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Elimination of agricultural nonpoint source pollution using a pre-dam in the Taihu Lake basin: perspective from a laboratory study

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ABSTRACT

This study analyzes the influence of a pre-dam reservoir on the quality of agricultural nonpoint source pollution. The results clearly indicated that the pre-dam reservoir under study significantly improved the quality of the water flowing through the pre-dam. The greatest reduction in the pollutant concentration, particularly for suspended solids, NH_4-N, and chlorpyrifos was observed in the settling area of the pre-dam reservoir. The results of this study indicate that pre-dams may be successfully applied to reduce agricultural nonpoint source pollution in the Taihu Lake basin. However, the pre-dam reservoir was not able to completely eliminate all the contaminants flowing into Taihu Lake.

Keywords: Agricultural nonpoint source pollution; Functional areas; Pre-dam; Water retention

1. Introduction

Taihu Lake, located at the center of the Changjiang delta region, is one of the five largest freshwater lakes in China. The lake and its effluent rivers are important sources of water for the 40 million inhabitants in the area and the rapidly increasing industrial factories in Shanghai, Jiangsu, and Zhejiang. Additionally, the Taihu Lake basin is an agriculturally based catchment, and water quality degradation is a serious issue because of the use of fertilizers and pesticides in rural areas. Compared with point source pollution, agricultural nonpoint source pollution is more complicated to treat and more difficult to control. Agricultural nonpoint source pollution is a direct contributor to the deterioration of the water environment of the Taihu Lake watershed.

Suspended solids (SS) compose the primary pollutant of the nonpoint source pollution in the basin, as approximately 80% of SS in the area is from nonpoint source pollution [1]. Pollution from widespread use of

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chemical fertilizers has led to high concentrations of nitrogen and phosphorus in the basin. Furthermore, residue from pesticides such as chlorpyrifos is used in agriculture to prevent and control pests and parasites for cattle and a wide variety of crops [2], remains in farmlands and eventually flows into Taihu Lake through run-off. Thus, agricultural nonpoint pollution negatively influences ecological health [3] and can harm human health by entering the food chain and contaminating drinking water, especially during the irrigation season [4].

Pre-dams are generally small reservoirs that have an average theoretical water retention time of a few days. They serve to improve the quality of inflowing water. Pre-dams are situated immediately above the large main reservoir [5]. The pre-dam approach has been implemented for removing nonpoint source pollution from the Taihu Lake basin [6,7]. However, the pre-dams that have been used in the actual system did not focus on the removal paths and the contribution of SS, nitrogen, and phosphorus in the different functional areas of the entire pre-dam system, but rather focused on the overall removal of TN and TP by comparing the concentrations of the influent and effluent [6,7]. Furthermore, few studies have evaluated pesticide removal in pre-dams.

The difficulties encountered in adjusting the operational parameters in a large facility after implementations are well known. The system can be analyzed more completely in the controlled environment of a lab system. It is necessary to investigate removal in each of the lab-scale subunits of the pre-dam system to achieve the best overall removal of pollutants and to determine the optimal operational parameters such as the retention time and the optimal functional structure.

Thus, from a laboratory perspective, the objectives of this study are (1) to determine the optimal operational parameters in pre-dams and (2) to understand the removal paths and relative contributions of agricultural nonpoint source pollution, including the pesticide chlorpyrifos in the different functional areas of the pre-dam by simulating the Taihu Lake basin. The research findings of this paper will provide technical support for pre-dam engineering applications.

2. Materials and methods

2.1. Composition and structure of the pre-dam system

The laboratory scale pre-dam, made from organic glass, was divided into six isolated areas based on their different functions, i.e. the settling area, the shallow wetland area, profundal zone 1, profundal zone 2, the deep wetland area, and the biological membrane purification area, which are labeled A, B, C, D, E, and F, respectively.

Within A, the larger-sized particles and other pollutants attached to the SS should settle out. In B and E, all agricultural non-point source pollutants should be removed, and in C and D, the smaller-sized particles should be removed. In F, the TN and NH_4-N should be eliminated.

Reeds and *Acorus Calamus* were planted in B and E at a density of 50 plants/m^2 and 55 plants/m^2, respectively. *Hydrilla varticillata* was planted in C and D at a density of 25 clumps/m^2 and 36 clumps/m^2. Fig. 1(a) shows the composition structure of the pre-dam. The actual operational device is shown in Fig. 1(b).

To improve TN removal, Ackermann packing (Fig. 1(c)) was used as the biofilm carrier. It was added to cover the surface of the purification area; the thickness of the Ackermann layer was 5 cm. The biofilm was cultivated via continuously feeding with prepared wastewater of which the biochemical oxygen demand was 400 mg/L, also it was seeded with anaerobic digestion sludge and aerated for 2 h per day. Thus, a certain amount of biofilm is adhered to the surface of the packing to promote the removal of nitrogen.

2.2. Pre-dam system operation

Water flowed through the six zones with overflow successively from A to B, to C, to D, to E, and to F. The water depth in the shallow wetland zone was 0.2 m, and that in the other areas was 0.3 m. To easily control the different concentrations in the influents, the pre-dam was fed with 140 L of freshly prepared synthetic wastewater (the composition of the synthetic effluent is described in Table 1(a), once using a submersible pump. The flow rate was 0.42 m^3/d as compared to the average run-off in the Taihu Lake basin [8].

Sediment from the Taihu Lake was used as the source of SS for the pre-dam system influent after a necessary dilution. The characteristics of the particle size distribution of the sediment are shown in Table 1(b).

Nutrients were supplied by adding 2.18 g of $NaNO_3$, 0.93 g of NH_4Cl, and 0.7 g of KH_2PO_4 to 140 L of tap water. The carbon source expressed as COD_{Cr} was prepared by adding 8 g of glucose (Table 1(a)). Chlorpyrifos, which is a high-impact, medium-toxicity, broad-spectrum organophosphorus pesticide [9], was added to the diluted solution.

Chlorpyrifos is one of the most widely produced and sold insecticides in the world and is the only organic phosphorus pesticide that is certified by the World Health Organization.

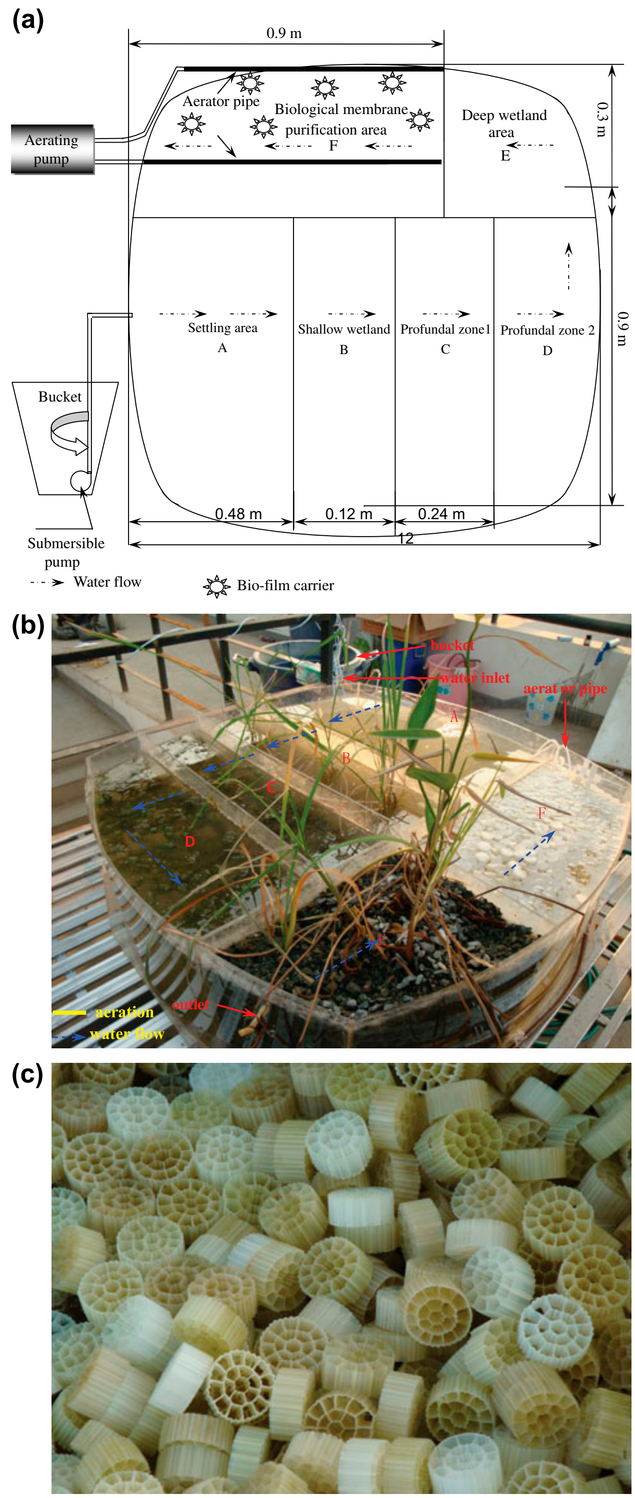

Fig. 1. (a) Schematic diagram and actual photograph of the pre-dam, (b) Actual photograph of the pre-dam, (c) Ackermann packing.

In China, starting from 1 January 2007, because of a total ban on the sale and application of highly toxic pesticides, such as parathion, phosphamidon, methamidophos, monocrotophos, and methyl parathion, the production of moderately toxic pesticides, such as chlorpyrifos, increased sharply. Chlorpyrifos then became the most widely used pesticide in China [10]. The concentration of chlorpyrifos was kept between 1.5 and 1.8 mg/L, which is similar to that of natural sources contaminated with chlorpyrifos in the Taihu Lake basin [10], though these concentrations are high in a global context. To distribute the synthetic wastewater more evenly in the system, an electric stirrer was used in the feeding tank, using a rotation of 45 rpm.

2.3. Measurements

Samples were taken every day until the effluent water quality became stable. Stability was achieved when the removal rates no longer varied. The water samples were analyzed for SS, TN, NH_4-N, NO_3-N, TP, TDP, and chlorpyrifos according to the water and wastewater monitoring method [11]. The water samples were collected in 100 mL polyethylene bottles, sent immediately to the laboratory, and transferred into brown glass bottles to wait testing. The samples were frozen until analysis.

Specifically, chlorpyrifos was quantified using an Agilent Technologies 6840 plus gas chromatographer, an microcell electron capture detector, a split/nonsplit automatic injector, an HP 5 column (5% phenyl-methylpolysiloxane), and helium as the carrier gas. The furnace temperatures were as follows: initial 60℃ for 0 min; ramp 1: 40℃ min^{-1} from 60℃ to 200℃ for 1 min; and ramp 2: 10℃ min^{-1} to 240℃ for 2 min. The injector and detector temperatures were 290℃ and 300℃, respectively [2]. The recovery and detection limit of chlorpyrifos were 81.2–96.5% and 0.01–0.02 μg/L, respectively.

3. Results and discussion

3.1. Removal efficiency and fate of SS in the pre-dam system

After only one day of retention time (the retention time refers to the amount of time that the polluted water was kept in a functional area), the SS concentration was difficult to measure in the effluent, indicating that the pre-dam system had considerable efficiency in SS removal. Fig. 2 shows the removal efficiency and the concentration of SS in the six functional areas of the pre-dam system. The average SS level of the input flow to the settling area was 242.3 mg/L. After passing through the basin, SS in the water flow

Table 1a
The qualities of pollutants and analysis

Concentrations (mg/L)	Pollutants	Sources (AR)	Analysis methods
4.30	TN	$NaNO_3$	Alkaline potassium persulfate digestion–UV Spectrophotometric method
1.74	NH_4–N	NH_4Cl	Nessler's colorimetric method
1.14	TP	KH_2PO_4	Ammonium molybdate spectrophotometric method
60.95	COD_{cr}	Glucose	Potassium dichromate method
200–250	SS	Sediment	Combustion method
1.5–1.8	Chorpyriphos	Chorpyriphos	Gas phase chromatography

Table 1b
Properties of the sediment to simulate SS in synthetic wastewater

Water content (%)	39.28
Organic matter[a] (%)	2.72
Particle size (μm)	
d(10)	2.236
d(50)	15.042
d(90)	53.091

[a]Ignition method [11].

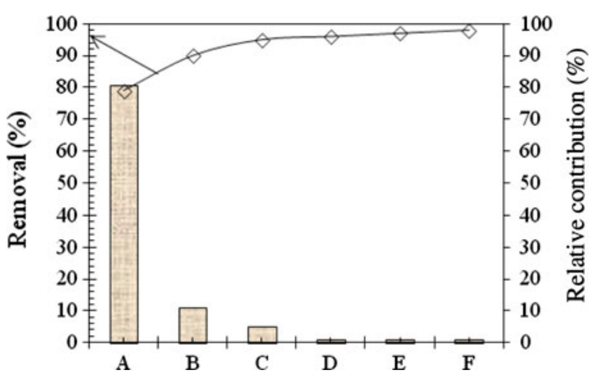

Fig. 2. Removal and relative contribution of SS in the different functional areas.
Note: A: Settling area; B: Shallow wetland; C: Profundal zone 1; D: Profundal zone 2; E: Deep wetland area; F: Biological membrane purification area.

entering the shallow wetland decreased to 28.5 mg/L. In the settling area, approximately 80% of SS were removed. The settling area eliminated SS primarily through settling. Furthermore, on a mass basis, the majority (approximately 95% of the total mass) of the trapped sediments in the settling area were larger particles. Therefore, because of the lack of vegetation, the settling area was not effective in retaining either light density plant organic matter aggregates or very fine clay particles, although it was highly effective in settling heavy particles. This result is similar to that obtained by Lothar et al. [12].

When SS in the water flow entered into the wetland area, emergent aquatic plants slowed down the water flow in the shallow wetland area, causing more SS to settle [13].

In the profundal areas, submerged plants were set to slow the water flow and increase the rate of interception of SS. SS were observed in the plant residues in the profundal areas. In the shallow and deep wetland areas, finer SS were removed from the water flow through the combined actions of substrates and plants. Thus, different mechanisms in the different functional areas contributed to the removal of various sizes of SS. However, the removal path of SS in the pre-dam system is still not completely clear. Further study, such as developing a model or a quantitative relation based on the flow rate and system configuration, which determines the flow pattern, would be useful to more completely understand the mechanisms.

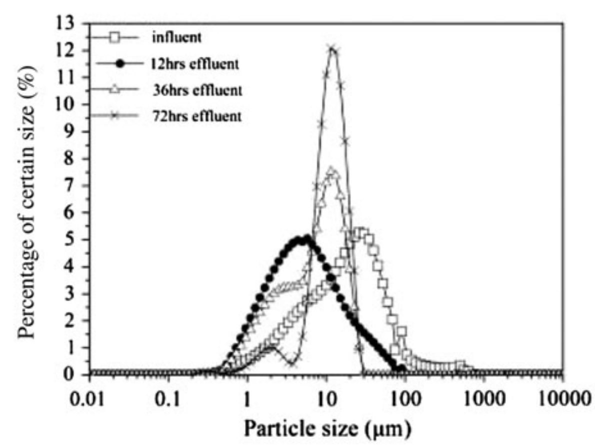

Fig. 3. Particle size distribution of inflow and outflow in the settling area.

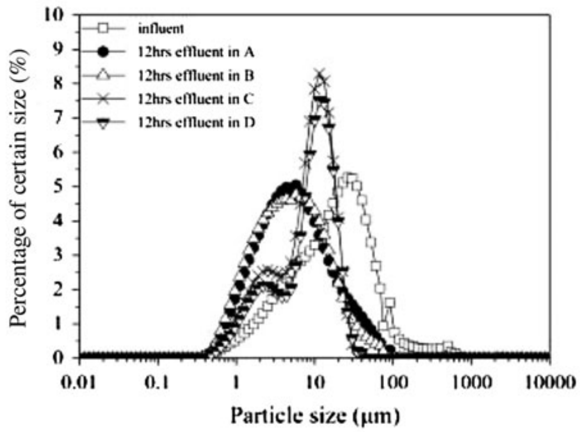

Fig. 4. Particle size distribution of the effluent in the subunit of the pre-reservoirs.
Note: A: Settling area; B: Shallow wetland; C: Profundal zone 1; D: Profundal zone 2.

As shown in Fig. 3, the particle size decreased as the retention time increased from 12 to 72 h. Therefore, optimizing the retention time for increased deposition and physical trapping by emergent vegetation may be the key to the removal of the particles and consequently to improve the overall performance of a pre-dam system in a wetland area.

The data in Fig. 4 shows that the effluent did not contain SS with particle sizes larger than 100 μm, with a continuous 12 h of influent, indicating that SS with particle sizes larger than 100 μm completely settled in the settling area. In addition, most SS with particle sizes larger than 20 μm in the influent settled within the whole system, as measured by comparing the influent with the effluent after 12 h (Fig. 4). Upon increasing the retention time, the settled of SS with particle sizes larger than 30 μm precipitated within 36 h. The content of small particles (2–7 μm) in the effluent after 36 h decreased from 8 to 5% compared with that in the effluent after 12 h. The particles that occupied the highest content became larger, as shown in Fig. 4. Thus, SS (10–30 μm in size) settled primarily in the settling area and the remainder was carried with the water flow to other functional areas.

As shown in Fig. 4, the particle size in the effluent decreased and then slightly increased from the settling area to the profundal zones, i.e. in the direction of the water flow. As shown in Fig. 5, approximately 3 cm of thick sediment, measured with a needle, was deposited in the settling area. The submerged plants and gravel were also covered with SS. First, the influent passed through the settling area where most of the larger particles (more than 30 μm) were deposited. After exiting the settling area, the water flow became slower, allowing slightly smaller particles to settle. In the profundal areas, the presence of particles with

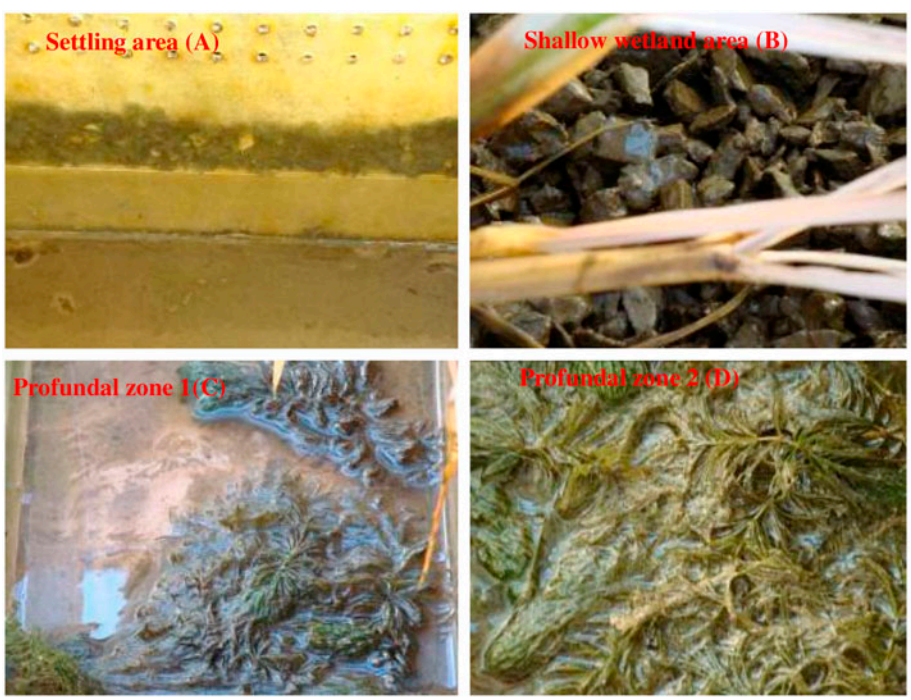

Fig. 5. Remaining SS in the subunit of the pre-reservoirs.

sizes of 5–8 μm was sharply decreased, whereas the presence of those with a size of approximately 10 μm increased from 4 to 10%, indicating that the SS (5–8 μm) were trapped by the plants. This result may have been observed because the plants were not able trap SS with particle size of 10 μm.

The data in Table 2 shows that SS larger than 30 μm were completely removed and that 95.24% of SS smaller than 5 μm were removed in the settling area. The data in Table 2 show that the lowest removal rate was for SS with particle sizes of 10–15 μm, at 74.77%. Thus, efforts aimed at complete SS removal should focus on enhancing the absorption, settling, or interception of SS in this range (10–15 μm).

3.2. Removal efficiency and fate of nutrients in the pre-dam system

The data in Fig. 6 shows the removal of nutrients with retention time. The pre-dam system showed increased removal efficiency for NH_4-N, TP, and TDP as the retention time was increased. Overall, the removal rate of NH_4-N was 60 to 80%, although it fluctuated slightly. The removal rates of TP and TDP were approximately 55 and 70%, respectively. These rates achieved stability within the first three days. The NO_3-N removal rate increased from 10 to 30% over the tested period, although there was a peak fluctuation at a removal rate of 50% for NO_3-N. The two main reasons why there was a fluctuation for NH_4-N and NO_3-N at day 3 were—first, it was affected by temperature fluctuations and second, random error existed during the experiments.

The high removal efficiency for phosphorus resulted from the numerous mechanisms for phosphorus removal throughout the pre-dam system, i.e. chemical adsorption, precipitation, and plant uptake [14]. For NH_4-N, both absorption and nitrification contributed to NH_4-N removal [15].

Table 2. Particle size distribution of the removed SS

Particle size (μm)	Removal (%)
>40	100
30–40	99.05
5–30	84.70
<5	95.24
20–30	94.64
15–20	85.38
10–15	74.77
5–10	84.19

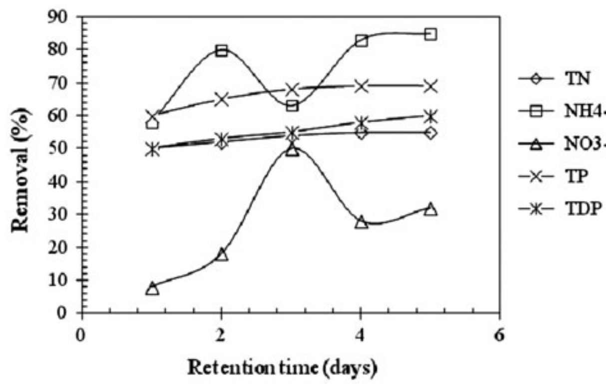

Fig. 6. Removal of nutrients of the whole the pre-dam with different retention times.

Although the ammonium concentrations were high, dissolved oxygen (DO) was very low at 0.1 mg/L in the shallow wetland. These two conditions, a high ammonium concentration and low DO, inhibited the denitrification activity in the wetland area. Thus, poor denitrification resulted in low removal rates for TN and NO_3-N [16].

The removal of nutrients in the different functional areas is shown in Fig. 7. The results are similar to those for retention time (Fig. 6). The removal rates of the whole pre-dam system improved gradually as the water flowed through the six functional areas. From Fig. 7, the pre-dam showed high removal efficiency for NH_4-N, achieving a removal rate of approximately 85%. In addition, removal rates for TP, TDP, TN, and NO_3-N were 68, 55, 52, and 46%, respectively.

To understand the quantitative contribution of the different functional areas to nutrient removal, the contributions of the different functional areas to TN, NH_4-N, NO_3-N, TP, and TDP removal are described

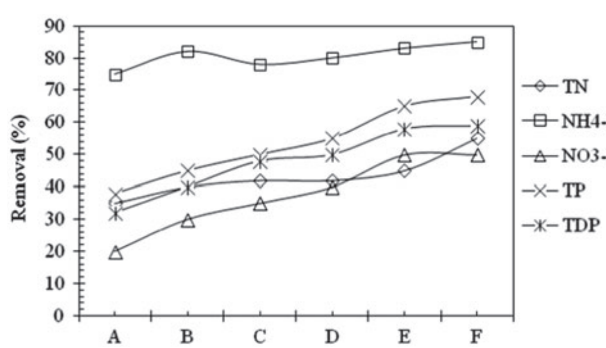

Fig. 7. Removal of nutrients in the different functional areas.
A: Settling area; B: Shallow wetland; C: Profundal zone 1; D: Profundal zone 2; E: Deep wetland area; F: Biological membrane purification area.

in Fig. 8. For both TN and TP, the settling area removed over half of the nutrients with a removal efficiency of 35% for TN and 38% for TP. The contributions of the shallow wetland area, profundal area 1, profundal area 2, the deep wetland area, and the biological membrane purification area to TN removal were 9, 4, 0, 5, and 18%, respectively, and those to TP removal were 10, 7, 7, 15, and 4%, respectively.

The removal rate of TN in the biological membrane purification area was higher than that in the other four functional areas. For NH_4-N, 75% was removed in the settling area and 8% was removed in the shallow wetland. This result is not surprising, as the primary mechanisms of NH_4-N removal are absorption and nitrification [17]. In this study, the influent flowed into the settling area, where NH_4-N was absorbed by the settled SS and nitrification from high DO (5.21–5.68 mg/L) occurs. For NO_3-N, the highest nitrate elimination rate of 40% was found in a very shallow pre-dam and was primarily the result of high denitrification in the sediment [18]. As the low DO (0.2–0.3 mg/L) in the biological membrane purification area inhibited nitrification, aerator pipes (used by an aeration pump with an aeration intensity of

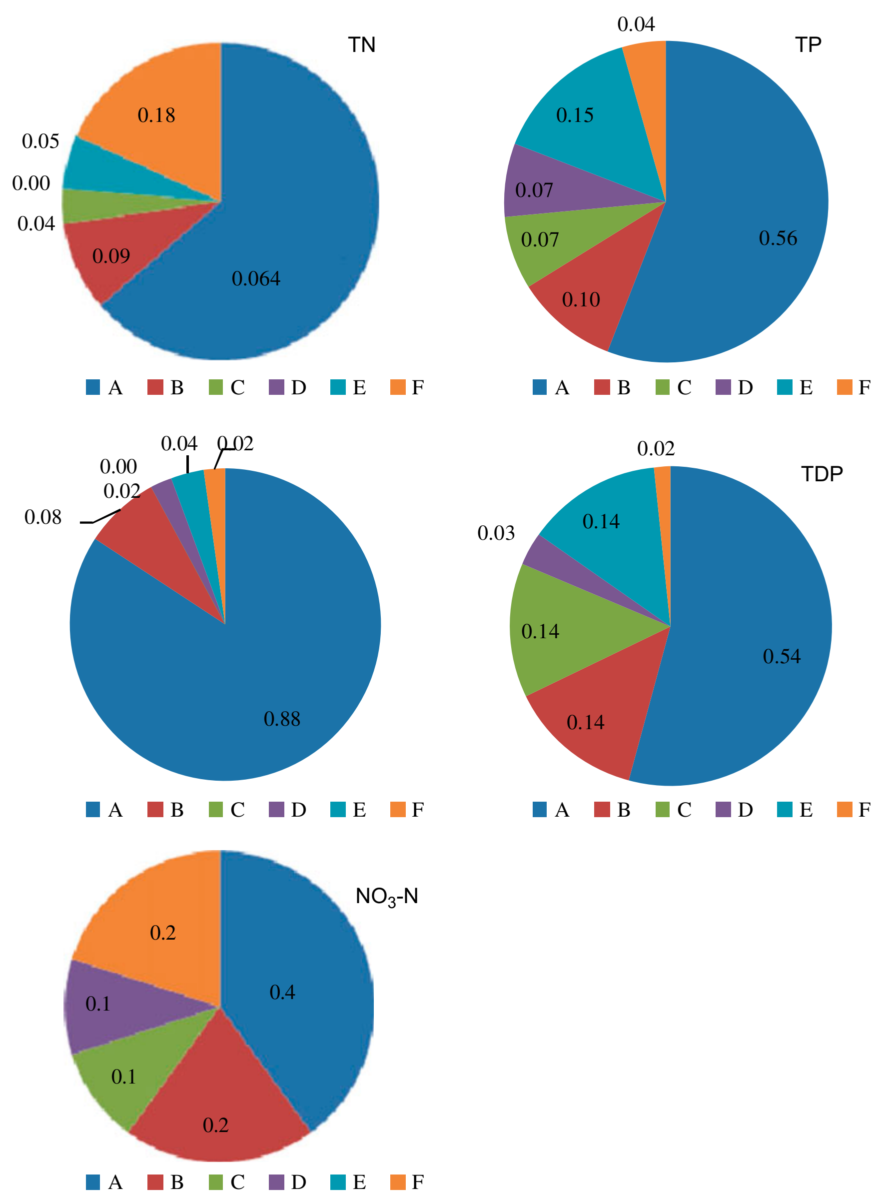

Fig. 8. Relative contributions to the removal of nutrients in the different functional areas.
Note: A: Settling area; B: Shallow wetland; C: Profundal zone 1; D: Profundal zone 2; E: Deep wetland area; F: Biological membrane purification area.

approximately 1.8 L/min) were used to increase the DO to approximately 4.0 mg/L. Increasing the DO improved the NO_3-N removal by 20%, accounting for the second largest contribution to the removal of NO_3-N. After 30 days of operation, NO_3-N removal was sharply increased to 90–95%, when abundant phytoplankton was observed. Surprisingly, the relative contribution of TN in the settling area was 64%. Two reasons may account for this result. First, the influent was diluted by the original water in the pre-dam when the influent flowed through the settling area. Thus, using the removal rate parameter to express the TN removal could make the relative contribution slightly higher than the actual removal quantity. This explanation can also apply to the other nutrients. Second, ammonia nitrogen is easily absorbed by and settles with SS. In the biological membrane purification area, the relative contribution of TN was 18%, which was also unexpected. Nutrient elimination in pre-dams is largely controlled by biological processes, primarily by phytoplankton growth, and it strongly depends on the retention time [19]. Therefore, a biological membrane system is required to improve TN removal, particularly for water with high NO_3-N content.

However, the situation is not the same for TP in the different functional areas. As shown in Fig. 8, the relative contribution by TP in the effluent was 25% in the shallow wetland area and in the deep wetland area. For TDP, the contributions of the shallow wetland area, profundal area 1, profundal area 2, the deep wetland area, and the biological membrane purification area to TN removal were 54, 14, 14, 3, 14, and 2%, respectively. The overall removal of TDP in the pre-dam system is consistent with that reported by Salvia-Castellvi et al. [20] and was probably caused by phytoplankton growth in the sediment. However, the mechanism of TDP removal must be studied further, as it currently is not well understood.

3.3. The removal efficiency and the fate of chlorpyrifos in the pre-dam system

In this study, when the influent was 1.8 mg/L, the chlorpyrifos concentration decreased sharply from 1.8 to 0.03 mg/L after 3 days of retention time. The removal efficiency of chlorpyrifos was studied in the pre-dam system; the results are presented in Fig. 9. As shown in Fig. 9, the removal rate of chlorpyrifos of the whole pre-dam system could be up to more than 95%. And the biggest relative contribution is about 56% in the settling area, followed by around 20% in shallow wetland area. The half-life of chlorpyrifos is reported to be approximately

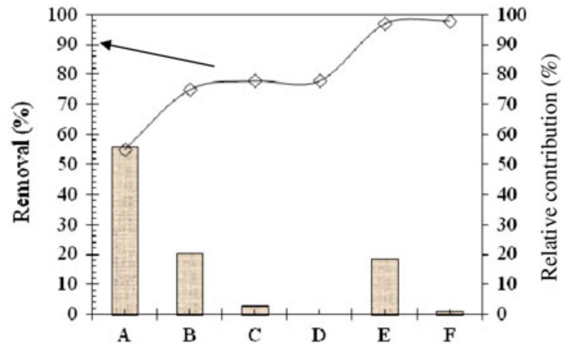

Fig. 9. Removal and relative contribution of chlorpyrifos in the different functional areas.

11–19 days in soil [21,22]. Compared to the published data, the half-life of chlorpyrifos in this study was much shorter, indicating that the elimination of chlorpyrifos in the pre-dam system was very effective.

When the retention time was extended to 5 days, almost no traces of chlorpyrifos were detected in effluent. Apparently, the entire pre-dam system can effectively remove chlorpyrifos.

The highest removal of chlorpyrifos (98.9%) was accomplished by the entire system. The contributions of the different functional areas to chlorpyrifos removal were analyzed. The highest percentage (approximately 56.12%) of chlorpyrifos was removed in the settling area and the second highest was removed in the shallow constructed wetland area at 20.41%. The relative contribution of chlorpyrifos removal in the other areas was 18.37%. These results were observed because the soils in the settled area and the gravel in the wetland had a wide range of moisture content, enhancing the adsorption of chlorpyrifos [23]. Moreover, bacteria may have developed in the shallow wetland area during the operation of the pre-dam system, degrading chlorpyrifos. The relationship between the particle size of SS and chlorpyrifos removal, the root exudates of vegetation in wetlands, and the degradation of chlorpyrifos by bacteria should be studied in the future. The wetland plant debris may affect chlorpyrifos transfer, at least temporarily, as indicated by its high sorption or potential desorption. Chlorpyrifos adsorption and desorption in the deposited sediments and wetland gravel and plants were not addressed in this study.

3.4. Contribution of the different functional zones to the removal of the corresponding pollutants

In the settling area, approximately 80% of SS; 60% of TN, particularly NH_4-N; 55% of TP; and 80% of chlorpyrifos were eliminated through chemical sorption and precipitation [23]. In the shallow wetland area, the water flow was streamlined by different densities of emergent aquatic plants (*Reeds*), which accelerated the settling of smaller particles. In the profundal zones, submerged plants (*H. varticillata*) increased the uptake of nutrients. After passing through the previous areas, the water flowed to the deep wetland area to further remove SS and nutrients, especially TDP and chlorpyrifos, through the combined chemical, biological, and plant mechanisms. Finally, considering the low NO_3-N removal efficiency, the water flowed to a biological membrane purification area, where a biofilm could be developed to treat wastewater with a high NO_3-N concentration.

Under the conditions of this study, the chlorpyrifos and solid particles were possibly bonded on site. Thus, experiments with tapered bottles were performed to determine the relationship between chlorpyrifos and suspended particles. After shaking and standing the bottles for 36 h, we added increasingly more suspended particles, and gradually, less chlorpyrifos was detected in the supernatant liquor. Thus, the predominant association of chlorpyrifos with suspended particles can be inferred, suggesting the importance of capturing and retaining the particles to maximize chlorpyrifos removal in the pre-dam system. However, the quantitative effects of suspended particles on chlorpyrifos removal should be further studied.

As previously discussed, the majority of the large particles were deposited in the settling area. The sediment in the top sediment layer (0–5 cm) was visually inspected. Sediments of approximately 3 cm thick were deposited in the settling area after a month of operation. Thus, sediments have to be removed regularly from pre-dams to prevent the removed pollutants from returning to the system.

In this study, only small contributions to the removal of agricultural nonpoint source pollution in profundal zone 1 and profundal zone 2 were observed. One reason for this result is that developing a complete and active ecosystem to remove pollution in a laboratory model is difficult. However, another reason could be that the contribution of *H. varticillata* to the contamination removal is actually very small. Thus, if agricultural nonpoint source pollution with high SS and chlorpyrifos content must be treated, enlarging the area of the settling basin and wetland is recommended; if water with high TN or NO_3-N content is treated, a biological membrane purification area should be added. In brief, the pre-dam in this study has six subunits to eliminate agriculture nonpoint source pollution, and the area of each subunit should change depending on the characteristics of the specific local pollutants.

It should be pointed out that in this study rather than focusing on the removal mechanism of the pollutants we focused on the removal efficiency and relative contributions of each subunit of the pre-dam to the removal performance. Therefore, based on different contributions of each pollutant in each functional area, we can optimize the functional areas to achieve the best overall performance of the pre-dam system for pollutant removal. If the contribution of removing one specific pollutant is very small, according to this study, we can reduce the size of the corresponding functional area. Furthermore, this study was a laboratory study, thus the scale was different from the actual full-scale project. The SS removal efficiency depends on the water flow rate and system configuration, which determines the flow pattern. However, the removal mechanism of other pollutants such as nutrient and chlorpyrifos are biochemical reaction, which would not change with the pre-dam scale. Thus, the operational parameters and suggested optimal functional areas in the pre-dam systems in this study can still provide valuable references to the practical engineering project. However, in the next step, the processes of operating in each subunit should be further studied to gain better understanding of the whole performance in the pre-dam.

4. Conclusions

The pre-dam system is an important contributor to improving the removal of agricultural nonpoint source pollution from water systems. The efficiency rates of the pre-dams are high, especially for the removal of chlorpyrifos, SS, and NH_4-N at 98, 95, and 85%, respectively. Retention times affect the purification process of agricultural nonpoint source pollution. The highest removal occurred in the settling basin when the retention time was 1–3 days. However, even with an increase in the retention time, high removal efficiency was observed for the same substances. Different functional areas contribute in differing relative proportions to the removal of pollutants. SS, NH_4-N, and chlorpyrifos were removed in large amounts in the settling and wetland areas at 80, 88, and 56%, respectively.

Pre-dam systems or settling zones are important for removing SS, nutrient, and other pollutants attached to SS. For this reason, to control nonpoint source pollution (agricultural or others), SS removal should be given increased research attention. However, a pre-dam system cannot eliminate all contaminants flowing into the system such as NO_3-N. Further research should focus on optimizing the use of the functional areas in a pre-dam system to determine the highest efficiency for eliminating agricultural nonpoint source pollution for different operational times and water qualities.

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