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Application of Vegetated Constructed Wetland with Different Filter Media for Removal of Ammoniacal Nitrogen and Total Phosphorus in Landfill Leachate

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Abstract—**Performance of vegetated horizontal subsurface-flow constructed wetland was evaluated for the removal of ammoniacal nitrogen (AN), total reactive phosphorus (TRP), and soluble reactive phosphorus (SRP) from landfill leachate. Four reactors were used, namely RI (granite without vegetation), RII, and RIII which consists of granite and gravel with different sizes respectively and RIV contained sand and 67.5 L of charcoal. RII, RIII, and RIV were planted with cattails. The leachate obtained from the site of Pulau Burung Sanitary Landfill, Penang was introduced into RI at flow rate of 18 mL/min which was continuous to flow through another three reactors. The leachate was analyzed for AN, TRP, and SRP before and after the treatment in each reactor by standard methods. The overall average removal efficiency of AN, TRP, and SRP were 86.7, 86.2, and 90.0%, respectively. Reactor IV performed the best for removal of all the parameter studies.**

Index Terms—**Ammoniacal nitrogen, landfill leachate, total phosphorus, vegetated constructed wetland.**

I. Introduction

Landfilling is the main method of municipal solid waste disposal in most countries. The composition of waste deposited in landfills is determined by the consumption habits and waste management systems as well as the changes in commonly used materials in the society [1]. Landfill leachates produced from these areas are classified as problematic wastewaters and represent a dangerous source of pollution for the environment due to its fertilizing and toxic effects [2].

Leachate is a high-strength wastewater formed as a result of percolation of rain water and moisture through waste in landfills. The composition of leachate greatly depends upon the landfill age [3], the quality and quantity of waste, biological and chemical processes that took place during disposal, rainfall density and water percolation rate through the waste in the landfill [4]. Leachate mainly consists of heavy metals, organics with different biodegradation and inorganic matters such as ammonia, sulfate and cationic metals [2]. The leachate produced gives a number of environmental problems primarily due to the heterogeneity of waste composition [5]. Therefore, treatment of the leachate is essential before it could be discharged directly into the receiving water bodies.

One of the sustainable low cost solutions for leachate management is constructed wetland system which utilizes anaerobic and aerobic reactions to break down, immobilize or incorporate organic substances and other contaminants from the leachate [6]. Treatment in the constructed wetland involves several processes such as microbial degradation, plant uptake, sorption, sedimentation, filtration and precipitation [7].

There are two basic designs for constructed wetlands which are subsurface-flow (SSF) and surface-flow (SF) wetlands. In the SSF, the water may flow horizontally (parallel to the surface) or vertically through the matrix and out of the system, whereas, the water moves above the substrates surface in SF [8].

The primary role of vegetation in a wetland treatment system is to recycle nutrients in the waste into a harvestable crop. The vegetation such as cattail, *typha latifolia,* is the support media for biological activity and maintains long term infiltration rates. Besides, the presence of vegetation increases the wetland evapotranspiration efficiency [6]. The plant active reaction zone is the root zone or rhizosphere where physicochemical and biological processes take place. The processes are induced by the interaction of plants, microorganisms, the soil and pollutants [9].

Biodegradable organic compounds and ammonia are constituents of leachate that pose the most significant environmental threats [10]. The processes of removal and retention of nitrogen in constructed wetland include NH_3 volatilization, nitrification, denitrification, nitrogen fixation, plant and microbial uptake, mineralization (ammonification), nitrate reduction to ammonium (nitrate-ammonification), anaerobic ammonia oxidation, fragmentation, sorption, desorption, burial and leaching [11]. Although processes for nitrogen removal are manifold, nitrification-denitrification is the main process for nitrogen removal [12].

Phosphorus occurs in natural waters and in wastewaters almost solely as phosphates. These are classified as orthophosphates, condensed phosphates (pyro-, meta-, and other polyphosphates), and organically bound phosphates. Phosphorus is essential to the growth of organisms and can be the nutrient that limits the primary productivity of a body

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of water. However, if excess phosphate enters the aquatic systems, algae and aquatic plants will grow wildly, choke up the aquatic system and use up large amount of oxygen. This condition is known as eutrophication or overfertilization of receiving waters. The rapid growth of aquatic vegetation can cause the death and decay of vegetation and aquatic life due to the decrease in dissolved oxygen levels [13].

In this study, the horizontal vegetated subsurface-flow constructed wetland was used. The leachate sample was flowed continuously through the four reactors, RI, RII, RIII and RIV. The RII, RIII and RIV were planted with cattails, *typha* latifolia. The removal efficiency of ammoniacal nitrogen and total reactive phosphorus and soluble reactive phosphorus were determined before and after the treatment.

II. Experimental

A. Preparation of media

The charcoal with diameter 4<Ø<10 mm was prepared by burning oil palm shells in furnace for 6 hours at 400 °C. The other filter media consists of granite (10<ϕ<25 mm), gravel (4< ϕ <10 mm), and sand (4<ϕ<10 mm) were washed thoroughly with tap water for 4 times before being used in order to remove all the impurities.

B. . Determination of leachate pH

The pH of leachate was measured with a digital portable HANNA instruments pH 211 Microprocessor pH meter.

C. Setting up of wetlands system

Four reactors, coded as RI, RII, RIII and RIV of dimension 50 cm (width) x 150 cm (length) x 60 cm (depth) were employed in this experiment. RI consists of granite (without plant), whereas, RII and RIII were set up with granite and gravel with different sizes respectively and RIV was filled with sand and 67.50 L of charcoal. Except RI, other reactors were planted with *typha latifolia.* The arrangements of reactors were in descending height order to allow leachate to flow along the wetland system from reservoir to RIV gravitationally. Each reactor was fixed with 5 perforated T-shape tubes as sampling points, labeled as Point 1 (inlet), Point 2, Point 3, Point 4 and Point 5 (Fig. 1).

D. Leachate treatment - determination of ammoniacal nitrogen (AN), total oxidized nitrogen (TNOx-N), and nitrite nitrogen (NO2-N),

Raw leachate collected from landfill site in Pulau Burung, Pulau Pinang was filled into reservoir and let flowed into RI at a predetermined rate of 18 mL/min to fill the RI within 7 days. After 7 days leachate sample in RI was collected from 5 sampling points and analyzed for determination of the concentration of ammoniacal nitrogen (AN), total oxidized nitrogen (TNOx-N), nitrite nitrogen (NO_2-N), total reactive phosphorus (TRP), and soluble reactive phosphorus (SRP). The leachate sample was left to flow into the following reactors by overflow pipe which was connected to the inlet pipe of the following reactors. For every 7 days continuously, the leachate was analysed for all three parameters at every sampling points for all reactors. The amount of rain fall was measured throughout the study.

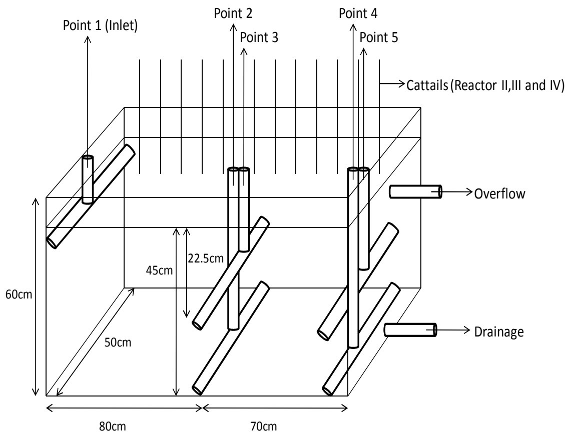


Fig. 1. Schematic diagram of reactor.

Analysis for all the parameters studies were conducted based on the Standard Method for the Examination of Water and Wastewater (APHA, 1985). AN was analyzed using the titrimetric method. Devarda's alloy reduction method was used to determine the concentration of TNOx-N. Whereas, the colorimetric method was used to determine the NO_2-N using UV-Vis Hitachi U-2000 Spectrophotometer at 543 nm with 1 cm cuvettes. Total phosphorus was determined from untreated leachate using the ascorbic acid method. Soluble reactive phosphorus was determined from filtered leachate through Advantec GC 50 glass-fibre filter paper size 70 mm (Toyo Roshi Kaisha, Ltd) using ascorbic acid method. Triplicate of analyses were performed for each sample of each parameter.

III. Result And Discussion

The combination performance of all reactors in the system for removal of ammoniacal nitrogen (AN), total oixidized nitrogen (TNOx-N) and nitrite nitrogen (NO_2-N) are shown in Fig. 2. The average concentration of AN, TNOx-N and NO_2-N at each sampling point for all reactors are shown in Figs. 3(a), (b) and (c) respectively. While the removal percentage of AN, TNOx-N and NO_2-N for each reactor at each sampling point are shown in Figs. 4(a), (b) and (c) respectively.

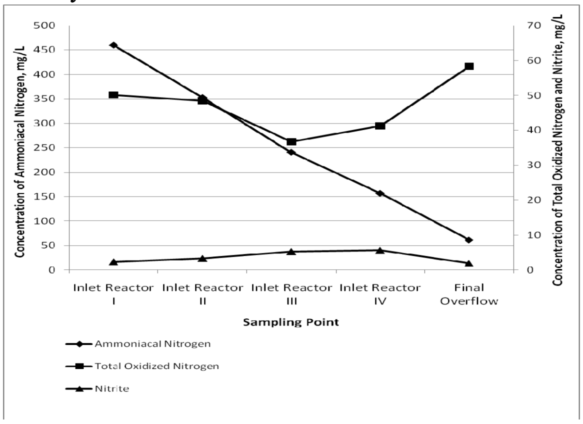


Fig. 2. The concentration of ammoniacal nitrogen, total oxidized nitrogen and nitrite nitrogen for Reactor I, II, III and IV.

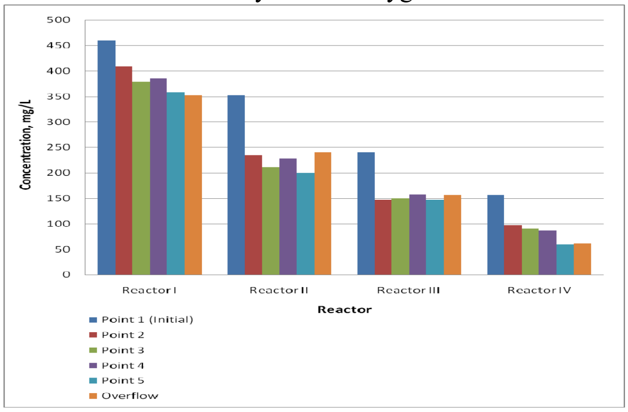
Results show that the average concentration of AN in raw leachate sample is 459.9 ± 52.68 mg/ L and decreased to 61.38 ± 3.206 mg/ L at the end overflow of RIV (Fig. 3(a)). This give the overall percentage removal of AN of 86.7 %. For unvegetated the removal of RI is 23.3 % and increased to 31.7 % of RII and then slightly increased to 34.9 % of RIII. The removal efficiency is the highest for RIV which is 60.9 % (Fig. 4(a)). The overall removal efficiency for AN is the combination of the efficiency of Reactors I, II, III and IV. The result shows that the overall removal efficiency is high when the four reactors are combined into a system.

Process of nitrification-denitrification is the most significant mechanism for biological nitrogen removal in constructed wetland [13]. The removal efficiency depends greatly upon the hydraulic retention time (HRT) which affects the duration of contact between the microbial population within the wetland system and the pollutants such as nitrate and organic compounds [14]. For AN, HRT plays a very significant role on removal efficiency. The longer the HRT, generates the higher removal efficiency of AN [15]. The combined system gives a total HRT of 28 days. As the longer time the leachate sample stays in the system, the nitrification and denitrification processes will be longer. As a result, the overall removal efficiency of AN achieves 86.7 %.

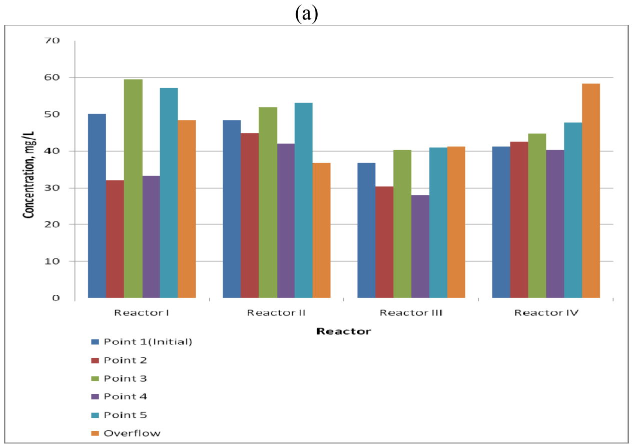
Cattail plants (*Typha latifolia*) which planted in Reactors II, III and IV play an important role in removing biological nitrogen compounds. The roots and rhizomes are hollow and contain air-filled channels that are connected to the atmosphere for the purpose of transporting oxygen to the root system [16]. The removal of AN is largely dependent on the oxygen supply in the media [17]. The plants transport oxygen to the rhizosphere, thereby creating aerobic microsites which is adjacent to the roots and rhizomes where ammonia will be oxidized to nitrite by nitrifying bacteria such as nitrosomonas and then to nitrate by nitrobacter [13].

The main difference between Reactor I, II, III and IV is the type of the media employed and its physicochemical characteristics. The removal efficiency depends on the porous media size and type. Finer porous media are more effective than coarser media [15]. The media with different size have different available surface area for nitrifier biofilm growth and different oxygen transfer rate. Reactor III and IV which fed with gravel of smaller and equal size with sand respectively provide larger surface area for nitrifier growth as compared to Reactor I and II which employ granite as filter media. Surface area for nitrifier biofilm growth can be a limiting factor upon nitrification on highly loaded system. However, inadequate of oxygen transfer rate are more likely to limit nitrification than substrate surface area availability [18].

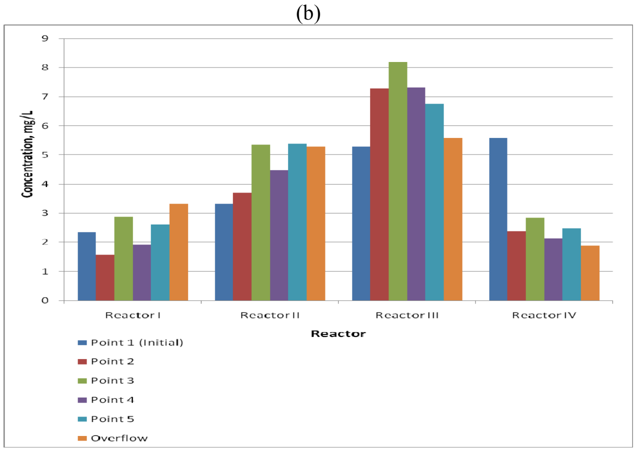
Aerobic and anaerobic processes are the main processes occur in each reactor of constructed wetland [19]. Five sampling points in each reactor where Point 3 and 5 are located at middle part of the reactor, while, Point 2 and 4 are located at bottom part of the reactor (Fig. 1). For AN removal, Point 3 and 5 show lower concentration of AN compared to Point 2 and 4. This is because Point 3 and 5 are located in aerobic region which is preferable for nitrification to occur. Most of the AN lost as nitrite and nitrate by nitrification process at Point 3 and 5 by used of oxygen.



(a)



(b)

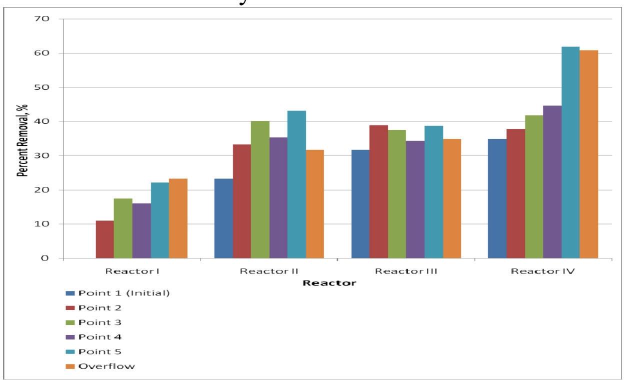


(c)

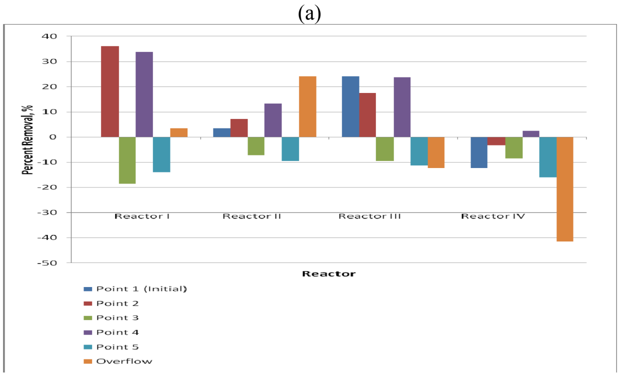
Fig. 3. The average concentration of (a) AN, (b) TNOx-N and (c) NO_2-N for Reactor I, II, III and IV at different sampling point.

At bottom part of the reactor which is anaerobic region, the removal of biological nitrogen is mostly due to conversion of nitrogen compounds to different form by denitrification process which is carried out by denitrifier bacterial. This process is supported by the lower concentration of TNOx-N at Point 2 and 4 as compared to Point 3 and 5. Anaerobic is the condition which lack of oxygen supply. Depletion of amount of dissolved oxygen could be proposed that denitrification is the major removal process [20]. In denitrification process, NO_3-N and NO_2-N are reduced to AN and further to N_2. Therefore, the concentration of TNOx-N is lower at anaerobic region.

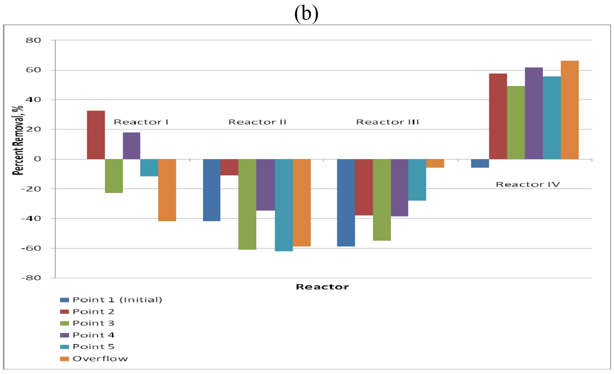
The average concentration of TNOx-N in raw leachate sample is 50.17 ± 12.83 mg/ L and increased to 58.33 ± 9.934 mg/ L (Fig. 3(b)). This result gives a negative effect for overall percent removal that is -16.3 % (Fig. 4(b)). Whereas, the average NO_2-N concentration in raw leachate sample is 2.345 ± 0.5950 mg/ L and decreased to 1.888 ± 0.4858 mg/ L at the end of the system (Fig. 3(c)). The overall percent removal of NO_2-N is 19.5 % (Fig. 4(c)). The result obtained shows that as there is a decrease in concentration of TNOx-N, the concentration of NO_2-N will be increased and vice versa. Besides, the data shows that there is NO_2-N accumulation in Reactor II and III. However, as the leachate sample is reached Reactor IV, the accumulation of NO_2-N is oxidized to NO_3-N. This is shown by the increased in average concentration of TNOx-N and decreased in average concentration of NO_2-N at the end of the system. The rate of conversion of NO_2-N to NO_3-N is low in the system. The average concentration of TNOx-N at Point 2 and 4 is lower compared to Point 3 and 5. This result indicated that at the Point 2 and 4 which located at bottom part of the reactor the denitrification process was enhanced and eventually reduced the amount of TNOx-N.



(a)



(b)



(c)

Fig. 4. Percentage removal of (a) AN, (b) TNOx-N and (c) NO_2-N for Reactor I, II, III and IV at different sampling point.

Ammonia volatilization is a physicochemical process. Losses of AN through volatilization are not serious if the pH value is below 8.0. But, if the pH values rise to values as high as 8.0 – 8.5, the process of ammonia volatilization increases [21]. In this study, the pH value of the leachate sample is 8.0. Therefore, the losses of AN through volatilization are possible. This process will cause the lost of amount of AN in the system.

From the data obtained, the role of charcoal in removing biological nitrogen in constructed wetland is less significant. Hence, the removal of nitrogen in this system mainly due to nitrification-denitrrification, plant uptake and media adsorption.

Fig. 5 shows the average concentration of total reactive phosphorus (TRP) in five trials for every sampling point which was indicating a decreasing trend from R1 to R4. The initial concentrations of TRP were in the range of 30-50 mg/L and after flowed through the reactors, the final concentration was reduced to less than 10 mg/L. The result obtained was in the agreement to results reported by Arda Yalcuk [2]. The average percentage removal is shown in Fig. 6, where the higher removal caused by the presence of the cattail plant and microorganisms which would uptake the phosphorus [23], [24]. It was believed that plant litter, providing additional organic material and thereby new sites for phosphorus adsorption was responsible for a better mass removal performance of the planted systems [25]. The average percentage removal for five trials in R3 was higher than R2 because the phosphorus was taken up by the cattail plant and the phosphorus adsorption capacity increases as the size of the gravel is smaller as compared to the size of the granite in R2 [26]. In R4 which was filled with the sand and charcoal and planted with cattail plant showed the highest average percentage removal, 67.7 %. This is because sand is a better media for a removal of phosphorus from the leachate as compared to granite and gravel [27].

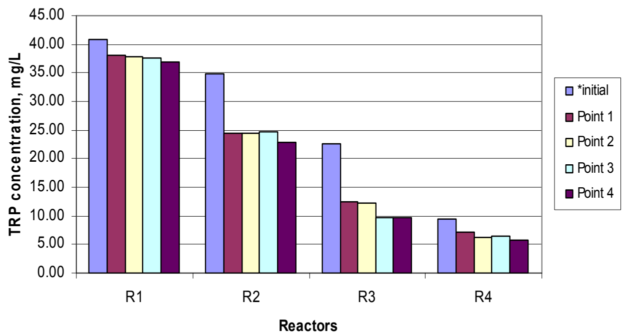


Fig. 5 Average concentration of TRP at various sampling points.

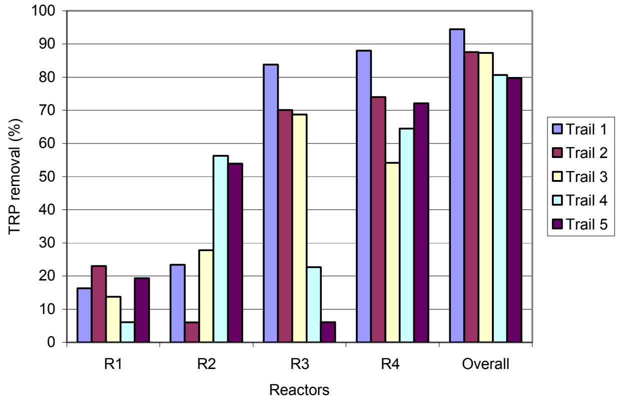


Fig. 6. Average percentage removal of TRP for series of treatments.

For soluble reactive phosphorus (SRP), the average concentration and percentage removal at each of the sampling point shows similar trend as for the TRP (Figs. 7, 8). This indicated that with the presence of cattail plant which would uptake the phosphorus by absorption process [23], [24]. Microbial uptake of phosphorus had been claimed that biotic processes have a considerable effect on the phosphorus removal which was influenced by the bioavailable carbon source in the sediment [28]. Results show that the availability of adsorption sites due to smaller media sizes which provided higher surface area that gave bigger capacity for the phosphorus binding site [26].

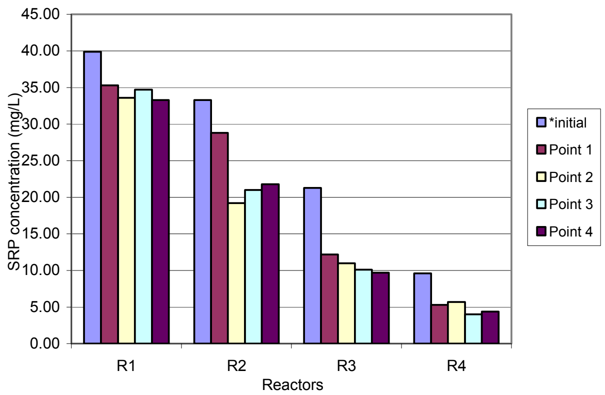


Fig. 7. Average concentration of SRP at various sampling points.

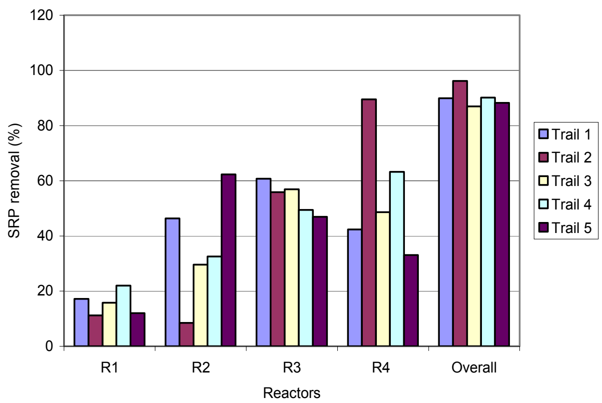


Fig. 8 Percentage removal of SRP at various reactors and on overall.

IV. Conclusion

Combination of four reactors into a combined system increased the hydraulic retention time and the overall removal efficiency in the constructed wetland. Nitrification coupled with denitrification is the main process occurs in biological nitrogen removal. Besides, cattail plant plays a very significant role in removing nitrogen in the system. Aerobic and anaerobic condition located at different sampling points in reactors greatly affects the efficiency removal of nitrogen.

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