mineral composition likely led to a reduced contribution of clay to phosphorus adsorption. In contrast, CEC, which is governed by both clay and OM content, emerged as the primary controlling factor promoting phosphorus absorption in the study area.

4.4 Sediment environmental quality and pollution assessment in Shamei Lagoon

Based on the results of the sediment environmental quality assessment level (Table 5), the sediment at some sampling points in the Shamei Lagoon has been contaminated to a certain extent. Even though this level is explained in the ecological risk assessment standards as “Lowest Effect Level ($600 \text{ mg} \cdot \text{kg}^{-1} \leq \text{TP} < 2000 \text{ mg} \cdot \text{kg}^{-1}$): sediments have been contaminated, but most benthic organisms can tolerate it”, the total phosphorus content poses a confirmed ecological risk to the environment and cannot be ignored. The pollution index results (Table 6) confirm the presence of phosphorus pollution in specific regions of the lagoon. Therefore, immediate actions are required to control pollution at its source, particularly by reducing phosphorus nutrient inflows. Such efforts are essential to prevent further deterioration of water quality,

control eutrophication risks, protect benthic habitats, and ultimately improve the overall ecological status of the lagoon.

5 Conclusions

This study focused on the Shamei Lagoon in Hainan Province and systematically investigated the occurrence characteristics of phosphorus speciation in lagoon sediments, assessed the dominant controlling factors and ecological risks, and reached the following conclusions. In the sediments of the Shamei Lagoon: (1) The total phosphorus (TP) content is comparable to levels observed in most regions of China, primarily existing in inorganic forms, with Fe/Al-bound phosphorus and Ca-bound phosphorus being dominant; (2) The spatial distribution pattern of phosphorus is closely related to multiple factors, including sediment sources, environmental conditions, and biological activities, and is mainly influenced by coastal agricultural cultivation, hydrological transport, aquaculture operations, vessel fuel leakage, and domestic sewage discharge from tourism areas; (3) Cation exchange capacity (CEC) is the primary factor controlling the spatial differentiation of phosphorus speciation (explained variance: 46.0%, $p < 0.01$), followed by organic matter, while pH exhibits a significant negative regulatory effect; (4) Sediment quality and pollution index assessments indicate that total phosphorus levels exceed the standard across the board, with 18 sampling sites, mainly located at river inlets and the lagoon center, classified as heavily polluted, and phosphorus pollution in some areas approaching or reaching hazardous levels.

Based on the above findings, the following measures are proposed from the perspectives of pollution prevention and environmental management.

1. Reduce phosphorus emissions at the source. For example, promote the use of organic fertilizers instead of some chemical fertilizers in Shamei Village and Lingxia Village, avoiding fertilization during the peak runoff period of the rainy season. At the same time, set up zeolite filled ecological ditches at the end of irrigation channels and plant root developed plants such as reeds at the entrance of the Lagoon to reduce and intercept phosphorus pollution from agricultural fertilizers flowing into the lagoon. Promote the 3D aquaculture model of symbiotic between submerged plants and shellfish in the aquaculture area at the mouth of Wanquan River, and set up calcium magnesium modified biochar filter beds in the feeding area to adsorb lipid bound phosphorus in residual feed. Mandatory use of P free detergents in tourist hotspots, laying artificial gravel at cruise ship docks, and utilizing biofilm on the surface of gravel to degrade lipid containing organic matter. Restricting the entry of old and high-risk oil spill vessels into lagoons, etc.
2. Improve the efficiency of pollutant treatment. On the one hand, the government should help industrial and agricultural enterprises understand relevant systems and establish the concept of combining efficiency with environmental protection. On the other hand, factories

should actively improve the efficiency of phosphorus treatment in wastewater by developing and applying advanced phosphorus treatment technologies, such as diversion scheduling, sediment dredging, and physical algae removal.

3. Ecological restoration and reconstruction. For example, artificial replanting of mangrove forests and seagrass vegetation communities for wetland reconstruction, regulating algae and fish communities through biological manipulation techniques, restoring ecosystem service functions, and controlling the degree of eutrophication in water bodies.
4. Establish a long-term monitoring and early warning system. Improve the construction of a lagoon environmental monitoring network in areas where human activities have an apparent impact, such as docks, central islands, and entrances to the Lagoon. Strengthen the quality supervision and management of environmental monitoring data, enhance environmental monitoring capacity building, establish an intelligent warning system, achieve accurate warning and rapid intervention, and prevent pollution problems from worsening.

Data availability statement

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to Kun Yuan, yk523057208@163.com.

Author contributions

KY: Conceptualization, Formal Analysis, Funding acquisition, Methodology, Writing – original draft, Writing – review & editing. XM: Methodology, Visualization, Writing – original draft. GX: Methodology, Visualization, Writing – original draft. KL: Visualization, Writing – original draft. WZ: Visualization, Writing – original draft. RW: Supervision, Writing – review & editing. SW: Supervision, Writing – review & editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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