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Effects of aeration modes on transformation of phosphorus in surface sediment downstream of a municipal sewage treatment plant

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ABSTRACT

This work was devoted to investigating the long-term cumulative effect of municipal sewage treatment plant (STP) effluents on the sediments, near the plant outlet, in Yunliang River in Nanjing, China. Laboratory experiments were performed to assess the effect of different aeration modes on the migration and distribution of phosphorus (P) in the sediments. The field investigation showed that the concentrations of organic matter, different P fractions, dehydrogenase activity and alkaline phosphatase activity in sediments near the STP outlet were higher than those in the upstream site. The results of laboratory studies documented that the retention of P in sediments mainly depended on the characteristics of sediments. Collectively, long-term intermittent aeration accelerated the conversion of parts of organic phosphorus to occluded Fe–P due to that Fe^{2+} were oxidized to Fe^{3+} gradually, inactivating the sediments P, which preventing P release completely even in the case of anoxic conditions. Compared with aerating to water, aerating to sediments was more effective on transferring total phosphorus and dissolved inorganic phosphorus (DIP) in overlying water to sediments. Additionally, microbial enzymes activity was directly related to the concentrations of DIP, indicating that enzymes activity could serve as sensitive indicators of nutrient dynamics in water ecosystems.

Keywords: Sewage treatment plant; Aeration modes; Sediments; Potentially mobile phosphorus; Enzymatic activity

1. Introduction

Phosphorus (P) is the key limiting nutrient controlling eutrophication in many aquatic systems [1], which in the water was adsorbed to suspended particles in overlying water and integrated into sediments [2]. Sediments are considered as a sink as well as a source for nutrients, releasing dissolved phosphorus, which can play a vital role in controlling the eutrophication of surface water bodies [3]. Besides, the migration and transformation of P at the water/sediment interface have been conceptualized as a buffer mechanism involving a dynamic equilibrium between the dissolved and adsorbed or precipitated phosphorus [4]. In order to better understand P cycling in the aquatic ecosystem, the sediment P was separately quantified [5], among of which, NH_4Cl–P, BD–P and NaOH–nrP

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were more readily released from the sediments [6]. Potentially mobile phosphorus (PMP) was calculated as the sum of NH_4Cl–P, BD–P and NaOH–nrP [7], acting as an important parameter for predicting the future internal loading. Thus, reducing the content of PMP in the sediments, inactivating the sediment P has far-reaching effect on improving the water quality.

In order to reduce the release flux of P from the contaminated sediments, various remediation strategies have been developed, including capping with clean soils *in situ*, artificial destratification, precipitation of P with Fe salts and hypolimnetic aeration [3,8–10]. Aeration is one of the most important measures to restore the damaged river. And some studies have focused on the migration and transformation of P in the river sediments under the condition of aeration [11,12]. Aeration can strengthen mass transfer of oxygen from water column to the benthic microbial community in river systems, which could accelerate the oxidation and decomposition processes of organic phosphorus (OP) afterward [1,12]. Both aerating to water [1] and aerating to sediments [13] were recognized as effective ways to improve the water quality partly.

Sewage treatment plants (STPs) effluents constitute the prominent source of P to the receiving rivers, which were recognized as one major point source of anthropogenic contamination to river systems [14,15]. Past researches proved that the effluents from the STPs have deteriorated physical and biochemical parameters in the recipient river [16–18], which has an further negative influence on the biogeochemical processes of the superfluous nutrients [19]. However, little is known about the long-term cumulative effect of effluents to sediments near the outlet of the STP in the recipient river. And few studies analyzed the effect of different aeration modes (aerating to overlying water and aerating to sediments) on P migration and transformation in the recipient river system near the outlet of STP. In particular, the characterization of these processes and the underlying mechanisms are still not well understood.

The aims of this study were (1) to investigate the characteristics of sediment P fractions near the outlet of the STP in the Yunliang River and study the cumulative effect of the effluents on the sediments; (2) conduct the batch laboratory experiments to determine the effect of long-term different aeration modes on transformation of P fractions in sediments, to analyze the relationship between the P fractions and the capacity of P release.

2. Materials and methods

2.1. Study sites

The research focused on two sites in Yunliang River, site 1 (hereafter referred to as S1, 32˚00′25.59′′ N 118˚85′93.17′′ E) was 35 m downstream of the outlet of the STP (32˚00′26.07′′ N 118˚85′89.29′′ E). Site 2 (hereafter referred to as S2, 32˚00′69.49′′ N, 118˚84′ 03.59′′ E) was located in the Qiqiaoweng Wetland Park, about 1,850 m upstream of the STP. S2 has a good ecological environment, which was used as control to assess the impact of the STP input.

The Yunliang River, 2.9 km long, approximately 2 m deep, is a tributary of the external Qinhuai River, located in Nanjing, China. This STP implemented in 1998 handles a 500,000 population equivalents, which performs biological secondary treatment with activated sludge, but does not include tertiary treatment for N and P removal. The discharge of the STP effluent is relatively constant over the year (mean = 2×10^5 m^3 d^{-1}).

2.2. Sample collection

Surface sediments samples (0–5 cm) were collected using a UWITEC sediment corer, placed into Ziploc bags, and returned to the laboratory, where they were sieved through a 2-mm mesh, homogenized. A portion of sediments were used to analyze their enzyme activity and chemical properties. The remaining sediments samples were stored at 4˚C (2–4 d) for subsequent laboratory experiments.

Overlying water samples were collected from 0.5 m depth at each site, kept in sterilized borosilicate glass bottles, placed on ice, and returned to the laboratory for further processing.

2.3. Laboratory experimental apparatus and treatment

P release of laboratory experiments was performed in six Perspex containers (13 cm diameter × 40 cm height) labeled as S1-1, S1-2, S1-3, S2-1, S2-2, and S2-3, respectively. Homogeneous sediments samples from the two sampling sites (S1, S2) were injected into the six containers to a height of 5 cm from the bottom, then filtered river water was slowly added to a height of 25 cm. The sediments and water in S1-1, S1-2 and S1-3 were taken from S1, and the others were taken from S2. Six containers were all irradiated under fluorescent tubes, Dark:light period was12:12 h.

For S1-2 and S2-2, two air pumps were used for aerating to overlying water respectively, and the air

pumps in S1-3 and S2-3 were used for aerating to sediments. S1-1 and S2-1 served as the control containers, respectively, which meant that no aeration was done to the sediments or water. The four air pumps mentioned above connected to the corresponding containers were operated from 09:00 to 10:00 every day with the airflow of 100 L/h. The experiments lasted for 40-d period.

Overlying water for analysis was sampled 24 h after aeration stopped every day, and the same amount of river water were added immediately to the original level after sampling each time. Meanwhile, distilled water was added every 5 d to maintain the water column height of 25 mm due to the evaporation of water. After the experiments, sediment subsamples were taken from each container, dried, ground, homogenized, and sieved with a standard 100-mesh sieve for chemical analysis [20].

2.4. Analytical methods

2.4.1. Water chemistry

The pH and dissolved oxygen (DO) were measured directly *in situ* (portable electrochemical Meters, SX751, China). Total phosphorus (TP) and dissolved inorganic phosphorus (DIP) were determined by molybdate–ascorbic acid according to an established method [21,22]. Total nitrogen (TN) and chemical oxygen demand (COD) followed the standard methods [22].

2.4.2. Sediment chemistry

The fractions of P in sediments were determined on wet samples by extracting P according to the scheme [7]. P was separated into: (1) NH_4Cl–P (loosely sorbed P), (2) BD–P (iron bound P), (3) NaOH–nrP (organic P including bacteria-incorporated P), (4) NaOH–P (mainly aluminum bound P), (5) HCl–P (calcium bound P), and (6) Res-P (mainly refractory organic P and inert inorganic P). The TP in sediments was the sum of all P fractions. The PMP was the sum of NH_4Cl–P, BD–P, and NaOH–nrP. Organic matter (OM) was determined by digestion with potassium bichromate plus sulfuric acid [23].

2.4.3. Extracellular enzyme activity

Microbial activity was determined by measuring alkaline phosphatase activity (APA) and dehydrogenase activity (DHA). APA and DHA were assayed by p-nitrophenylphosphate (pNPP) [24] and triphenyltetrazolium chloride (TTC) [25] reduction, respectively.

3. Results and discussion

3.1. The effects of the STP effluents on the sediments in the receiving river near the plant outlet

The concentrations of nutrients (TN, TP, DIP, and COD) of the overlying water in S1 were higher. However, the DO was lower than that in S2 (Table 1). Table 2 shows the concentrations of TN, TP, OM, APA, and DHA of the sediments in S1 (2,550 mg/kg, 750 mg/kg, 3.32%, 1.28 mg g^{-1} h^{-1}, and 38.02 mg g^{-1} h^{-1}) were higher than those in S2 (780 mg/kg, 410 mg/kg, 1.36%, 0.31 mg g^{-1} h^{-1} and 8.23 mg g^{-1} h^{-1}). It should be noted that the values of DHA, APA in S1 were three times higher than those in S2.

The mean concentrations of the different P fractions and the relative contribution of each fraction to TP of the sediments in S1 and S2 were presented in Table 3. The BD–P and HCl–P both made the largest percentages to the sediment P in S1 and S2, while the concentrations of them both were higher in S1 than those in S2, what's more, NaOH–nrP followed by BD–P and HCl–P both in S1 and S2, which concentration was higher in the sediments of S1. The concentration of PMP in S1 was 397.5 mg/kg, higher than that in S2 (174.25 mg/kg) (Table 2), indicating that the risk of sediment P release was higher in sediments of S1. In particular, the BD-P was the dominant internal P source to water bodies, released from anaerobic sediments, which concentration was almost twice higher in sediments of S1 than that of S2, probably related to the nutrient states of the sediments and Fe concentration [3,26] of overlying water in the two sampling sites (Table 3). This should be ascribed to that the long-term effluent discharge caused elevated concentrations of stream water P and N in S1 compared to that in the upstream site (S2) [27].

Table 1
Main characteristics of overlying water at the two sampling sites in Yunliang River in October 2011

Site	DO (mg/L)	TP (mg/L)	DIP (mg/L)	pH	TN (mg/L)	COD (mg/L)
S1	3.50	0.79	0.32	7.52	14.84	55.86
S2	5.40	0.32	0.15	6.94	5.96	21.50

Table 2
Main characteristics of the two studied sediments

Site	TN (mg/kg)	TP (mg/kg)	PMP (mg/kg)	OM(%)	DHA (mg g^{-1} h^{-1})	APA (mg g^{-1} h^{-1})	C/N
S1	2,550	750	397.5	3.32	38.02	1.28	17
S2	780	410	174.25	1.36	8.23	0.31	30

Table 3
Phosphorus fractions concentration in the river sediments (mg P/g ± SEMs, n = 3)

	NH_4Cl–P		BD–P		Al–P		NaOH–nrP		HCl–P		Res-P	
Site	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%
S1	15.00	2	247.50	33.2	105	14	135	17.8	225	30	22.50	3
S2	3.69	0.9	120.95	29.5	70.52	17.2	49.61	12.1	141.86	34.6	23.37	5.7

The DO concentration in S1 was low (Table 1), resulting in low oxidation and decomposition rate of the OM, along with accumulated pollutants discharged from STP. This may directly affect the species composition, variation in biomass and succession of aquatic organisms [28]. Besides, it would further affect the enzymatic activity, especially alkaline phosphatase played an important role in the degradation of OP in sediments [29,30]. The values of DHA and APA in S1 were four times higher than those in S2, meanwhile the contents of OP and OM were also higher in the heavily polluted S1 (Table 2). The high concentrations of OM served as a carbon source for the microbes and caused high DHA [31], which may be explained by the mechanism that APA and DHA were induced by OP and OM [24,29]. The sediments in S1 have potential ability for biological self-purification, which was already inhibited by the relatively low DO saturation level, and further threatened by the wastewater [32].

The concentrations of TN and TP of sediments in S1 were almost four times higher than those in S2. The STP effluent was a kind of direct pollution input source for anthropogenic pollution, which had long-term cumulative effects on the outlet of STP, modified physical and biochemical parameters (the DO, nutrients, and enzyme activity) in the recipient river (Tables 1 and 2), mainly ascribed to the input of the STP effluents without tertiary treatment for N and P removal. The results also indicated that the high concentration of DIP of the overlying water in S1 was related to the nutrient states of the sediments [11], which was verified once more by the following laboratory experiments.

3.2. *The effects of aeration modes on the concentrations of P fractions, APA, and DHA in the sediments*

The concentrations of TP and different P fractions before and after the aeration process were shown in Fig. 1. The TP and BD–P concentrations of the sediments in S1-1 and S2-1 under the anaerobic condition (no aeration) were significantly lower than their initial values. In contrast, under the aeration conditions, the concentrations of BD–P, HCl–P, and TP all increased, compared with the initial values, the Al–P concentrations were relatively stable in all containers during the aeration process, the concentration of NH_4Cl–P in those sediments declined slightly, and NaOH–nrP declined significantly, compared with the initial values (S1, S2). It should be noted that the relative variation

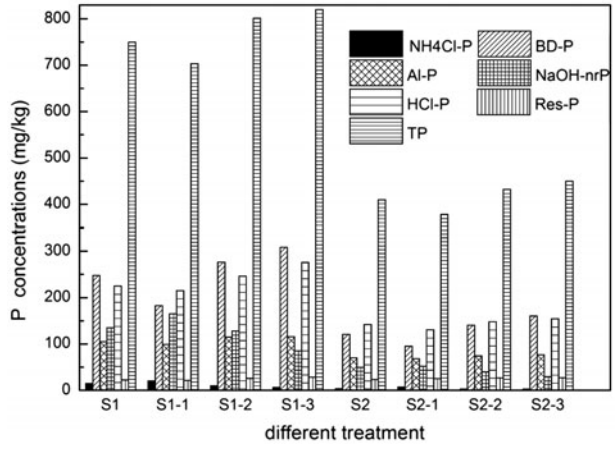

Fig. 1. Variations of P fractions in sediments.

of the P fractions in the sediments of S1-3 and S2-3 were larger than those of S1-2 and S2-2, respectively. Furthermore, biofilm was all observed on the surface of the sediments after the aeration experiments in S1-1, S2-1, S1-2, and S2-2.

The results above showed that P release from the sediments occurred in S1-1 and S2-1 under the anaerobic condition. In contrast, the long-term intermittent aeration facilitated the migration of P from the overlying water to the sediments eventually. This may be explained by the fact that (1) the BD–P made the largest percentage of the sediment P in S1 and S2, demonstrating that BD–P was the dominant compound of released P under the anaerobic condition due to the reduction process of occluded Fe^{3+} oxide to soluble Fe^{2+} oxide. This is also supported by the previous reports [20,33,34]. (2) The long intermittent aeration facilitated oxidation of soluble ferric (Fe^{2+}) compounds to insoluble ferric (Fe^{3+}) compounds [34]. Moreover, the P in the overlying water could also be greatly adsorbed by the $Fe(OH)_3$ when the ratio of Fe/P was high [29]. (3) The alternate operation of anaerobic–aerobic resulted from the intermittent aeration was advantageous for growth of the polyphosphate accumulating organisms (PAOs). It had been proved that the PAOs uptake P more than the released P in the sediments [35], since they were able to convert the dissolved phosphorus to insoluble phosphorus in the sediments [36].

After the aeration experiments, the TP concentrations of sediments in S1-3 and S2-3 were higher than those in S1-2 and S2-2, respectively (Fig. 1). This phenomenon may be explained that aerating to overlying water only accelerate aerobic metabolism through the active biofilm layer above the sediments [37]. But aerating to sediments can lead to higher penetration oxygen into the sediments, making the OM and OP become more degradable. There was a drop in the concentration of NaOH–nrP, meanwhile the BD–P of S1-3 and S2-3 increased significantly, so it can be inferred that a part of phosphate mineralized from OP was mainly converted into occluded Fe–P. Some study has indicated that the higher ratio of occluded Fe–P in the BD–P was the greater of P retention ability became [20]. The non-occluded Fe–P and occluded Fe–P in BD–P should be determined in the future, which is of more practical significance to analyze the migration of P in sediments.

The concentrations of NH_4Cl–P in the sediments of S1-1 and S2-1 increased significantly, which were probably due to the adsorption of extracellular polymeric substance (EPS) in the biofilm on the surface of sediments in the static water. Some studies have shown that EPS promoted the nutrient uptake from the overlaying water [38–40]. In contrast, under the condition of aeration, the concentrations of NH_4Cl–P in S1-2, S1-3, S2-2 and S2-3 decreased. The reason may be that shear force would interfere with the structural stability of the particles, which resulted in the desorption of P from particles in the surface sediments [41,42].

The concentrations of HCl–P increased slightly after the experiments, which may be induced by the slight alkaline pH (approximate 7.7). This fact was related to their own sediments which contained calcium phosphate minerals that favorable to HCl-P growth [43].

The appearance of the biofilms in S1-1, S1-2, and S2-1 was green, while the color was darker in S1-1. But the appearance of the biofilms in S2-2 was khaki. The color of the biofilms may be related to the community structure and diversity, which should be studied further.

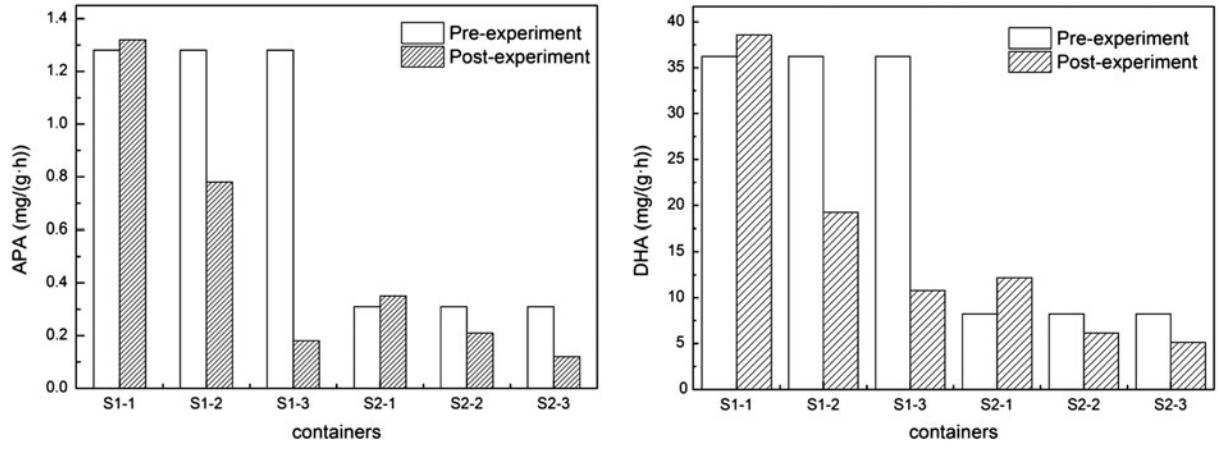

Fig. 2. Variations of APA and DHA in sediments.

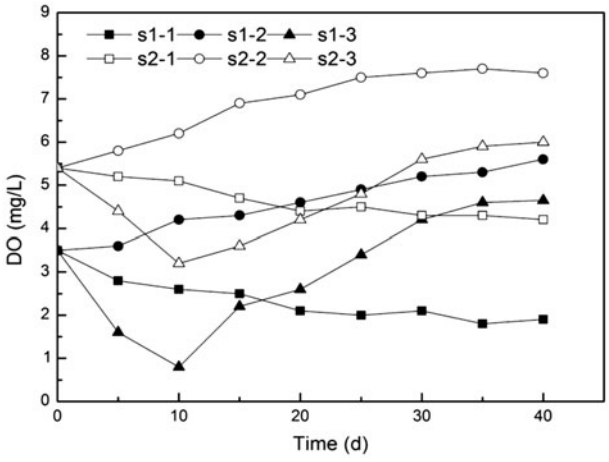

Fig. 3. Variations of DO in overlying water.

The initial values of DHA and APA of the sediments in S1 were four times higher than those in S2. After the laboratory experiments, the DHA and APA of sediments in S1-1 and S2-1 increased mildly, but their values in S1-2, S1-3, S2-2, and S2-3 dropped. The reduction rates of DHA and APA in S1-3 and S2-3 were higher than those in S1-2 and S2-2, respectively (Fig. 2), which may attributed to the sharp degradation of OP and OM under the influence of DO [44]. As the substrate of dehydrogenase and alkaline phosphatase, the OM and OP in sediments could reflect the trophic state, which were linked with the release of potential P of river sediments. The results further supported the idea that APA and DHA were induced by the high content of OP and OM [29].

3.3. The effects of aeration modes on DO, TP, and DIP in the overlying water

During the period of experiments, the mean concentrations of DO in S2 series were always higher than those in S1 series (Fig. 2). There were similar change tendency between two experimental groups (S1 and S2), the concentrations of DO in S1-1 and S2-1 declined gradually, the concentrations of DO in S1-2 and S2-2 slightly increased during the test period, reached up to 5.6 and 7.6 mg/L, respectively. However, the concentrations of DO in S1-3 and S2-3 declined rapidly during the first 10 d, particularly occurred in S1-3, the lowest value was 0.8 mg/L, and then, the DO increased to 4.65 mg/L eventually, higher than the initial value.

From Fig. 3, it is clear that there were mild variations of DO in S1-2 and S2-2 compare with S1-3 and S2-3, which may be explained by that aerating to water could result in lower penetration of oxygen into the sediments, boosting the aerobic metabolism just throughout the active biofilm layer [37]. By aerating to the sediments intermittently, the rate of hydrodynamic transport reached a higher level, resulting in the penetration of oxygen to the sediments became stronger, accelerating the mineralization of sediment OM and OP, which led to a large amount of DO consumed [45]. This was the reason that the concentrations of DO in S1-3 and S2-3 declined rapidly, accompanied with the concentrations of TP and DIP rapidly increased within first ten days of the experiments. Then, the concentrations of DO in S1-3 and S2-3 begun to rise after the fifteenth day, which may be due to large amounts of OM containing OP have been

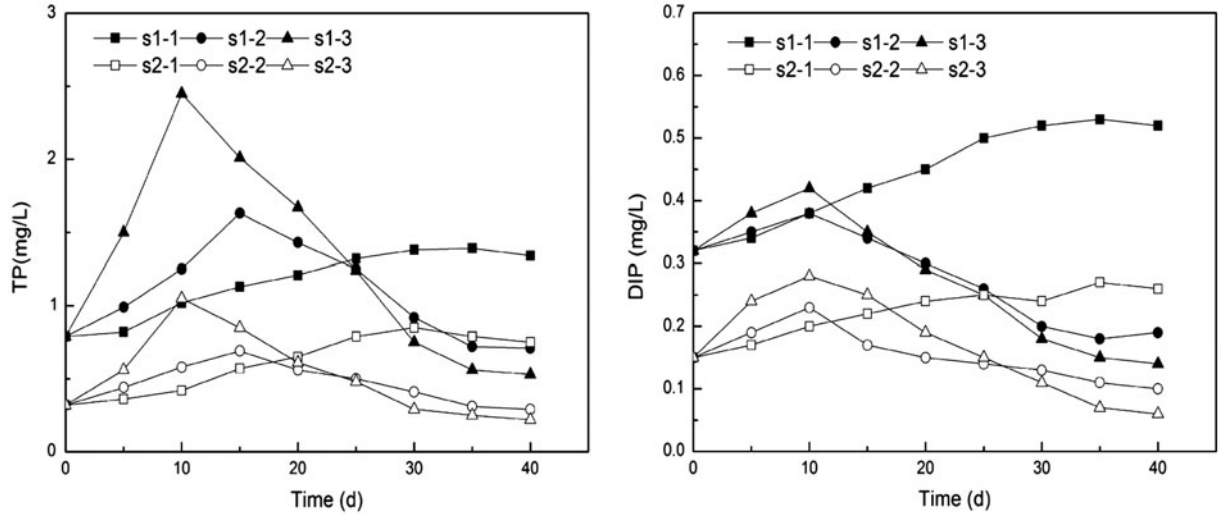

Fig. 4. Variations of TP and DIP in overlying water.

oxidized gradually, so the rate of oxygen filling by the aeration was greater than oxygen consumption.

There were the larger variation ranges of TP and DIP in S1-3 than those in S2-3 (Fig. 4), which attributed to the worse nutrient state and higher enzyme activity of the sediments in S1 [11,30]. In addition, the sediments with higher concentration of OM can offer a certain amount of carbon and nitrogen source for microbial reaction, producing more DIP [29], which were consistent with the decrease of TP and BD–P in the sediments after the aeration process (Fig. 1). From the above result, we can deduce a conclusion that the DIP release resulted, partially at least, from the desorption of the redox sensitive BD-P and the hydrolysis of some liable OP mediated by APA, which may lead to the release of bioavailable phosphate from the sediments as well [30]. Therefore, the transformation of P was a result of the combined action of biogeochemistry. The result supported the application of ecological stoichiometric theory in freshwater benthic ecosystems and emphasized the significance of the balance of biologically important elements for regulating organisms' response to their surrounding environment [46].

4. Conclusions

The STP effluent discharge caused elevated concentrations of P, N, DHA, and APA in stream water and sediment near the outlet site compared to the upstream site. The P retention of river sediments strongly depended on the characteristics of the sediments. The seriously polluted sediments could induce higher DHA and APA, resulting in the release of internal phosphorus.

Long-term intermittently aerating to sediments facilitated the migration of P from the overlying water to sediments. And the variations of the concentration values of the different target analytes were more evident than that by aerating to water intermittently, ascribing to that more oxygen penetrated into the sediments. Collectively, under the condition of long-term intermittent aeration, parts of phosphate mineralized from OP were probably converted into occluded Fe–P due to Fe^{2+} were oxidized to Fe^{3+} gradually; PAOs in sediments increased largely, up taking P from overlying water, which converted the dissolved phosphorus from water body into difficult soluble phosphorus to sediments. Then, the DIP in the overlying water decreased eventually, reducing the risk of water eutrophication.

It should be noted that the BD–P supplied the internal loading. However, not all BD–P can be released; the occluded Fe–P can enhance the P retention and even prevent P from being released completely in case of anoxic conditions. Thus, not all BD–P should be viewed as PMP under certain conditions. The non-occluded Fe–P and occluded Fe–P in BD–P should be further determined in future, which is of more practical significance to analyze the migration of P in sediments.

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