Optimization of low-cost phosphorus removal from wastewater using co-treatments with constructed wetlands

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Abstract Eighteen wastewater treatment systems were operated for one year to investigate phosphorus (P) removal. Systems paired co-treatment reactors containing iron or calcium drinking water treatment residuals with vertical-flow constructed wetland mesocosms planted with *Schoenoplectus tabernaemontani* (K.C. Gmel.) Palla. For secondary municipal wastewater, soluble reactive P (SRP) concentrations were reduced from 0.70 to 0.03 mg L^{-1} (95%) or 0.01 mg L^{-1} (98%) by systems with the calcium or iron co-treatments, respectively (compared to 0.09 mg L^{-1} or 87% by controls). Total P (TP) concentrations were reduced from 1.00 to 0.07 mg L^{-1} (93%) and 0.05 mg L^{-1} (95%) by the same treatments (compared to 0.16 mg L^{-1} or 84% by controls). For anaerobically digested dairy wastewater, SRP was reduced from 7.68 to 6.43 mg L^{-1} (16%) or 5.95 mg L^{-1} (22%) by the systems with calcium or iron, respectively (compared to 7.37 mg L^{-1} or 4% by controls). For this wastewater, the TP was reduced from 48.5 to 22.5 mg L^{-1} (53%) and 22.7 mg L^{-1} (53%) by the same treatments (compared to 24.1 mg L^{-1} or 50% by controls) but performance improved substantially with a design modification tested.

Keywords Anaerobically digested dairy wastewater; bulrush; drinking water treatment residual; iron; lime sludge; municipal wastewater

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Introduction

The US-EPA has identified agriculture as a source of excess phosphorus (P) damaging surface waters. Management of excess P with conventional wastewater treatment can be cost-prohibitive to the agricultural industry (Sharpley, 1999). Natural wetlands are known to buffer the impact of excess P in effluent and runoff on downstream waters (Reddy and Gale, 1994; Richardson, 1999). Constructed wetlands (CW) are an accepted low-cost technology for removing P from wastewater (US-EPA, 1993). However, questions of mechanisms, predictability and sustainability persist (Richardson, 1999) and there is a need to optimize P removal (Kadlec and Knight, 1996).

Current surface-flow CW treatment systems rely on the sequestration of P in organic and inorganic sediments (Kadlec, 1997) and thus become ultimately unsustainable as the wetland eventually fills in. This process of accretion may take many years. However, treatment wetlands can decline in performance over the years. Although CW may be managed to act as a sink for P, their functional longevity and cost effectiveness are limited by their size. There is also a need to find ways to capture P from wastewater and return it to agriculture as a nutrient source in order to balance inputs and outputs of P in agricultural systems (Sharpley, 1999).

Researchers have examined the use of various materials as potential substrates to improve P removal by wetland treatment systems (Brix *et al.*, 2001; Grüneberg and Kern, 2001). Other researchers have investigated the addition of P-sorbing materials in separate rechargeable cells to improve the ability and sustainability of constructed wetlands to remove phosphorus from wastewater (Arias *et al.*, 2003; Zhu *et al.*, 2003).

By sequestering P with non-toxic materials, it could possibly be reused by agriculture. The use of by-products in co-treatment cells could be a low-cost way to improve the performance and longevity of CW or could be used to reduce the wetland area required for a given level of treatment. Laboratory and greenhouse experiments were used to test several available by-products as potential wetland co-treatment substrates. Based on those results, a 52-week mesocosm study was conducted to evaluate two of the most promising materials at a more realistic scale with both an agricultural and a municipal wastewater. This paper focuses on the effluent phosphorus and relevant total suspended solids (TSS) data from that mesocosm study.

Materials and methods

Co-treatment substrates and wetland root-bed media

Earlier laboratory and greenhouse column experiments with several potential co-treatment substrates guided the design of this mesocosm study. Phosphorus sorption isotherms were conducted with aluminium, iron, calcium and magnesium materials in 100 mg-P L^{-1} solu tions. Phosphorus sorption potentials ranged from 805 to 990 mg kg^{-1}. Lime and iron drinking water treatment residuals (DWTR) adsorbed 893 and 952 mg kg^{-1}, respectively. Factors including turbidity in solution, wide availability, previous use as agricultural soil amendments, and non-toxicity were also considered in addition to P removal performance. The two DWTR were chosen based on all of these relevant factors. The first was a lime sludge (primarily calcium carbonate) by-product of using calcium oxide for drinking water softening. The second was an iron sludge by-product of using ferric sulfate to remove color from drinking water supplies, which is low in sulfur and high in dissolved organic carbon. The mineral fractions of the lime and iron DWTR were 95% silt-sized and 83% sand-sized particles, respectively. However, the iron material held approximately twice the water content, perhaps due to the higher organic fraction. Coarse sand with low P sorption ability (66 mg kg^{-1}) and high hydraulic conductivity was used as the root-bed media in the wetlands.

Wastewaters

Both agricultural and municipal wastewaters were used to test the treatment design over a range of matrix characteristics. These included secondarily treated municipal wastewater from the local wastewater reclamation facility and anaerobically digested flushed dairy manure from the University of Florida Dairy Research Unit. The flushed dairy manure underwent mechanical solids separation and settling prior to being treated in a fixed-film anaerobic digester (Wilkie *et al.*, 2004). Standard methods for wastewater analysis (*Standard Methods*, 1998) were used to measure pH, total suspended solids (TSS), dissolved oxygen, conductivity, salinity and oxidation-reduction potential (E_h) for characterization of the two wastewaters. To measure dissolved organic carbon (DOC), effluents were filtered through 0.45 μm membrane filters before analyzing for non-purgeable organic carbon. Soluble reactive phosphorus (SRP) samples were likewise filtered and analyzed by standard automated colorimetric methods. Total phosphorus (TP) samples were digested in sulfuric acid and potassium persulfate before analysis.

Experiment I: Pre-wetland co-treatment

The experimental systems combined batch-fed co-treatment reactors (CTR) containing either iron or lime DWTR in series with vertical-flow constructed wetland mesocosms (CWM) containing coarse sand and the native soft-stem bulrush *Schoenoplectus tabernae montani* (K.C. Gmel.) Palla (= *Scirpus validus* Vahl) (Figure 1). The horizontal surface area in the CTR was 0.23 m^2 and in the CWM was 0.70 m^2. Eighteen of these systems

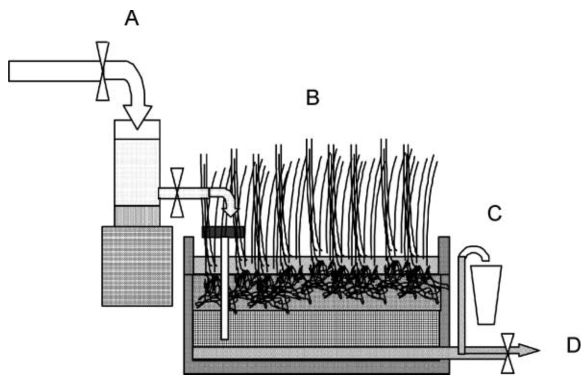

Figure 1 Schematic diagram of the experimental units. Wastewater was loaded to the co-treatment reactor (A) then drained to the constructed wetland mesocosm (B) before exiting the unit (D). A storm event rainfall overflow collection system (C) was employed to ensure that a phosphorus mass balance could be maintained

were built and operated for one year to treat the two different wastewaters, using two DWTR from Florida plus controls, with three replicates of each in a complete randomized design at two sites in Alachua County, Florida, the wastewater reclamation facility and the Dairy Research Unit. Each CTR consisted of a 208 L plastic barrel with a side outlet drain that directed outflow into a CWM. A dry-weight equivalent of 10.4 kg of co-treatment substrate was placed in the barrel below the side outlet drain level to allow wastewater drainage with minimal substrate loss. Both DWTR substrates were used at the water contents as obtained from the water treatment plant storage fields. In practice, drying of the DWTR reduces mass for transport but increases handling costs. The optimal water contents would be site and situation-specific so equivalent masses, rather than volumes, were used in this study. At the equivalent mass, the lime occupied 0.015 m^3 and the iron 0.023 m^3. The CTR substrates at the dairy were replaced every four months (i.e. twice) during the 52-week study as P removal performance declined. They were replaced only once at the municipal site (after 8 months) due to storm damage and not due to a decline in performance. Each control CTR had no substrate added but was filled with water. Each treatment CTR was initially topped off with water, to maintain equivalent volumes. Each CWM was a 567 L plastic tank with a bottom outlet drain. Each tank had a 7.6 cm diameter plastic perforated agricultural drainage pipe in the bottom covered by 10 cm of washed gravel, a geo-textile cloth (to prevent sand seepage), and 20 cm of course sand, and was planted with bulrush. This was a gravity-driven system without electric mixers, aerators or valves. Pumps were only used to move wastewater from the source to the CTR at each site. Clogging was not a problem in the vertically drained CWM sands and drainage rates were unchanged after 52 weeks.

A treatment cycle began by batch-feeding 132.5 L of wastewater to the CTR (Figure 1), where it was held for 7 days. Then the CTR was drained into the CWM, which remained flooded for another 7 days for a total system hydraulic retention time (HRT) of 14 days. This cycle was repeated every 14 days with CTR and CWM cells alternating between flooded and drained conditions. The hydraulic loading rate to the CTR cells was 0.08 $m^3\,m^{-2}\,day^{-1}$ and to the CWM cells was 0.03 $m^3\,m^{-2}\,day^{-1}$ (due to the larger surface area of the CWM). The overall hydraulic loading rate to the combined systems, based on total area and total HRT, was 0.01 $m^3\,m^{-2}\,day^{-1}$.

The average P mass loading rates at the municipal and dairy sites were 0.01 and 0.49 $g\,m^{-2}\,day^{-1}$, respectively. These rates were calculated from the 52-week mean

P mass in the inflowing wastewaters, the total areas of the co-treatment and wetland cells, and the total HRT of 14 days.

Experiment II: Post-wetland co-treatment

The dairy wastewater had much higher TSS and DOC and it was suspected that this reduced the co-treatment's efficiency of P removal from this wastewater. The co-treatments preceded the wetland cells in the 52-week experiment but, for wastewaters with high TSS, the co-treatment may remove P more efficiently when used following an initial wetland cell in the treatment sequence. An additional short-term experiment was conducted using dairy wastewater. Effluents were collected from the three control wetland mesocosms at the dairy. This effluent had lower TSS but had not been exposed to lime or iron co-treatments. Sub-samples of a composite sample of this effluent were added to nine smaller scale co-treatment containers (three each of control, lime, and iron, randomly assigned) with proportionally the same ratio of wastewater to co-treatment substrate as in the 52-week study. The wastewater was held in these containers for 7 days, just as in the larger study, and then analyzed for P concentration.

Results and discussion

Characteristics of wastewaters

Phosphorus and other parameters measured (Table 1) differed by orders of magnitude. The pH was very similar although the buffering capacity differed greatly. The dairy wastewater had been treated anaerobically so oxidation-reduction potential and dissolved oxygen were correspondingly much lower.

Experiment I: Phosphorus removal with pre-wetland co-treatment

For municipal wastewater, SRP concentrations were reduced from 0.70 to 0.03 mg L^{-1} (95%) or 0.01 mg L^{-1} (98%) by systems with the lime or iron co-treatments, respectively (compared to 0.09 mg L^{-1} or 87% by controls). For the same wastewater, TP concentrations (Figure 2) were reduced from 1.00 to 0.07 mg L^{-1} (93%) and 0.05 mg L^{-1} (95%) by the same treatments (compared to 0.16 mg L^{-1} or 84% by controls). For the dairy wastewater, average SRP was reduced from 7.68 to 6.43 mg L^{-1} (16%) or 5.95 mg L^{-1} (22%) by the systems with lime or iron, respectively (compared to 7.37 mg L^{-1} or 4% by controls). The TP (Figure 3) was reduced from 48.5 to 22.5 mg L^{-1} (53%) and 22.7 mg L^{-1} (53%) by the same treatments (compared to 24.1 mg L^{-1} or 50% by controls).

Phosphorus mass removal rates were calculated for CTR and CWM cells separately and combined. For the dairy wastewater, the 52-week mean combined CTR and CWM system phosphorus mass removal rates were very similar for control, lime, and iron systems at 0.27, 0.28, and 0.29 g m^{-2} day^{-1}, respectively. The P mass removal rates by

Table 1 Characteristics of wastewaters loaded to experimental systems at each site

Parameter (units)	Anaerobically digested flushed dairy manure	Secondarily treated municipal wastewater
SRP (mg L^{-1})	3.8–15.6	0.44–1.8
TP (mg L^{-1})	33–49	0.47–2.5
pH	6.7–7.4	6.7–7.4
TSS (mg L^{-1})	2390	<1
DOC (mg L^{-1})	453	<7
DO (mg L^{-1})	~0.1	~8
Conductivity (mS cm^{-1})	4.5	0.7
Salinity (ppt)	2.2	0.3
E_h (mV)	−45	$>+350$

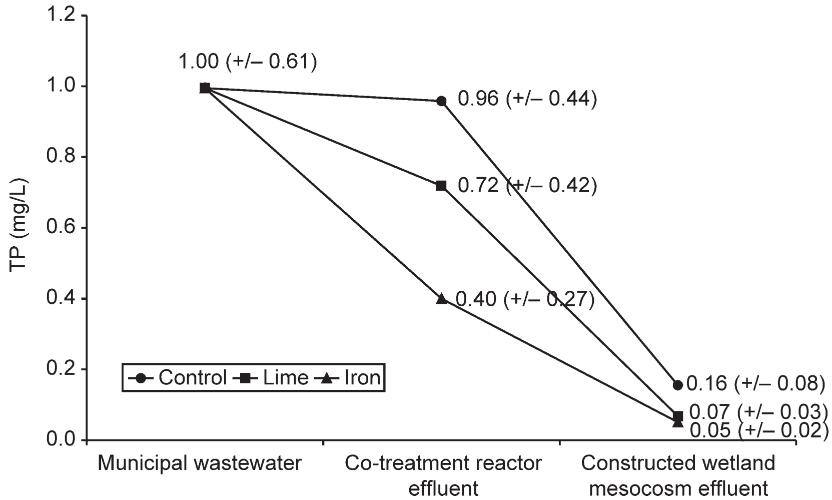

Figure 2 Effluent total phosphorus (TP) (± 1 std. dev.) 52-week means for the municipal wastewater

the CTR cells alone were likewise very similar for control, lime and iron treatments at 1.1, 1.2, and 1.3 g m^{-2} day^{-1}, respectively. For the municipal wastewater, the 52-week mean combined rates for control, lime, and iron systems were also similar at 0.008, 0.009, and 0.010 g m^{-2} day^{-1}, respectively. However, the difference between treatments and controls was more evident in the P mass removal rates by the CTR cells alone. For control, lime, and iron CTR cells at the municipal site, the rates were 0.003, 0.023, and 0.049 g m^{-2} day^{-1}, respectively. The percent P mass reductions by the CTR cells alone also more clearly illustrate the differences between treatments and controls. For the control, lime, and iron CTR cells at the municipal site, the 52-week mean P mass reductions were 4%, 28%, and 60%, respectively. Before loading, the lime had 34 mg P kg^{-1}. When the lime was replaced at the dairy, it averaged 229 mg P kg^{-1}, as compared to only 59 mg kg^{-1} at the municipal site. The iron had 1489 mg P kg^{-1} pre-loading and 1715 mg kg^{-1} post-loading with municipal wastewater. Post-load with dairy wastewater, the iron contained 2216 mg P kg^{-1}. Neither substrate was P-saturated at the municipal site but the iron material may have been at the dairy.

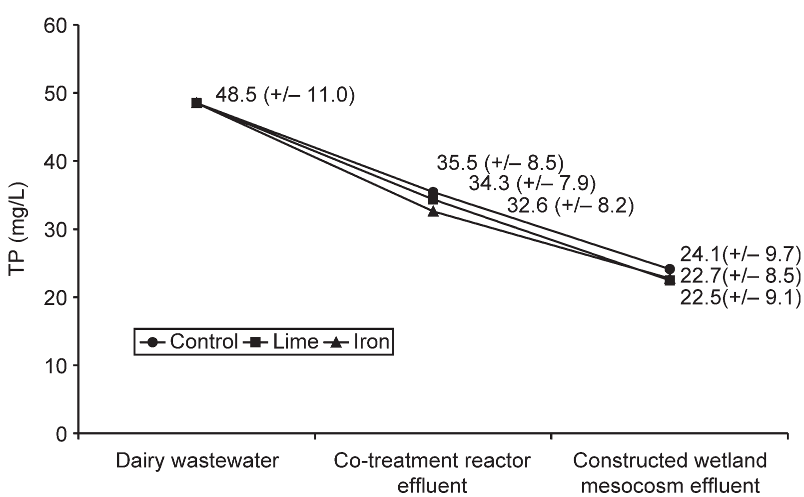

Figure 3 Effluent total phosphorus (TP) (± 1 std. dev.) 52-week means for the dairy wastewater

Newly constructed wetland cells alone are fairly efficient at removing P from wastewater in the first year of operation. This "start-up" effect may have masked the contribution of the co-treatments in this 52-week study since the overall system removal rates were similar for controls and treatments. However, the data from the co-treatments alone suggests that larger differences would eventually be evident in the overall system removal rates. It should also be noted that the P removed by the co-treatments does not stay in the system but is removed when the co-treatment substrates are changed out. This suggests that, in the long-term, wetlands with co-treatments will become saturated with P more slowly and thus have a longer functional lifespan for P removal.

Total suspended solids

Total suspended solids (TSS) were measured in the inflowing wastewaters and the CTR and CWM effluents at each site. The municipal wastewater had less than $1\,mg\,L^{-1}$. The dairy wastewater had relatively high TSS, but these levels were greatly reduced (Figure 4) by the CTR and CWM cells.

Experiment II: Phosphorus removal with post-wetland co-treatment

This experiment supported the hypothesis that placing a CTR after a wetland cell was more effective for P removal from wastewaters with high TSS. One can compare these short-term experimental results with the CTR effluent TP levels of the first week of the 52-week study. Initial TP reductions in the 52-week study at the dairy (Figure 5), with CTR preceding wetlands, were from 43.8 to $25.6\,mg\,L^{-1}$ (41%) and $20.4\,mg\,L^{-1}$ (53%) by the CTR with lime or iron, respectively (compared to $24.8\,mg\,L^{-1}$ or 43% by controls). However, for the post-wetland CTR experiment, initial TP reductions (Figure 6) were from 28.4 to $12.3\,mg\,L^{-1}$ (56%) and $8.1\,mg\,L^{-1}$ (71%) by the CTR with lime or iron, respectively (compared to $24.5\,mg\,L^{-1}$ or 14% by controls). The SRP data also highlighted the contribution of co-treatments with reductions from 7.3 to $3.5\,mg\,L^{-1}$ (52%) and $0.3\,mg\,L^{-1}$ (96%) by the CTR with lime or iron, respectively (compared to $3.8\,mg\,L^{-1}$ or 48% by controls). The TP minus SRP equals particulate P. We observed that particulate P, not just SRP, was reduced by treatments compared to controls. This suggests some particulate P was captured by the DWTR and thus, sedimentation and filtration were not the only mechanism of P removal from dairy wastewater.

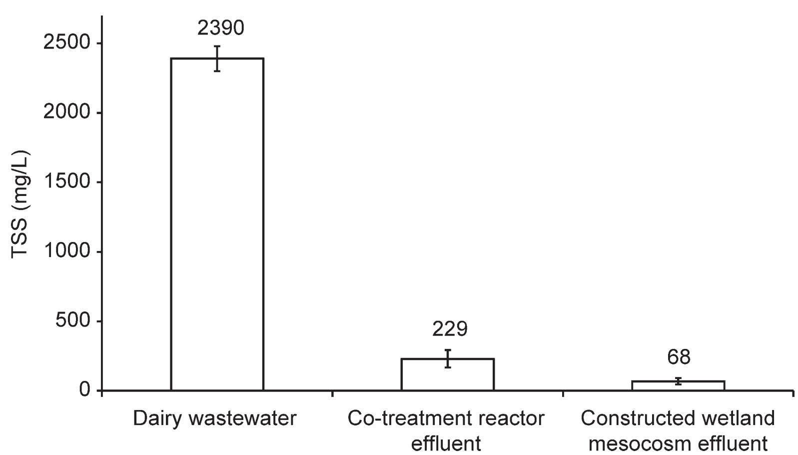

Figure 4 Mean total suspended solids (TSS) ($\pm$1 std. dev.) in effluents at dairy site. Means include all replicates of controls and treatments

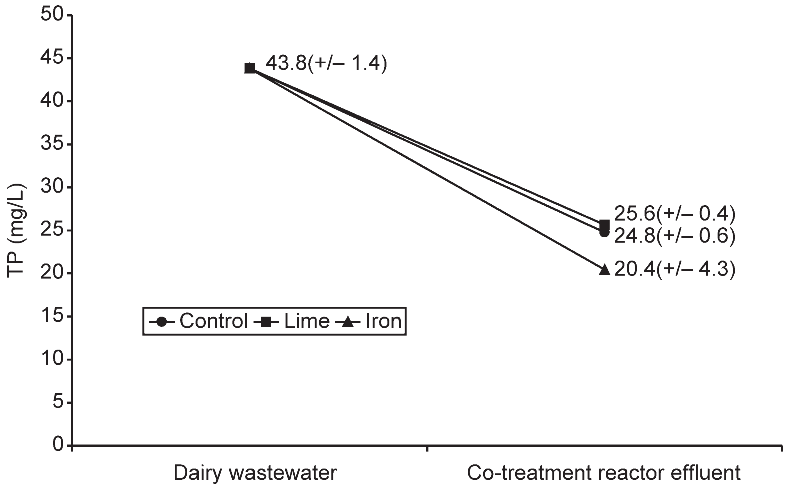

Figure 5 Total phosphorus (TP) (± 1 std. dev.) in pre-wetland CTR effluents for first cycle only of 52-week study with dairy wastewater

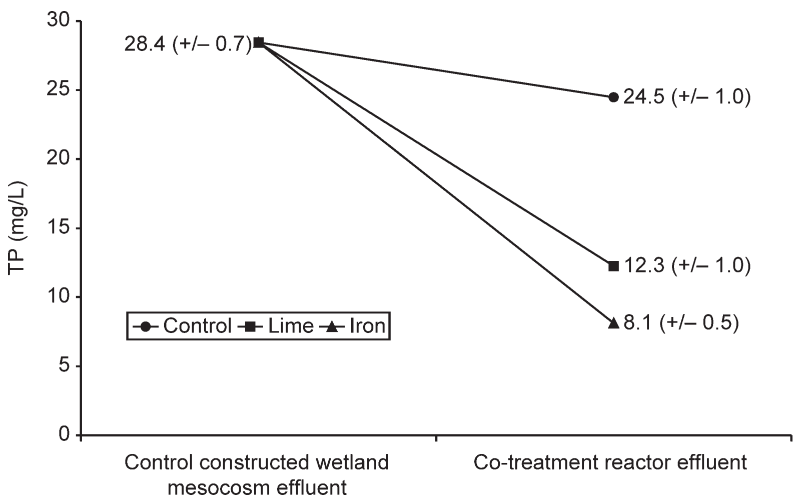

Figure 6 Total phosphorus (TP) (± 1 std. dev.) in post-wetland CTR effluents for the 1-week experiment with dairy wastewater

Conclusions

Wetlands paired with co-treatments generally removed P as well as, or much better than, control wetland systems. Results from the parallel experiments run with both dairy and municipal wastewater provided "proof of concept" for the use of co-treatments with wetlands, and added insight for improved design for specific wastewaters. It was demonstrated that for wastewaters with high TSS, co-treatments would work best when placed after a cell that reduced TSS. As with the standard multiple cell CW systems, an initial settling basin for solids removal would be advisable. Placing the co-treatment after a solids removal cell is compatible with current agricultural CW system design recommendations (USDA-NRCS, 2002). Also, to prevent long-term clogging problems surface drained, rather than vertically drained, CW would be more appropriate for treating high TSS wastewaters.

The use of co-treatments with locally available inexpensive and non-toxic by-products, such as the DWTR used in this experiment, has potential for increasing the sustainability of P removal by CW systems. The iron and calcium-based DWTR used were chosen in part based on their potential to be used as agricultural soil amendments and fertilizers after they have been saturated with phosphorus in this system. Both DWTR used in this study have been applied to agricultural lands in Florida. This feature of reuse could increase both the environmental and economical feasibility of this design. A less mechanized, less energy-intensive system that utilizes non-toxic by-product co-treatments with CW, and returns P back to agriculture, could conserve valuable resources. The ultimate application of this wastewater treatment system design will depend on the type and quality of wastewater being treated. Consideration of many local factors would also be required to optimize the design and operation to ensure the economic feasibility and positive environmental impact in a given situation. Co-treatments might be incorporated into either existing or new wastewater treatment systems.

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