



Organic phosphorus regeneration enhanced since eutrophication occurred in the sub-deep reservoir[☆]

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ABSTRACT

Lake eutrophication remains a serious environmental problem of global significance, and phosphorus (P) plays a key role in lake eutrophication. Internal P loading, as a result of P release from sediments, is gathering more and more recognition as an important source governing the P availability in these ecosystems. Anoxic condition can promote the release of P associated with Fe oxides, which has already been a consensus. However, it is still unknown whether the anoxic conditions induced by eutrophication act to intensify or weaken the regeneration of organic P (P_{org}) in sediments. We selected the Hongfeng Reservoir, a typical sub-deep lake, to study the regeneration behaviours of C and P in the sediments buried before and after eutrophication. The results showed that P_{org} did not significantly increase with the rapid increase in organic C (C_{org}) since eutrophication occurred. Furthermore, the organic C/P ratio was much higher in sediments buried after eutrophication than in those buried before, which indicated that P_{org} regeneration had been significantly enhanced since eutrophication occurred. Based on C/P ratios, our estimation suggested that the P_{org} regeneration and P release from sediment to water approximately enhanced $45.2\% \pm 8.7\%$ and $34.5\% \pm 9.8\%$, respectively. Elevated primary productivity (algae) and the corresponding hypoxic/anoxic condition, both caused by eutrophication, promoted P biogeochemical cycle in the sub-deep reservoir. This study further verifies the significant contribution of regenerated P_{org} to the internal P load, and highlights the importance of controlling P release from sediments in order to restore clear water ecosystems in sub-deep lakes or reservoirs.

1. Introduction

Lake eutrophication is a serious environmental problem of global significance. In most freshwater lakes, phosphorus (P) is a key limiting nutrient, and algal productivity is directly linked to its availability (Qin, 2009; Schindler et al., 2008; Zhu et al., 2013). As such, the reduction in P loading is a promising strategy for controlling eutrophication in lake ecosystems. Despite significant reduction in external P loading, P concentrations have not noticeably declined in many lakes (Søndergaard et al., 1999; Zhang et al., 2017). This is mostly attributed to internal P release from sediments (Burger et al., 2007; Elsbury et al., 2009; Paytan et al., 2017; Qin, 2009; Rockwell et al., 2005; Søndergaard et al., 2003, 2013). Fe oxides reductive dissolution (Chen et al., 2018b; Ding et al., 2016; Katsev, 2016; Wang et al., 2016; Yuan et al., 2020) and organic matter decomposition (Joshi et al., 2015; Kwak et al., 2018; Li et al., 2018; Ni et al., 2019; Xie et al., 2019; Zhang et al., 2021) both have a

profound influence on P exchange across the sediment-water interface in coastal environments and freshwater lakes.

One serious consequence of eutrophication is the decrease of dissolved oxygen (DO) of lake bottom water (Diaz and Rosenberg, 2008). DO of lake water and sediments are closely linked with trophic state (Moosmann et al., 2006). High primary production leads to high DO consumption in lake water. DO levels in the bottom water of lake can affect the oxygen penetration depth in sediments, thus plays a key role in the sedimentary P recycling (Conley et al., 2009; Wang et al., 2016; Yu et al., 2019). When the bottom water is oxic, P released from OM degradation is potentially trapped by Fe oxides in the upper portions of the sediment. However, when the bottom water becomes anoxic, P released from OM degradation and liberated from Fe oxides via reduction diffuses easily to the overlying water.

One another consequence of eutrophication mentioned here is the change of OM source in sediments. Both content and proportion of

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autochthonous OM (phytoplankton) increase as a result of eutrophication. The autochthonous OM is relatively easy to decompose (Burdige, 2007), and release C, N and P (Zhang et al., 2021). Organic C burial efficiency also depends on the oxygen exposure time, and anoxia favors organic C preservation (Sobek et al., 2009). In this respect, organic P may be coupled to preserve in anoxic environments. For example, Stratford et al. (2000) assumed that the remineralization of both organic C and P was slower under anoxic conditions compared to oxic conditions in the model calculation of nutrient cycling in sapropel times in the eastern Mediterranean Sea. Whereas some investigations in marine environments revealed that hypoxia/anoxia of bottom water reduced the burial efficiency of organic P and promoted the burial of organic C (Ingall et al., 1993; Knauer et al., 1979; Krom and Berner, 1981; Viktorsson et al., 2013a, 2013b). As eutrophication occurs in lakes and reservoirs, the sediment-water interface always becomes hypoxic/anoxic in summer, especially for the sub-deep lakes and reservoirs (Steinsberger et al., 2020). However, it is still unknown whether the anoxic conditions induced by eutrophication intensified or weakened P_{org} regeneration in sediments.

Decomposition of OM continues after deposition onto lake bottom sediments. However, the decomposition rate decays exponentially, and most of the C_{org} mineralization occurs within 5 years after deposition (Galman et al., 2008). After that, the decomposition rate is very low. Thus, C_{org} accumulation rates derived from the sediment core, except the surficial sediments, can faithfully record the changing burial rates (Anderson et al., 2014). Therefore, the comparison of the organic C/P ratio in the sediments buried before and after eutrophication may provide valuable information on the variation of P_{org} regeneration degree, as C and P are both important component of OM.

There are more than 98,000 reservoirs in China, including almost half of the large reservoirs worldwide (www.mwr.gov.cn). Reservoirs are the main utilisation form of surface water resources in China. They have a total storage capacity of 794 km³, which is triple that of natural lakes (Yang and Lu, 2014). Most reservoirs are classified as sub-deep lakes. Although there is no explicit definition of a 'sub-deep lake', they generally have an average water depth of 10–50 m, and differ from 'shallow lakes' in the presence of a thermocline during summer. They are distinguished from 'deep lakes' by the existence of seasonal convective mixing between the hypolimnion and epilimnion, as most deep lakes exhibit permanent thermal stratification (Chen et al., 2018a). Sub-deep lakes are assumed to be the most susceptible to eutrophication. Furthermore, they are too deep to be protected by macrophytes and too shallow to mitigate internal P loading through dilution in the permanent hypolimnion, which makes their restoration more difficult (Chen et al., 2018a; Genkai-Kato and Carpenter, 2005). In addition, the sediment-based O₂ consumption by OM, and release of reduced substances, inhibit the restoration of hypolimnetic O₂ concentrations in sub-deep lakes. As such, following a reduction in the total phosphorus (TP) load, hypolimnetic O₂ concentrations increase at a slower rate compared to those in much deep lakes (more than ~100 m) where the sedimentary OM is much more refractory (Steinsberger et al., 2020). Studies focused on the internal P cycle of sub-deep lakes are relatively scarce (Wang et al., 2012, 2021). Therefore, understanding the mechanisms that govern the internal P cycle is crucial for the prevention and control of eutrophication in sub-deep lakes.

The Hongfeng Reservoir, constructed in 1960, is a typical freshwater sub-deep lake. It underwent the transition from an oligotrophic state to a eutrophic state, and the sediment well recorded its trophic state history (Chen et al., 2019; Chen et al., 2020). Accordingly, the Hongfeng Reservoir was selected as the study site. We aimed to investigate the content of P species and organic C as well as the C/P ratio in the sediments buried before and after eutrophication, and elucidate whether organic P regeneration was enhanced after eutrophication.

2. Material and methods

2.1. Study site

The Hongfeng Reservoir (N 26°26′–26°36′, E 106°19′–106°28′) is located about 32 km west of Guiyang, the capital of Guizhou Province, in Southwest China. Originally constructed in 1960, it has a catchment area of 1596 km², an average water area of 32.15 km², and an average water depth of 10.5 m (max. 45 m). The water volume is 2.81×10^8 m³, with a mean water residence time of 0.33 years. It is monomictic, and usually stratified in summer.

The Hongfeng Reservoir is the main source of drinking water for Guiyang City, and supplies water to over four million people. However, it has suffered water quality deterioration and eutrophication due to increasing external nutrient input since the 1990s (Chen et al., 2020). The Conservation Regulation of Water Quality for the Hongfeng Reservoir was promulgated by the government in 1996, and a variety of restoration strategies have since been implemented. Although external nutrient loading has been drastically reduced, algal blooms have still occurred frequently in recent years (Wang et al., 2016).

2.2. Sample and methods

One sediment core was retrieved from the northern central section of the Hongfeng Reservoir (Fig. 1) in July 2017 (Summer), October 2017 (Autumn), January 2018 (Winter), and April 2018 (Spring), respectively. In order to ensure that the positions of the four cores collected in different seasons are close to each other, GPS was used to fix the sample position. In the field, the sediment cores were sectioned at 1 cm intervals to a depth of 20 cm, and then sectioned at 2 cm intervals thereafter. The sectioned sediment samples were placed into polyethylene centrifuge tubes. Immediately before the sediment cores collection, temperature (T) and dissolved oxygen (DO) measurements were performed *in situ* from the surface water to bottom water at the intervals of approximately 1 m using a YSI 6600 V2 multi-parameter instrument (YSI 6600 V2).

Within 12 h after collection, samples from the upper 20 cm of sediment cores were centrifuged (3800 r, 15 min) to extract porewater. The supernatant was filtered using acetate-fibre membranes (0.45 μm) and stored in 15 mL polyethylene centrifuge tubes. Aliquots of 5 mL porewater were extracted to determine the total dissolved phosphorus (TDP) concentration. Briefly, samples were digested by potassium persulfate in an autoclave (120 °C, 30 min); thereafter, the colorimetric molybdenum blue method was used to measure TDP concentration. One group of parallel samples were measured out of every 5 samples for this, and the relative standard deviation (RSD) was less than 10%.

After porewater extraction, the sediment samples were instantly freeze-dried and homogenised. As the distribution of organic C and P varied in a narrow range in different seasons, the entire sediment core retrieved in July 2017 was solely selected for the analysis of TP, organic C (C_{org}), total nitrogen (TN), and P chemical speciation. The P chemical speciation in the sediments was determined using a five-step sequential extraction procedure. It separates sedimentary P into five components: loosely sorbed or exchangeable P (P_{ex}); ferric bound P (P_{Fe}); authigenic P (P_{authi}); detrital P ($P_{detrital}$); and organic P (P_{org}) (Ruttenberg, 1992).

A 200 mg sample of dried and ground sediment was washed with 20 mL of (1) 1 M MgCl₂ (pH 8, twice at 4 h) for P_{ex} ; (2) citrate dithionite bicarbonate buffer (pH 7.6, 8 h) for P_{Fe} ; (3) 1 M sodium acetate buffer (pH 4, 6 h) for P_{authi} ; (4) 1 M HCl (16 h) for $P_{detrital}$; and (5) 1 M HCl (16 h) after combustion at 550 °C (2 h) for P_{org} . After steps (1) to (4), the samples were rinsed repeatedly with 10 mL 1 M MgCl₂ solution and 10 mL deionised water to prevent the reabsorption of phosphate. TP in the samples was measured using the method validated in the frame of standards, measurements and testing programme (Ruban et al., 1999). P concentrations in known volumes of extractants were determined directly using the spectrometric phosphomolybdate blue method. All samples were analysed in triplicate and the data were expressed as

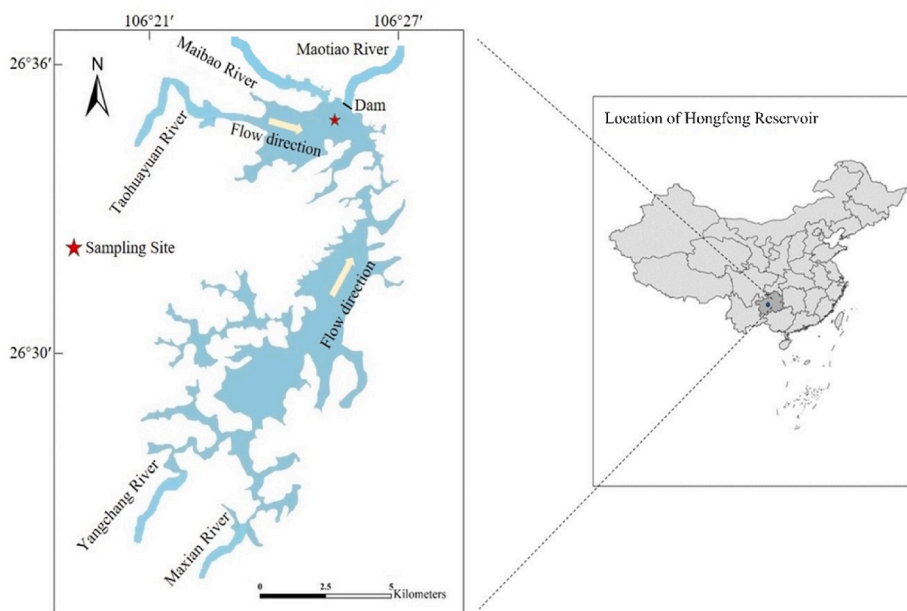


Fig. 1. Location of Hongfeng Reservoir and sampling site.

average values. The RSDs from replicate analyses of a given sample for all P components were less than 10%, and most of them were less than 5%. And the recovery of P sequential extraction ranged from 78% to 108%, most of them were between 80% and 90%.

For the analysis of C_{org} , the dried and ground sediment samples were soaked with 1 M HCl for 24 h to remove carbonates, and then washed with deionised water and dried again. These prepared samples were used to determine concentrations of C_{org} and TN using a PE2400-II elemental analyser (PE2400-II), and to measure stable isotope ratio of C_{org} ($^{13}C_{org}$) using the Mat 253 mass spectrometer.

3. Results

3.1. Temperature and DO in water column, and TDP concentration in porewater

Temperature and DO variations of the water profile in different seasons in Hongfeng Reservoir are shown in Fig. 2. The thermocline and oxycline form in spring and stabilize in summer (Fig. 2a and b). In summer, the DO concentration varied significantly from approximately 8.0 mg L^{-1} to less than 0.5 mg L^{-1} in the oxycline, which created hypoxic conditions ($<2 \text{ mg L}^{-1}$) in the hypolimnion (Diaz, 2001; Wang et al., 2016). In addition, the elevated concentration of DO in the hypolimnion might be attributed to the water discharge in the sampling period of summer. However, in autumn and winter, the DO mixed well in vertical profile of the lake.

The TDP concentration in porewater was very high in spring and

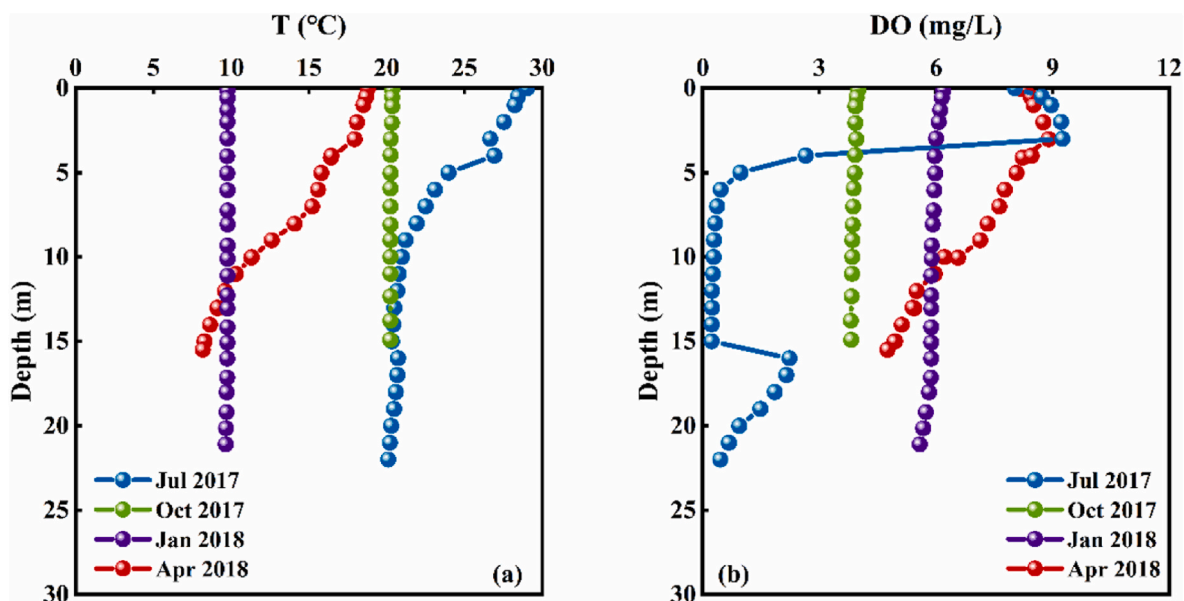


Fig. 2. Temperature (T) (a) and dissolved oxygen (DO) (b) variation of water column.

summer (Fig. 3), and reached a maximum of 1.4 mg L^{-1} in the upper profile in summer, indicating that the sediment had a high potential for P release. In contrast, the TDP concentration was much lower in autumn and winter.

3.2. Sediment P speciation

TP concentration varied from 873 to $1905 \text{ } \mu\text{g g}^{-1}$ throughout the entire sediment core from the Hongfeng Reservoir (Fig. 4a). It was low from the bottom up to the depth of 15 cm , rapidly increased to their peak value at the 10 cm depth, and then gradually declined up to the sediment-water interface. P_{org} was the major P component with concentrations ranging from 391 to $643 \text{ } \mu\text{g g}^{-1}$, accounting for 38.3 – 64.4% of TP in sediments (Fig. 5). The second largest P fraction was P_{authi} , with concentrations ranging from 131 to $502 \text{ } \mu\text{g g}^{-1}$ (13.4 – 33.5% of TP). Following P_{authi} , P_{Fe} and P_{detrital} had concentrations ranging from 51 to $274 \text{ } \mu\text{g g}^{-1}$ (5.3 – 15.9% of TP) and 86 – $202 \text{ } \mu\text{g g}^{-1}$ (6.6 – 16.0% of TP), respectively. The minimum fraction was P_{ex} , with concentrations ranging from 2 to $72 \text{ } \mu\text{g g}^{-1}$ (0.2 – 5.4% of TP).

3.3. Variations of C, N and P concentrations in sediments

As shown in Fig. 6, $\delta^{13}\text{C}_{\text{org}}$ values were relatively positive from the bottom of the sediment core to the depth of 26 cm , where the $\delta^{13}\text{C}_{\text{org}}$ values ranged from -23.65‰ to -22.57‰ . And then $\delta^{13}\text{C}_{\text{org}}$ became relatively depleted, ranging between -29.03‰ and -24.53‰ from the depth of 24 cm to the surface. The organic C/N ratios fluctuated between 13 and 17 from the bottom of the sediment core to the depth of 26 cm , and then sharply decreased to 11 at the depth of 24 cm . The organic C/N ratios of the upper 24 cm sediment are relatively stable and low. Based on the combined analysis of $\delta^{13}\text{C}_{\text{org}}$ and organic C/N ratio, the result indicated that the OM in the upper 24 cm sediments was mainly from autochthonous source and the other part was mainly from allochthonous source (Fig. 6).

C_{org} concentration varied between 2.97% and 7.45% throughout the

entire sediment core from the Hongfeng Reservoir (Fig. 4a). It was low from the bottom up to the depth of 15 cm , and rapidly increased to the peak value at the 8 cm depth, then followed by a slight decrease. The variation of C_{org} concentration corresponded well to the evolution of trophic state in the Hongfeng Reservoir (Chen et al., 2019).

The biogeochemical cycles of C, N and P are intimately coupled through lake primary production and OM degradation (Bratkic et al., 2012). A large proportion of C, N and P are delivered to the sediment-water interface as organic C, N and P in particulate OM. Patterns of C_{org} and TN were generally similar in the sediment core (Fig. 4a). TP showed synchronous fluctuations with C_{org} , except in the surface 0 – 10 cm sediment layer where C_{org} maintained high and constant values, and TP decreased gradually from $1905 \text{ } \mu\text{g g}^{-1}$ at a depth of 10 cm – $1250 \text{ } \mu\text{g g}^{-1}$ in the top sediment layer (Fig. 4a).

Fig. 4b shows the results of another sediment core that was collected in northern central region of the Hongfeng Reservoir in May 2017 and reported in Chen et al. (2019). Due to the complexity of lake topography in karst region, there were some differences in TP, C_{org} and TN contents; as well as the depth at which significant changes in their contents occurred. However, TP, C_{org} , TN, and organic C/N ratio of the two cores showed a similar variation trend (Fig. 4a and b).

4. Discussions

4.1. The variation of organic C/P ratios in the sediment core: evidence for enhanced organic P regeneration

Variation in organic C/P ratios in particulate matter provides important information for comparing the regeneration behaviours of C and P during OM decomposition (Boyd and Trull, 2007; Christian et al., 1997; Ingall et al., 1993; Krom and Berner, 1981). Generally, organic C/P ratio depends on the initial OM composition and differentiated transformation of C and P during early diagenesis in the sediment. As shown in Fig. 7a, organic C/P ratios were relatively constant below 15 cm in the sediment core, with an average value of 180 . They then sharply increased between 10 and 15 cm in the sediment profile, with high organic C/P ratios fluctuating around an average value of 300 in the 0 – 10 cm sediment layer. The higher organic C/P ratios in the surface sediment may be related to multiple factors such as the OM source (Ingall et al., 2005), as well as the difference in regeneration between C_{org} and P_{org} during OM degradation in early diagenesis (Emeis et al., 2000; Ingall et al., 1993; Slomp et al., 2004).

The mixing of OM with different C/P ratios determines the original organic C/P ratios of lake sediment. Generally, OM that originates from terrestrial plants has higher organic C/N and C/P ratios than planktonic OM (Ingall and Cappellen, 1990; Meyers, 1997; Meyers and Ishiwatari, 1993). Therefore, a higher proportion of OM sourced from terrestrial plants could lead to high organic C/P ratios; furthermore, we would expect high organic C/N ratios to occur synchronously in the surface sediment. However, Fig. 4a shows that organic C/N ratios decreased gradually from the depth of 15 cm to the top sediment, which is in contrast with the trend of organic C/P ratios (Fig. 7a). A significant negative correlation between organic C/N and C/P ratios ($r = -0.478$, $p < 0.05$, $n = 26$) in the sediment profile ruled out the above explanation. In general, the higher organic C/P ratios in the upper sediment is not dominated by the OM source.

In addition, dissolved nutrients stoichiometry can influence the C/P ratio of endogenous organics (Franz et al., 2012; Klausmeier et al., 2004; Persson et al., 2012). Under nutrients-depleted condition, phytoplankton would assimilate “what be supplied” and regulate their physiological functions for growth (Moreno and Martiny, 2018), leading to higher C/P ratio in phytoplankton (Hecky et al., 1993; Wang et al., 2018). While, when the nutrients are replete, phytoplankton would eat “what they need”, and the stoichiometry of nutrients might not significantly affect the C/P ratio in particulates. High input of agriculture pollutants and active cage culture resulted in rapid water eutrophication

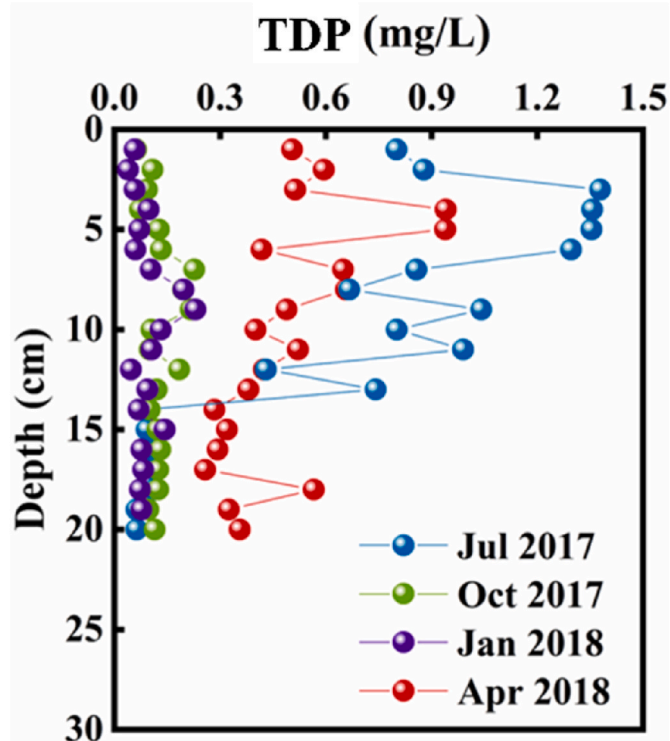


Fig. 3. Total dissolved phosphorus (TDP) variation of porewater in different seasons.

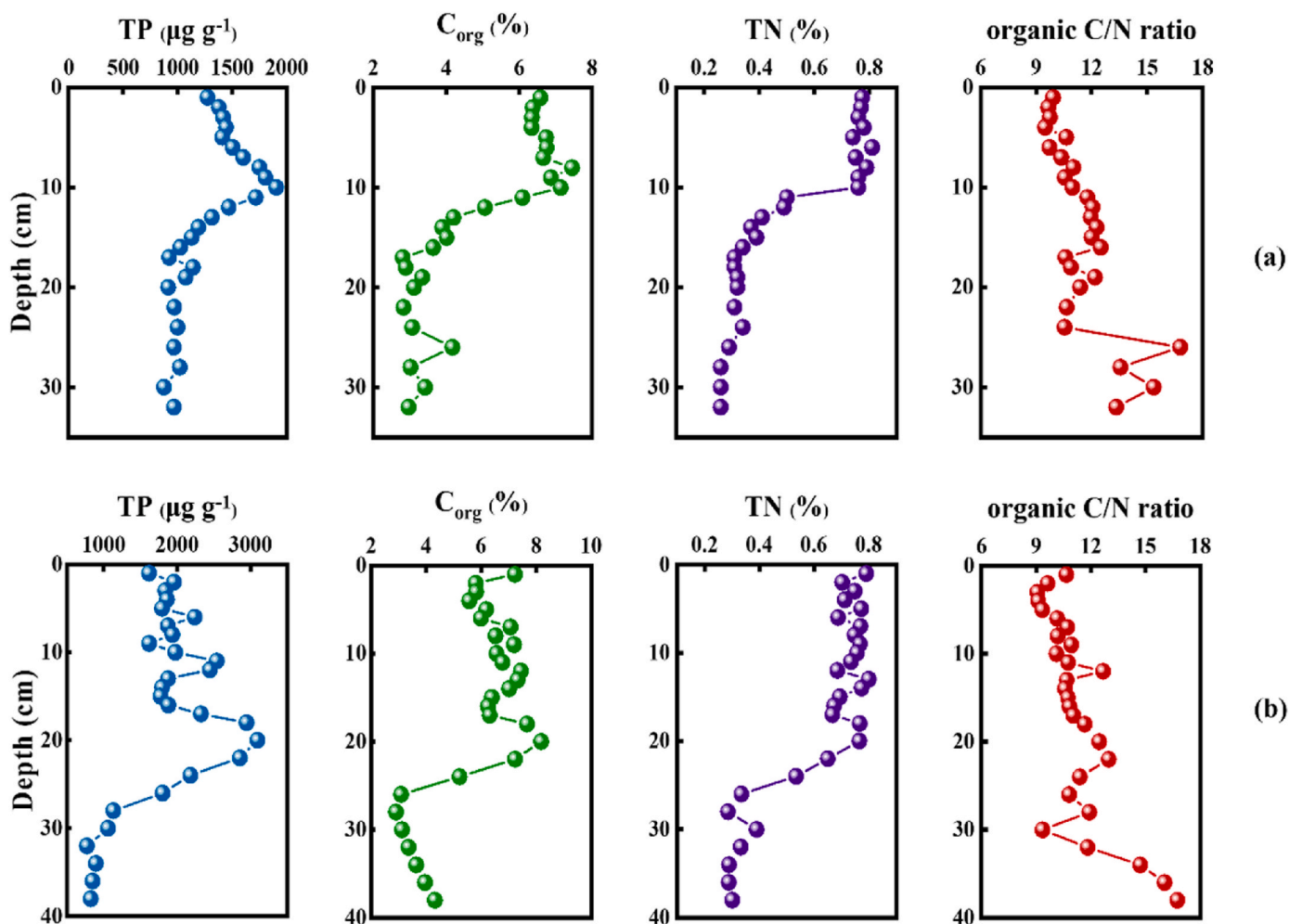


Fig. 4. Depth profiles of concentrations of total phosphorus (TP), organic carbon (C_{org}), total nitrogen (TN), and organic C/N molar ratios of the sediment core in this study (a) and another sediment core also collected in north central region of Hongfeng Reservoir in May 2017 (Chen et al., 2019) (b).

and algae blooms from the mid-1980 to mid-1990 in Hongfeng Reservoir (Yang et al., 2016). However, the organic C/P ratios in the sediments of Hongfeng Reservoir deposited in the corresponding period did not become lower than that in sediments before (Fig. 7a). On the contrary, the organic C/P ratios of the sediments increased since eutrophication occurred. This indicates that the stoichiometry of nutrients is not the dominant factor controlling the increase of organic C/P ratio in the sediments of Hongfeng Reservoir.

Besides original C/P ratios of OM, differentiated transformation rate of C and P during OM decomposition can also lead to higher organic C/P ratios in the surface sediment. Many studies have shown that P is preferentially regenerated relative to C during OM decomposition under anoxic conditions in marine environments (Ingall et al., 1993; Knauer et al., 1979; Krom and Berner, 1981; Slomp et al., 2002; Viktorsson et al., 2013a, 2013b). Thus, it is reasonable to expect that P can be preferentially regenerated relative to C during OM degradation in freshwater lakes. In our study, low organic C/N ratios (8–12) and relatively depleted $\delta^{13}\text{C}_{\text{org}}$ in the 0–24 cm sediment layer suggest that the sedimentary OM stems primarily from autochthonous phytoplankton. The steep increase of C_{org} from the sediment depth of 15 cm to the depth of 8 cm indicates that primary production in the lake increased rapidly. A previous study on the evolutionary history of the trophic state in the Hongfeng Reservoir also indicated that the rapid increase of C_{org} in the middle part of the sediment core resulted from increased primary productivity as a result of eutrophication (Chen et al., 2019). Increased primary production results in higher fluxes in OM deposition, further

accelerating hypoxia in the hypolimnion, as evidenced by the bottom water hypoxia in the Hongfeng Reservoir during summertime (Fig. 2b).

In contrast to C_{org} , the variation of P_{org} is relatively moderate and P_{org} concentration did not increase significantly as the sharp increase of C_{org} concentration. A previous study on the core sediments in the eastern Mediterranean Sea revealed that P regeneration had been enhanced relative to C during OM decomposition under anoxic conditions, leading to much higher organic C/P ratios (Slomp et al., 2002). Recently, a simulated study with lake water indicated that the transformation rate of dissolved organic P was significantly higher under anoxic conditions than that under oxic conditions (Bai et al., 2021). Accordingly, higher organic C/P ratios in the upper part of the sediment core in Hongfeng Reservoir support an enhanced P regeneration relative to C during OM decomposition under anoxic conditions which was induced by eutrophication.

As shown in Fig. 5, the content of organic P through the whole core is relatively stable, especially for the upper 20 cm sediment. When eutrophication occurred, the primary production significantly increased. However, organic P which is an important component of OM did not increase significantly as eutrophication occurred. The distribution of organic P in the sediment core of Hongfeng Reservoir further verified that organic P regeneration had been enhanced since eutrophication occurred. Microorganisms play a key role in the enhanced P regeneration relative to C from organic matter under anoxic conditions. Anaerobic microorganisms are inefficient to accumulate and store P compared to aerobic microorganisms and lose part of their intracellular

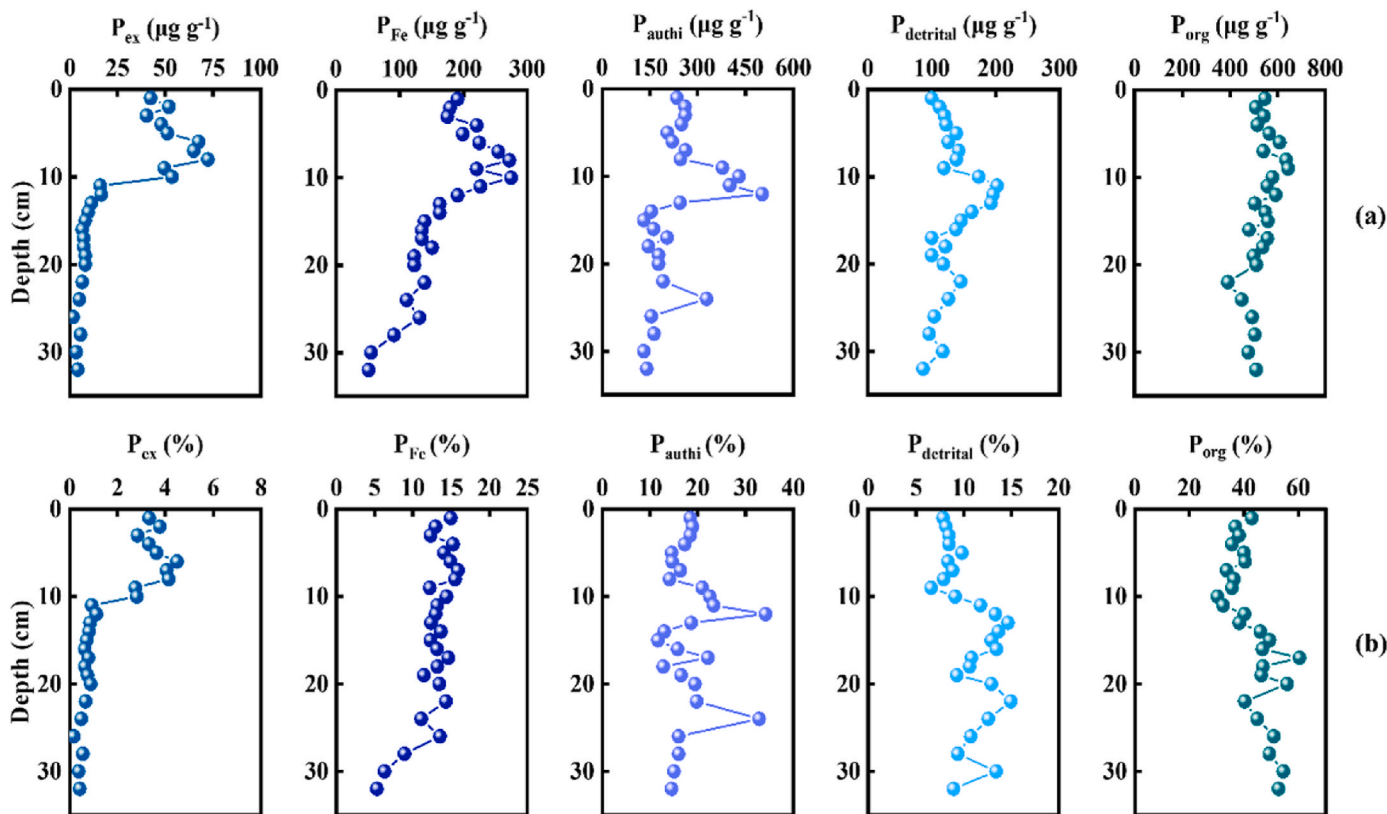


Fig. 5. Depth profiles of concentrations and percentages of P speciation including loosely sorbed or exchangeable P (P_{ex}), ferric bound P (P_{Fe}), authigenic P (P_{auth1}), detrital P ($P_{detrital}$), and organic P (P_{org}).

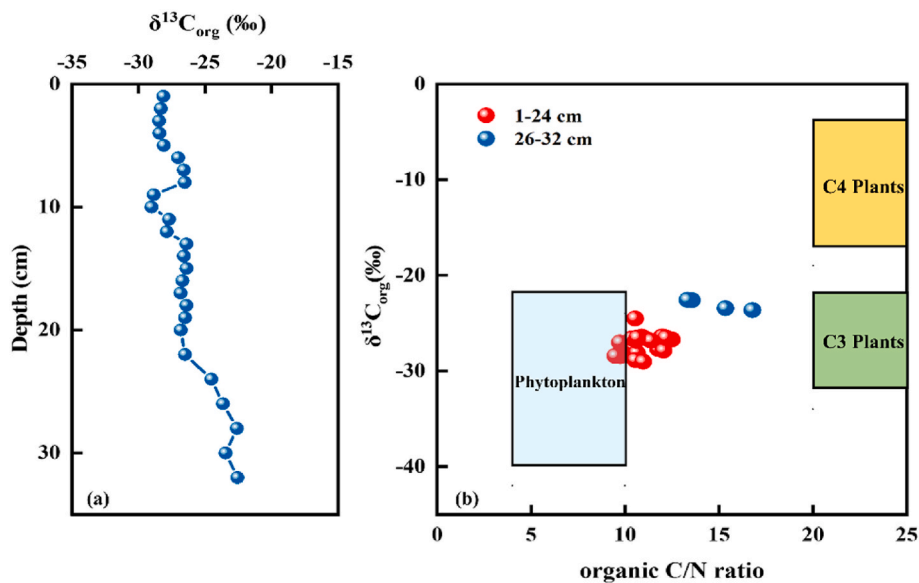


Fig. 6. Depth profile of $\delta^{13}C_{org}$ values (a) and organic C/N molar ratios vs $\delta^{13}C_{org}$ values (b) in the sediment core of Hongfeng Reservoir. Organic C/N molar ratio of phytoplankton ranges from 4 to 10, while organic C/N molar ratio of terrestrial organics exceeds 20 (Meyers, 1994; Yamaguchi et al., 2010). The $\delta^{13}C_{org}$ value of phytoplankton ranges from -42‰ to -24‰ , while the $\delta^{13}C_{org}$ value of C3 plants and C4 plants fluctuates from -34‰ to -24‰ and -19‰ to -6‰ , respectively (Smith and Epstein, 1971; Kendall et al., 2001).

P (Gachter et al., 1988; Slomp et al., 2002; Wentzel et al., 1986). In addition, Li et al. (2016) and Yuan et al. (2019) found that the elevated primary productivity increased alkaline phosphatase activity (APA). APA is a part of cell-membrane-associated enzymes, which can hydrolyze dissolved and particulate monoester P into orthophosphate (Trowsdale et al., 1990; Feng et al., 2018). Therefore, the increased APA under eutrophic condition can also promote P_{org} regeneration, further accelerating P recycling. Besides, the association of P_{org} with humic substance would affect enzyme hydrolysis of P_{org} (Zhao et al., 2020). It

has been established that the interaction of P_{org} with humic substance and/or metals can suppress the enzyme hydrolysis rate of P_{org} (Burdige, 2007; Zhu et al., 2015, 2018). The proportion of P_{org} associated with humic acid to TP becomes lower with increasing eutrophication (Wang et al., 2008). Huang et al. (2003) indicated that the proportion of P_{org} associated with humic acid to total phosphorus was only 1%–2% in the sediments of algae bloom zone, while most P_{org} associated with humic acid in the sediments of the lake where algae bloom never occurred. These studies collectively implied that P_{org} was more liable to be enzyme

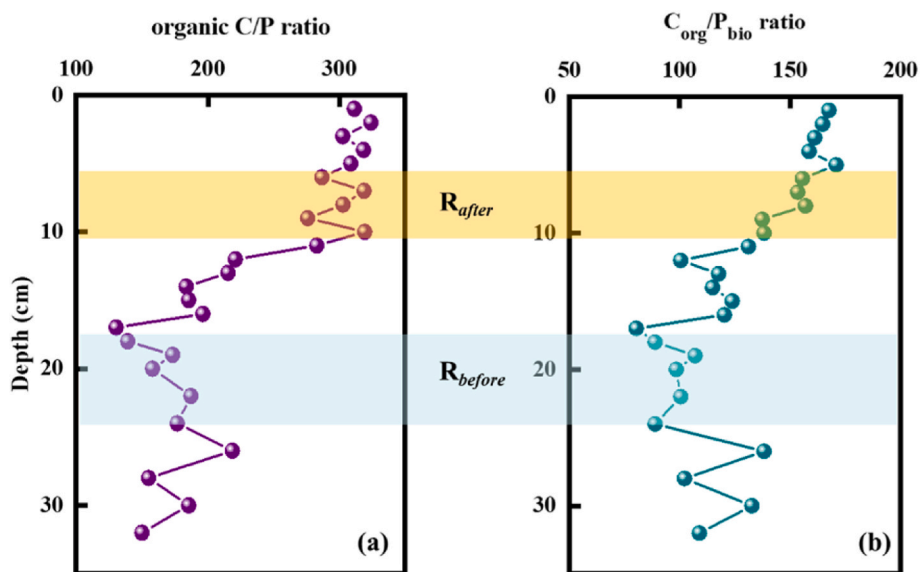


Fig. 7. Variations of organic C/P ratios and C_{org}/P_{bio} ratios. $P_{bio} = P_{ex} + P_{Fe} + P_{authi} + P_{org}$.

hydrolyzed under eutrophic state.

4.2. Estimation of organic P regeneration enhancement

Since eutrophication occurred, P_{org} regeneration has been enhanced in the Hongfeng Reservoir, which may provide important contributions

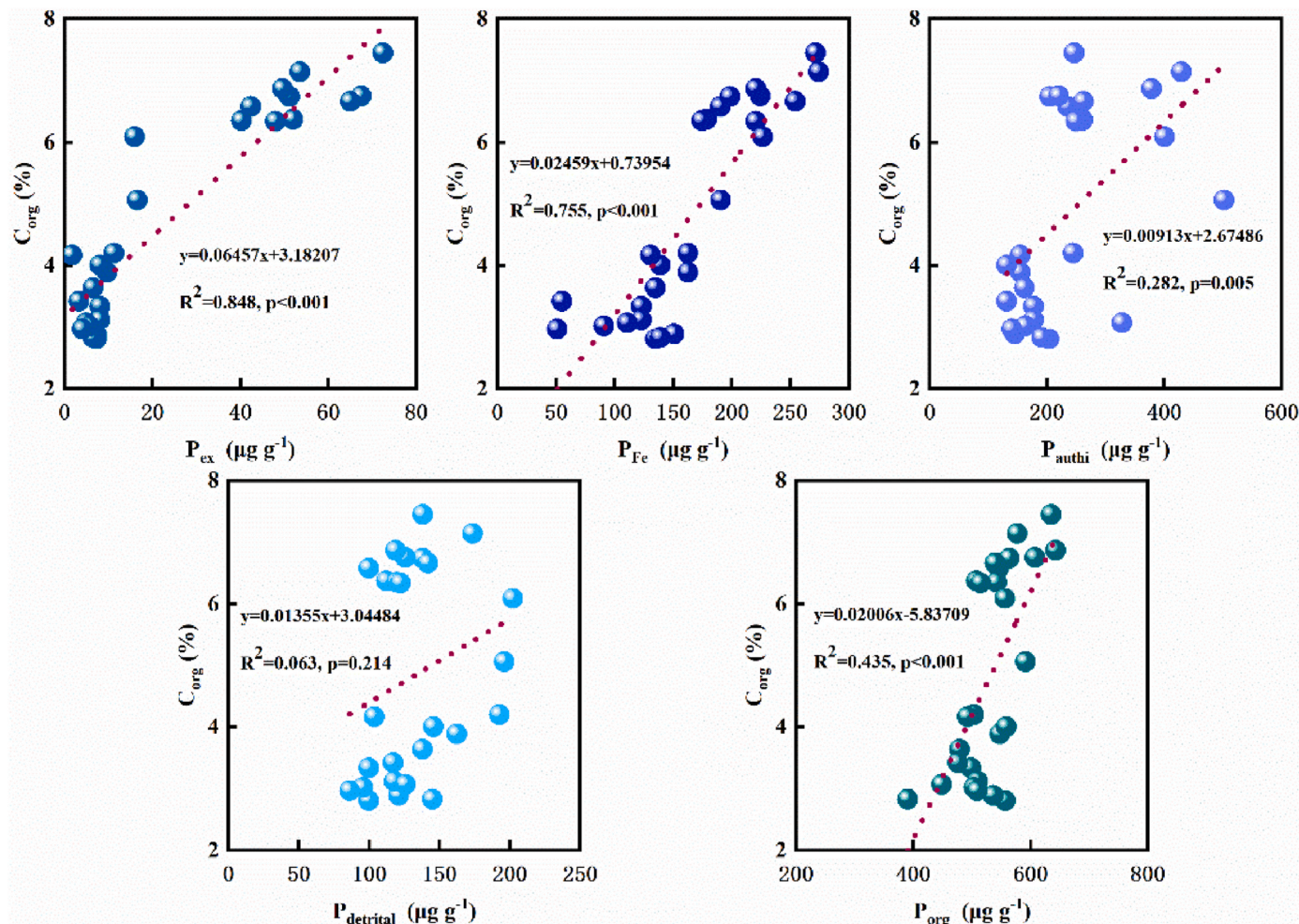


Fig. 8. Relationship between C_{org} and different P speciation. Except $P_{detrital}$, P_{ex} , P_{Fe} , P_{authi} and P_{org} all significantly correlated with C_{org} , respectively.

to the water column P inventory. In order to understand internal P cycling, it is important to quantitatively estimate the P released from P_{org} regeneration in sediments to the overlying water column.

Except P_{detrital} , P_{ex} , P_{Fe} , P_{authi} and P_{org} all respectively correlated with organic C significantly (Fig. 8), which implies that P_{ex} , P_{Fe} , P_{authi} and P_{org} all probably originates from OM. P released from OM decomposition, and thereafter, enters the sediment porewater, resulting in the supersaturation of dissolved inorganic P in sediment porewater. Dissolved inorganic P could precipitate as P_{authi} minerals such as carbonate fluorapatite, biogenic apatite and vivianite. Previous studies have demonstrated that P released during OM decomposition can be directly transformed to P_{authi} (Ruttenberg and Berner, 1993). Joshi et al. (2015) used authigenic phosphate isotope ($\delta^{18}\text{O}-\text{PO}_4^{3-}$) data to show that the regeneration of inorganic P from OM degradation was the predominant pathway for P_{authi} precipitation in the sediments of Chesapeake Bay. Besides, dissolved inorganic P in sediment porewater can also be trapped by Fe oxides. A recent study indicated that settled cyanobacteria OM could promote the formation of P_{Fe} in lake sediments (Wang et al., 2019), demonstrating that P_{Fe} in sediments is from OM decomposition.

P_{detrital} exists in inert minerals, and is biologically inaccessible. Biogenic P (P_{bio}) is used to characterize sediment P that mainly originates from OM. P_{bio} in the sediment contains three labile fractions (P_{ex} , P_{Fe} , and P_{org}), and one refractory fraction (P_{authi}). Regardless of whether P generated by OM degradation exists as P_{ex} , or is transformed to P_{Fe} and P_{authi} , all these fractions belong to P_{bio} . Thus, based on the comparison of $C_{\text{org}}/P_{\text{bio}}$ ratios in the sediments deposited before and after eutrophication, the enhanced proportion of P release can be estimated. In the same way, the enhanced proportion of P_{org} regeneration can also be estimated with organic C/P ratio.

As mentioned earlier, when the dominant source of C_{org} just transferred to autochthonous phytoplankton, the nutrition state of Hongfeng Reservoir was assumed to be oligotrophic, based on the low and constant C_{org} concentration and low organic C/N ratio between the depths of 16–24 cm (Fig. 4a). The average organic C/P ratio and $C_{\text{org}}/P_{\text{bio}}$ ratio of this part of sediment core are 165 ± 24 and 97 ± 13 , respectively (Fig. 7). The sediments between the depth of 6–10 cm, rather than the surficial sediments, were selected to represent the sediments deposited after eutrophication, as the OM in the surficial sediments would experience relative strong early diagenesis (Galman et al., 2008; Radbourne et al., 2017). The average organic C/P ratio and $C_{\text{org}}/P_{\text{bio}}$ ratio of the sediments between the depth of 6–10 cm are 301 ± 19 and 148 ± 10 , respectively. The enhanced proportion (w , %) can be estimated as follows (Chen et al., 2019; Emeis et al., 2000):

$$w = \frac{P_0 - P}{P_0} \times 100 = \frac{R_{\text{after}} - R_{\text{before}}}{R_{\text{after}}} \times 100,$$

where P_0 is the P_{org} or P_{bio} calculated by multiplying the accumulated C_{org} in the sediment deposited after eutrophication with $1/R_{\text{before}}$. P is the P_{org} or P_{bio} in the sediments deposited after eutrophication. R_{after} and R_{before} are the organic C/P ratios or $C_{\text{org}}/P_{\text{bio}}$ in the sediments deposited after eutrophication and before eutrophication, respectively. The enhanced proportion (w , %) of P_{org} regeneration was calculated with organic C/P ratios, while the enhanced proportion (w , %) of P release was calculated with $C_{\text{org}}/P_{\text{bio}}$ ratios.

The calculated results show that P_{org} regeneration enhanced approximately $45.2\% \pm 8.7\%$ and P release from sediment to water enhanced approximately $34.5\% \pm 9.8\%$ since eutrophication occurred in Hongfeng Reservoir, compared to before eutrophication. The enhanced regeneration of P during OM decomposition provides a considerable and sustainable nutrient resource for refuelling primary productivity, and has profound influences on the reservoir's trophic status. A positive feedback loop may develop among elevated primary productivity, higher flux in OM deposition, hypoxia in the hypolimnion, and enhanced P release from sediments. Similar relationships could be expected in other sub-deep reservoirs, such as Aha Reservoir, Baihua

Lake, and Lake Erie (Anderson et al., 2020; Lan et al., 2017; Wu et al., 2001). Furthermore, the internal P loading from OM degradation seems to be an important factor, which determines the sensitivity of water column P levels to external loading reduction, and further defines the ecological trajectories of reservoir ecosystems. Restoration of these sub-deep reservoirs becomes complicated by high internal P loading and the associated positive feedback loops. It is particularly critical to prevent the initial occurrence of eutrophication. Once eutrophication occurs, restoration of clear water ecosystems by reducing external loading is less achievable. Stronger controls on both external and internal P loading (such as applying passivant and bubble plumes) are required to limit eutrophication and maintain high water quality in sub-deep reservoir systems (Lan et al., 2017; Yang et al., 2020).

5. Conclusions

This is the first study delineating that P_{org} regeneration has increased significantly since eutrophication occurred in the reservoir. The P_{org} regeneration and P release from sediment to water approximately enhanced $45.2\% \pm 8.7\%$ and $34.5\% \pm 9.8\%$, respectively, in the Hongfeng Reservoir. Enhanced regeneration of P during OM degradation can provide a considerable and sustainable nutrient resource for refuelling primary productivity. We believe that a positive feedback loop may develop among elevated primary productivity, higher fluxes in OM deposition, hypoxia in the hypolimnion, and enhanced P release from sediments. Elevated primary productivity (algae) and the corresponding hypoxic/anoxic condition, both caused by eutrophication, promoted P biogeochemical cycle in the sub-deep reservoir. Restoration of the sub-deep reservoir becomes complicated by high internal loading and the associated positive feedback loop. It is particularly critical to prevent eutrophication. Once eutrophication happens, it would be unlikely to restore clear water ecosystem by separately reducing external loading. Our study highlights the importance of controlling P release from sediments in order to restore clear water ecosystems in the sub-deep reservoir.

CRedit authorship contribution statement

Jia Yu: Investigation, Data curation, Formal analysis, Writing – review & editing. **Yan Zeng:** Conceptualization, Methodology, Writing – original draft, Writing – review & editing. **Jingan Chen:** Conceptualization, Methodology, Writing – review & editing, Project administration. **Peng Liao:** Writing – review & editing. **Haiquan Yang:** Investigation. **Chao Yin:** Investigation, Formal analysis.

Declaration of competing interest

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

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