

Advanced phosphorus removal for secondary effluent using a natural treatment system

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ABSTRACT

Mechanisms for low concentrations phosphorus removal in secondary effluent were studied, and a process was developed using limestone filters (LF), submerged macrophyte oxidation ponds (SMOPs) and a subsurface vertical flow wetland (SVFW). Pilot scale experimental models were applied in series to investigate the advanced purification of total phosphorus (TP) in secondary effluent at the Chengjiang sewage treatment plant. With a total hydraulic residence time (HRT) of 82.52 h, the average effluent TP dropped to 0.17 mg L^{-1} , meeting the standard for Class III surface waters. The major functions of the LF were adsorption and forced precipitation, with a particulate phosphorus (PP) removal of 82.93% and a total dissolved phosphorus (TDP) removal of 41.07%. Oxygen-releasing submerged macrophytes in the SMOPs resulted in maximum dissolved oxygen (DO) and pH values of 11.55 mg L^{-1} and 8.10, respectively. This regime provided suitable conditions for chemical precipitation of TDP, which was reduced by a further 39.29%. In the SVFW, TDP was further reduced, and the TP removal in the final effluent reached 85.08%.

Key words | advanced treatment, phosphorus, purification, secondary effluent

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INTRODUCTION

In China, the Emission Standard for Urban Sewage Water Treatment Plant (GB18918-2002) constrains effluent total phosphorus (TP) concentration to no higher than 1.0 mg L^{-1} in the most stringent Grade 1-A. However, the ordinary Surface Water Quality Standard (GB3838-2002), which divides surface waters into five classes with total phosphorus values of 0.02, 0.10, 0.20, 0.30, and 0.40 mg L^{-1} , has a much lower allowable TP concentration. Therefore, acceptable secondary effluent will still pollute surface waters and should be further treated before being discharged into natural waters. Additionally, purified secondary effluent is of high reuse value, especially in drought and water-deficient areas. In this paper, we aimed to develop a low-cost and efficient process to further treat secondary effluent to reach the third class of GB3838-2002, and prevent treated effluent from being a pollutant to most rivers and lakes.

International studies on advanced purification of secondary effluent are continually developing. Early in 1964, alum flocculation and lime precipitation were used by Malhotra *et al.* (1964) to dispose of secondary effluent; in the 1970s, filtration (Biskner & Young 1977) and soil

treatment (Rice 1974; Lo & Clayton 1978) began to prevail; in 1981, membrane technology was adopted by Inoue *et al.* (1981) to conduct advanced purification of secondary effluent. Once mature, membrane technologies were more widely applied in the reuse of industrial water (Masson & Deans 1996; Oron *et al.* 2004; Gozalez-Zafrilla *et al.* 2008). Such processes encountered easy clogging or high operation costs, which caused difficulties in practical application.

Related studies of secondary effluent treatment using natural ponds and wetland systems have become popular since 1988 (Brodrick *et al.* 1988; Vidales-Contreras *et al.* 2010). TP in secondary effluent that meets the discharge standard of Class 1-A is generally around 1 mg L^{-1} . Wetland treatment processes designed for this range of TP have appeared in recent years.

Pilot-scale constructed wetlands were used by Thomas *et al.* (1995) to dispose of secondary treated sewage effluent using four wetland units growing different species of plants, resulting in a P removal of only 13%. A free surface flow wetland with an area of $2,800 \text{ m}^2$ was adopted by Lu *et al.* (2009) to deal with agricultural runoff. With a

hydraulic retention time (HRT) of 2 d and an influent TP of 0.87 mg L^{-1} , the removal reached 59.0%. The limited capacity of a single type of wetland prevented it from further reducing TP. By using only one single wetland type, phosphorus removal was bound to encounter a bottleneck when low effluent concentration of TP was required. In contrast, composed wetlands possess complementary advantages allowing phosphorus to be removed in different states.

Some research has found that calcium substrates are effective for phosphorus removal. To find a suitable substrate for use in constructed wetlands, Brooks *et al.* (2000) examined P removal using columns filled with wollastonite with HRTs between 15 and 180 h. The average dissolved phosphorus in the outflow was 0.28 mg L^{-1} , while the average influent was 3.4 mg L^{-1} . In another study conducted by Calder *et al.* (2006), three types of sorptive media were selected and mixed as a filter unit after a subsurface flow wetland. It was found that the unit filled with slag and gravel had the greatest effect, with the mean total dissolved phosphorus (TDP) decreasing from 0.72 to 0.27 mg L^{-1} . After a study on the phosphorus removal of a calcium substrate filter between pond systems, Strang described the suitable conditions for the chemical precipitation of dissolved phosphorus, namely, a calcium ion concentration of 60 mg L^{-1} (Strang & Wareham 2006). An alkaline pH range was also conducive to phosphorus precipitation. The pH dynamic process of hydroxyapatite (HAP) precipitation, formed by phosphate and calcium ions, was studied by Inskip & Silvertooth (1988). The precipitation rate constant obtained by calculation was $173 \pm 11 \text{ L}^2 \text{ mol}^{-1} \text{ m}^{-2} \text{ s}^{-1}$. Alkaline conditions often exist in pond systems. It is known that the equilibrium of CO_2 solubility would be shifted with the CO_2 uptake by algae or macrophytes during the photosynthesis. Tadesse *et al.* (2004) found that, under the effect of algal photosynthesis, the pH in wastewater pond systems rose to 8.6~9.4. The maximal pH reached by *Hydrilla verticillata* in pH-drift experiments (Kahara & Vermaat 2003) was found to be close to 10.5, which was similar to the highest values published elsewhere for submerged macrophytes (Maberly & Spence 1983; Sand-Jensen *et al.* 1992; Madsen *et al.* 1996).

Based on the above studies, including the enhanced precipitation associated with calcium substrates and the presence of alkaline conditions in macrophyte pond systems, a process was developed for the advanced treatment of secondary effluent. The process employed the following units in series: limestone filters (LF), submerged macrophyte oxidation ponds (SMOPs), and a subsurface

vertical flow wetland (SVFW). The objective was to examine the applicability of the process for decreasing TP to meet the Class-III surface water standard. In this study, a pilot-scale experimental model was adopted for verification of this process, and favourable phosphorus removal efficiency was achieved.

MATERIALS AND METHODS

Experiment site

The experimental site was located at the Chengjiang Sewage Treatment Plant (CSTP) in Yunnan Province, China. Located in the central region of Yunnan, Chengjiang has a subtropical monsoon climate. During the study period, the mean temperature observed was 16.2°C . The highest temperature in summer was 26.7°C and the lowest temperature in winter was 7.3°C . The temperature was suitable for the growth of wetland plants and water purification with wetland systems.

The process used in this sewage treatment plant was Intermittent Cycle Extended Aeration (ICEAS). The Grade 1-A standard of GB18918-2002 was implemented, and the highest TP concentration limit was 1 mg L^{-1} .

Raw water

The secondary effluent of the CSTP was selected as the subject of advanced purification. The raw water for experiment was pumped from the effluent ditch of the CSTP and stored in the 3.0 m^3 tank (the white cube in Figure 1(b)). To keep a constant water pressure, the pump was always running and the excessive water ran away through an overflow pipe on the upper of the tank; thus the water level was kept at a fixed position. Raw water characteristics are shown in Table 1.

Process units

The process used in the pilot scale experiment was as follows (Figure 1(a)): secondary effluent (Influent#1) to a 4-stage SMOP (units #2, #3, #4, #5, for 4 times area of #1) and then to a subsurface vertical flow wetland (SVFW, unit #6) before being recovered as effluent.

The six experimental units were built with high-density polypropylene (HDPP) tanks (Figure 1(b)). The diameter was 1,300 mm at the bottom and 1,400 mm at the top, and the height was 1,380 mm. The six tanks

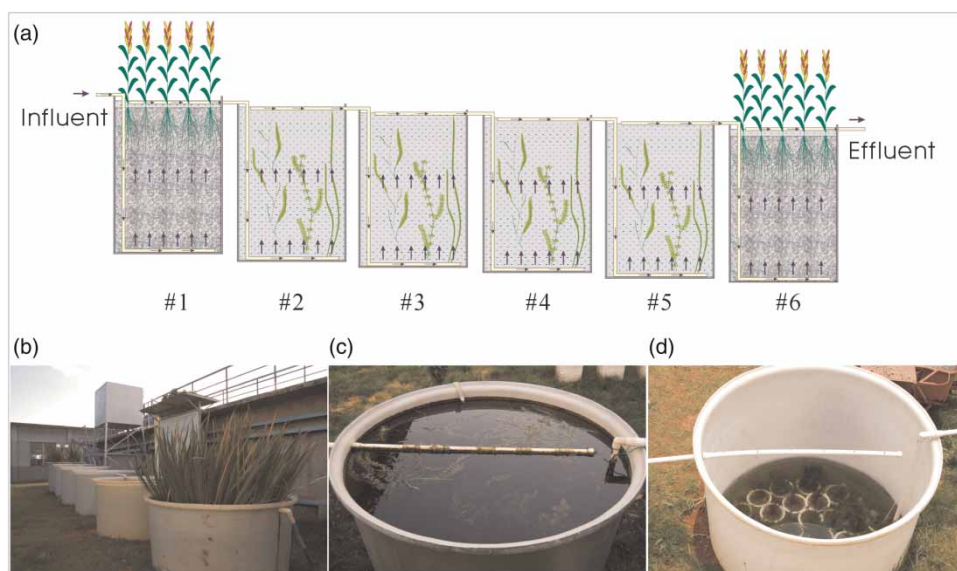


Figure 1 | Pictures of experimental model. (a) Process diagram; (b) Appearance of the model; (c) SMOP unit began to come into effect; (d) The cultivation form of submerged macrophytes.

Table 1 | Average raw water quality^a

Parameters	DO/mg L ⁻¹	pH	SS/mg L ⁻¹	TP/mg L ⁻¹	TDP /mg L ⁻¹
Value	4.56 ± 0.28	7.32 ± 0.10	53.82 ± 13.45	1.14 ± 0.25	0.56 ± 0.11

^aFrom August 20, 2008 to June 10, 2009.

were set into the ground at increasing depths from unit #1 to unit #6. The altitude difference between neighbouring tanks was 100 mm, as shown in Figure 1(b). A 32 mm diameter pipe composed of pentatricopeptide repeats (PPR) was installed vertically along the wall of each tank. The bottom of the inlet pipe was connected to a spiral-shaped water distributor made of 20 mm diameter high-density polyethylene-aluminium composite pipe. Holes were drilled in the water distributor after hydraulic computation, and the end of the water distributor was blocked. The outlet pipe was composed of 32 mm diameter PPR plastic pipe and was installed horizontally 50 mm from the top of the tank. The outlet pipe was connected to the inlet pipe of the next tank. Triangle-shaped weirs were cut in the top side of the outlet pipe to collect outflow water.

Units #1 and #6 were filled with limestone gravel (particle size 30–50 mm, porosity 37%) as the substrate. The height of the gravel substrate was 1,200 mm. During normal operation, a free water layer extended 100 mm above the substrate. The emergent plant *Acorus calamus*

was grown in the limestone in the form of a soilless culture. Units #2, #3, #4 and #5, namely the SMOP units, had the same operation water depth of 1,300 mm. At the bottom of each SMOP unit, there were 30 round plastic containers with a height of 150 mm, upper diameter of 190 mm and lower diameter of 150 mm. The containers were filled with soil collected from Fuxian Lake, and four species of submerged macrophyte were grown: *H. verticillata*, *Ceratophyllum demersum*, *Myriophyllum spicatum* and *Vallisneria spiralis*. Additionally, two to four carp and several shrimps were placed in these units to create an ecological equilibrium.

Water flow in each unit was the same. First, water flowed through the inlet pipe to the distributor pipe and came out from the distributor hole. Then it rose vertically from the bottom of the tank to the surface. After that it came together into the outlet pipe through the triangle-shaped weirs and flowed to the inlet pipe of the next level. The water level in the tank was controlled at about 1,300 mm through controlling the height of the outlet pipe.

Operating conditions

The construction and adjustment of the experimental system began on June 10, 2008. Normal operation started on August 20, 2008. The six units were operated in series and the water treatment capacity was $1 \text{ m}^3 \text{ d}^{-1}$.

Sampling and analysis

Between August 20, 2008 and June 10, 2009, the effluent samples from each unit were collected 57 times in total generally at approximately 10:00 AM. The analysis of water samples included the following: temperature, dissolved oxygen (DO), pH, suspended solids (SS), TP and TDP. Particulate phosphorus (PP) was calculated as TP minus TDP. Chemical analysis was conducted following standard methods (APHA 1998).

On August 18, 2010, the stems and leaves of aquatic plants in units #1 and #6 were harvested. They were weighed before and after drying to obtain wet and dry weights. At the same time, the water in units #2, #3, #4 and #5 was drained. The aerial parts of the submerged macrophytes were collected with scissors and weighed before and after drying. Dried plant samples were milled, and the phosphorus content was determined by a standard method (APHA 1998).

The data presented in the paper are arithmetic mean values, and the error is the standard error. Figures were produced with the software Origin Pro 8.0 SR4.

RESULTS

Phosphorus

Table 2 shows the concentrations and removal of TP, TDP and PP in the raw water and each unit's effluent. The

Table 2 | Different forms of P through the process

Position	TP/mg L ⁻¹	Removal/%	TDP/mg L ⁻¹	Removal/%	PP/mg L ⁻¹	Removal/%
Influent	1.14 ± 0.25	–	0.56 ± 0.11	–	0.41 ± 0.17	–
#1 Effluent	0.43 ± 0.04	62.28	0.33 ± 0.04	41.07	0.07 ± 0.02	82.93
#2 Effluent	0.30 ± 0.03	11.40	0.21 ± 0.03	21.43	0.07 ± 0.01	0.00
#3 Effluent	0.26 ± 0.03	3.51	0.18 ± 0.03	5.36	0.07 ± 0.01	0.00
#4 Effluent	0.22 ± 0.03	3.51	0.13 ± 0.02	8.93	0.07 ± 0.01	0.00
#5 Effluent	0.19 ± 0.02	2.63	0.11 ± 0.02	3.57	0.06 ± 0.01	2.44
#6 Effluent	0.17 ± 0.02	1.75	0.10 ± 0.02	1.79	0.06 ± 0.01	0.00

removal is the ratio of the decrease in concentration of neighbouring units and the concentration in raw water. From Table 2, PP was mainly removed in unit #1, with little removal in the other units. TDP was also primarily removed in unit #1, but other units also contributed to the removal and further reduced the TDP concentration.

SS

Table 3 shows the SS concentration in the raw water and effluent of each unit. The removal is the ratio of the concentration reduction between units and the concentration in raw water.

DO and pH

Figure 2 shows DO and pH values of the raw water and effluent of each unit.

The average influent DO was 4.56 mg L^{-1} . Then DO decreased to 3.43 mg L^{-1} in unit #1, and then increased due to the oxygen production of the submerged macrophytes in the four SMOP units. Some oxygen was consumed in unit #6, and DO fell to 5.84 mg L^{-1} .

The variation of pH showed the same trend as DO. The average influent pH was 7.32. After unit #1, pH decreased to

Table 3 | SS variation through the process

Position	SS/mg L ⁻¹	Removal/%	Cumulative removal/%
Influent	32.67 ± 8.26	0.00	0.00
#1 Effluent	6.06 ± 1.22	81.45	81.45
#2 Effluent	4.06 ± 0.46	6.13	87.58
#3 Effluent	4.22 ± 0.54	-0.49	87.09
#4 Effluent	4.34 ± 0.54	-0.37	86.72
#5 Effluent	4.53 ± 0.59	-0.60	86.12
#6 Effluent	3.54 ± 0.45	3.05	89.18

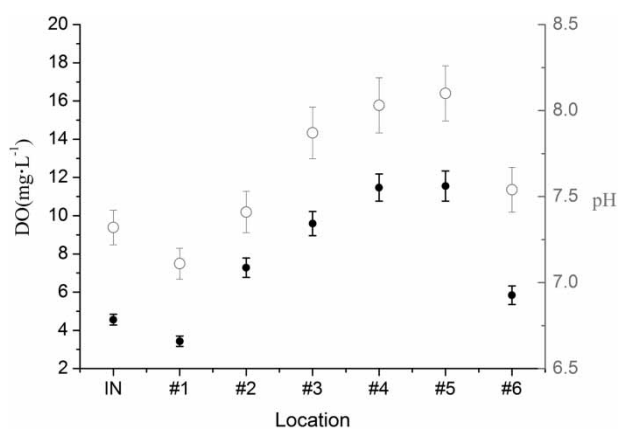


Figure 2 | DO and pH variation through the process

7.11. In four SMOP units, pH gradually increased, before decreasing again in unit #6 to 7.54.

Plant growing and harvesting

Located in the middle part of Yunnan, Chengjiang belongs to the subtropical monsoon climate, with abundant sunshine all the year round. Emergent macrophytes grew lushly in the experimental system (Figure 1(b)). Additionally, submerged macrophytes also grew above the water surface (Figure 1(c)).

Table 4 shows the plants harvested from the six units. The total biomass was weighed during the harvest, but the species were not sorted and weighed separately. The biomass of *Calamus* harvested in experimental models #1 and #6 was 12.20 and 2.11 kg m⁻², respectively. The yield of submerged macrophytes harvested in units #2 to #5 was

Table 4 | Plants harvested from the experimental models

Unit	Total fresh weight/kg	Yield/kg m ⁻²	Phosphorus content/g m ⁻²	Species
#1	18.78	12.20	4.14	<i>Calamus</i>
#2	4.67	3.03	0.53 ^a	<i>Hydrilla verticillata</i> , <i>Ceratophyllum demersum</i> , <i>Myriophyllum spicatum</i> , <i>Vallisneria spiralis</i>
#3	10.13	6.58	0.71 ^a	Same as #2
#4	11.35	7.38	1.78 ^a	Same as #2
#5	13.58	8.82	0.99 ^a	Same as #2
#6	3.24	2.11	0.33	Same as #1

^aSamples of mixed submerged plants were analysed.

between 3.03 and 8.82 kg m⁻². The biomass of submerged macrophytes in units #2 and #3 was less than in the other two SMOP units. In unit #2, a large amount of *Spirogyra* was found, wound around *H. verticillata* and *C. demersum*.

The phosphorus content in *Calamus* was 4.14 g m⁻² in unit #6 and 0.33 g m⁻² in unit #1. The phosphorus content in mixed submerged macrophytes was between 0.53 and 1.78 g m⁻².

DISCUSSION

Phosphorus removal mechanisms

The removal of TDP and PP showed different trends. PP could be precipitated through filtration, sedimentation and other means, while TDP would generally be removed when combined with calcium, magnesium and some metal ions to form PP under alkaline conditions. Therefore, PP removal is relatively easy, and rapid reduction of PP can be obtained through precipitation in LF. TDP is more difficult to remove. When TDP decreases to a certain concentration (as shown in Table 2, about 0.3 mg L⁻¹), further removal becomes difficult.

Without active precipitation measures, the sedimentation of PP is very slow. Ulen (2004) studied the particle-size distribution and sedimentation rate of phosphorus-containing particulate matter. He found that 45% of TP existed in the form of particulates and that the average sedimentation rate was very low, with a range of 0.0034–0.68 cm d⁻¹. Therefore, intensified sedimentation measures were applied in unit #1. PP decreased by 82.93% in unit #1, with only 0.07 mg L⁻¹ remaining in the effluent of #1. At the same time, SS was reduced by 81.45% here. In the subsequent units, the removal of SS and PP was quite low. One cause of the PP removal in unit #1 was due to the adsorption capacity of the limestone substrate of #1. Experiments done by Craft & Eichholz (1970) confirmed that, in a rapid uniform filter, van der Waals forces were dominant for void ratios below 4, while interstitial sieving took effect for void ratios above 4. Adsorption by the roots of plants (Sovik & Syversen 2008) also reduced PP as the water flowed upwards through the plants' roots. The combined effects of these factors resulted in a high PP removal capacity for unit #1.

The phosphorus content in plants was low, between 0.33 and 4.14 g m⁻² (Table 4). Studies have shown that neither absorption by plants nor immobilisation by microbes was the primary mechanism for phosphorus removal

(Mander *et al.* 2003). The TDP removal was mainly achieved through chemical precipitation (Ferguson *et al.* 1973). For chemical precipitation, suitable metal ions and appropriate DO and pH conditions are needed. Suitable metal ions can combine with phosphate and form insoluble substances, such as hydroxyapatite, a calcium phosphate compound. HAP is one of the most insoluble compounds with a solubility product of 6.8×10^{-37} . Strang & Wareham (2006) found that calcium and phosphorus content of sediments in a waste-stabilisation pond used for the removal of TP was appropriate for hydroxyapatite formation. That meant precipitation as hydroxyapatite would be the main incidence for phosphorus. In general, calcium carbonate from the limestone substrate reached dissolution equilibrium in water. Even in the case when pH was higher than 7, sufficient calcium ions were available for the formation of hydroxyapatite (Compton & Pritchard 1990). At the same time, the dissolution and precipitation of calcium on the surface of the limestone substrate also contributed to the precipitation of dissolved phosphorus (Clark & Turner 1955). Under the conditions required for chemical precipitation, DO was always maintained above 3.43 mg L^{-1} . Moreover, the DO in the four SMOPs continuously increased to 11.55 mg L^{-1} , exceeding the saturated concentration (7.54 mg L^{-1} at 20°C). An advantage of the high DO was that the release of phosphorus from bacteria (Kernjespersen *et al.* 1994), sediments, and microbial reactions (Gachter *et al.* 1988) under anaerobic conditions was avoided. Another advantage was that increasing CO_2 uptake by macrophyte photosynthesis increases the pH. The increased pH further promoted an environment for the formation of hydroxyapatite (Ferguson *et al.* 1973). When there are calcium ions and the required alkaline environment, the precipitation of hydroxyapatite occurs.

Treatment units and processes

Based on the mechanisms discussed above, the treatment units developed in the study successfully reduced TP concentration and steadily controlled effluent TP below the limit line for Class-III surface waters. LF was the first unit in the P removal process. Its primary functions are the following: (1) to achieve intensified sedimentation of particles in water through filtration and adsorption by the substrate; (2) to improve water transparency by SS reduction so as to be conducive to the growth of submerged macrophytes in the follow-up SMOP units; and (3) to provide calcium ions for the precipitation of TDP in the following

units by changing the dissolved matter balance and increasing the alkalinity of the water. The function of the SMOPs is to alter the acid-base balance and increase the pH of the water by CO_2 uptake by the submerged macrophytes through photosynthesis. Therefore, it can provide appropriate basic conditions for the chemical precipitation of PP. Additionally, the high DO prevents phosphorus release due to anaerobism. The functions of the SVFW are the following: (1) to provide further TDP reduction and fluctuation buffering capacity so that effluent water quality can be improved to meet the Class-III surface water standard reliably; and (2) to reduce the impact of algae from SMOPs on the effluent through filtration.

The area ratio of LF to SMOPs to VFW used in the process was 1:4:1. The hydraulic residence time in the three sections was 18.94, 44.64 and 18.94 h, respectively. The overall HRT of the process was 82.52 h (3.44 d). The multi-stage nature of the SMOPs had certain significance. They enlarge the area of the SMOP section. According to the ionisation balance and dissolution equilibrium in water, the pH value will be increased to a certain extent (in the SMOPs the effluent pH value of the #5 was 8.10).

Additionally, adequate calcium must be provided to improve the removal of TDP. It would be beneficial to add a small amount of limestone or calcite to the SMOPs so that additional calcium could be provided.

CONCLUSION

In this study, a process using composed wetlands was developed to provide appropriate conditions for the advanced phosphorus purification in secondary effluent based on an understanding of phosphorus removal mechanisms. Pilot scale experiments were conducted with a system composed of an LF, Submerged Macrophyte Oxidation Ponds (SMOPs), and a Subsurface Vertical Flow Wetland (SVFW) connected in series. The HRT of the total process was 82.52 h. The average TP in the final effluent was lower than 0.20 mg L^{-1} , achieving the requirements of Class III surface water quality standard (GB3838-2002).

Though the pilot scale experiment showed good efficiency, it is necessary to conduct long-term tests before application to solve common problems. One problem may relate to the ageing of the wetlands. Substrates in LF and SVFW units readily encounter clogging problems when there are suspended solids in the influent, or when there is a proliferation of microorganisms. Certain measures could be used to clear and wash the substrates; for example,

spraying water and compressed air through a long hollow tube which could be dug into the bottom of the substrates. Then the adhering matter would be washed away and brought up by the air and water stream. Another problem may be uniform water distribution in the full-scale applications. The unit area increases greatly in the real project, which means a large number of distribution pipes should be used in the bottom. Proper measures should be used to ensure uniform water outflow at the beginning and end of the long water distribution pipes.

Additionally, the water quality standard for the effluent of urban waste water treatment plants is growing stricter. We will also encounter problems if we want to further reduce the TDP concentration in secondary effluent to below 0.10 mg L^{-1} . Future work will focus on reducing problems associated with long-term operation and optimising the process to allow application in the future.

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