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ORIGINAL ARTICLE

Application of Phytoremediation Techniques to Treat Reverse Osmosis Concentrate using Wetlands

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Abstract

Reverse Osmosis (RO) plants have been introduced by many organizations to Chronic Kidney Disease of Unknown Etiology (CKDu) affected areas as an effective drinking water treatment method. The liquid byproduct of RO system known as RO concentrate or reject is considered as the major environmental and economic drawback of RO process and normally released to the environment without any treatment. Main objective of this study was to investigate the potential of phytoremediation techniques in treating RO concentrate through pilot scale constructed wetlands (CWs). Four plant species; Vetiver (*Vertiveria zizanioides*), Cattail (*Typha augustifolia*), Cannas (*Canna indica*) and Bulrush (*Scirpus californicus*) were planted in small plastic containers (60 x30 x 30 cm) and soil without amendments was served as the control. There were 3 replicates for each treatment. The experimental units were treated with continuous flow of concentrates obtained from RO plant installed in Faculty of Agriculture, Rajarata University of Sri Lanka at a rate of 2.3 ml s^{-1} for three months period. The hydraulic retention time was 52 h. Water samples were collected from inlets and outlets of each experimental unit by two weeks interval and analyzed for pH, Electrical Conductivity (EC), Total Dissolved Solids (TDS), concentration of Na^+, K^+, Ca^{2+}, Mg^{2+}, NO_3^--N, PO_4^{3-}-P and NH_4^+-N. The results revealed that removal efficiencies (RE) of all pollutants were increasing with time. Cattail plants showed highest removal efficiencies for PO_4^--P, NO_3^--N and NH_4^+-N by 45%, 30% and 39% respectively. Sodium Adsorption Ratio of all treatment plants were within the irrigation water quality standards. Therefore, it can be concluded that the quality of RO concentrate can be improved using phytoremediation techniques.

Keywords: *Irrigation water quality, Phytoremediation, Removal efficiency, Reverse Osmosis concentrate*

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1. Introduction

In recent years, a significant number of patients has been observed with Chronic Kidney Disease of unknown etiology (CKDu) especially in North Central Province of Sri Lanka. Highest prevalence of CKDu occurs in the largest rice farming areas in Sri Lanka and it is reported that approximately 99 % of CKDu patients are farmers (Paranagama et al. 2013). The high content of fluoride in groundwater, contamination of the water supply with artificial fertilizers used for paddy cultivation, use of aluminum utensils instead of clay pots for cooking and toxin released from blue green algae are some of the suspect reasons for the crisis (Abeygunasekera and Wickremasinghe 2013).

Groundwater is the main drinking water source for the dry zone of Sri Lanka and more than 85% of drinking water demand is fulfilled from shallow and deep wells (Paranagama et al. 2013). Many researches revealed that there is a significant relationship between water quality and occurrence of CKDu in the dry zone. Therefore, Reverse Osmosis (RO) plants were established all over the North Central Province by various government and non-government organizations to provide safe drinking water to the community.

RO systems remove much smaller dissolved particles than ultra-filtration or any carbon filters. Unlike the latter two, the RO systems remove trace elements, such as cadmium, arsenic, lead, copper and volatile organic compounds, sodium, nitrates, phosphate, fluoride, total dissolved solids (TDS) and agrochemicals (Wimalawansa 2013). Therefore, the RO technology is an important solution for generating safe potable water.

Reverse osmosis concentrate is a major problem in treatment plants due to its high salinity and possible content of toxic elements (Shanmuganathan 2016). The chemical characteristics of RO concentrate is highly depending on the quality of raw water source, RO pre-treatment methods used and the mode of RO system operation (Squire 2000). Discharging RO reject streams into the natural environment without any treatment can cause severe environmental complications and pollution of groundwater by salts and harmful chemicals (Qurie et al. 2013). Hence, it is necessary to improve the quality of RO concentrate by using simple and cost-effective method before releasing into the outer environment. This pilot study was aimed to investigate the possibility of improving the quality of RO concentrates by using phytoremediation techniques with the use of model wetland units.

2. Methodology

Phytoremediation is one of the biological wastewater treatment method which can be defined as a method which efficiently use of plants to remove, detoxify or immobilize environmental contaminants in a growth matrix (soil, water or sediments) through the natural biological, chemical or physical activities and processes of the plants (Unep.or.jp, 2018). The selection of appropriate plants is most important in implementing phytoremediation. The plants should have high uptake of both organic and inorganic pollutants, grow well in polluted water and easily controlled in quantitatively propagated dispersion. Based on phytoremediation capabilities and availability of plant materials, *Vertiveria zizanioides* (Vertiver grass), *Typha augustifolia* (Cattail), *Canna indica* (Cannas) and *Scirpus maritimus L.* (Bulrush) were selected for the study.

2.1 Study Location

The study was conducted using RO concentrate discharged from the water purification plant installed in Faculty of Agriculture Rajarata University of Sri Lanka. Average daily intake of the RO plant is 4000 L and about 3000 L of RO concentrate is discharged back to the nearby lowlands without any treatments.

2.2 Wetland Design

Plastic containers with 60 cm length, 30 cm width and 30 cm depth were used as experimental units. Inlets of the wetland units were set at the top of over end of the container and the outlets were set at the bottom (Fig. 1). Each container was filled with 5 cm soil aggregates layer and 15 cm top soil layer as the substrate with the pH value of 8.02. Four plant species: *Vertiveria zizanioides* (Vertiver grass), *Typha augustifolia* (Cattail), *Canna indica* (Cannas) and *Scirpus maritimus L.* (Bulrush) were planted in small scale constructed wetland while one unit was left plant-free as the control. Three replicates were used for each treatment.

Figure 1: Experimental wetland unit

2.3 Plant Establishment

Bulrush, Cattail and Vertiver shoots with rhizomes and Cannas rhizome cuttings were planted in polythene bags and after 2 weeks, well grown, healthy and uniform plants were selected for the experiment. Vertiver and Bulrush were established at the spacing of 15 cm x 30 cm while Cannas and Cattail were established at the spacing of 30 cm x 30 cm. There were 06 plants in each Vertiver and Bulrush containers and 03 plants in each Cannas and Cattail containers.

2.4 Water Supply

A continuous flow of concentrate (2.3 mls^{-1}) was maintained to each experiment unit throughout the study period. The hydraulic retention time of the system was 52 h. The pipe line system over the wetland units was maintained the continuous flow of concentrate from storage tank using gravitational force.

2.5 Sample Collection

Water samples were taken from both inlet and outlet of each experimental unit at two weeks interval for three months into 250 ml water sampling bottles. Soil samples were taken before planting and two months after planting from each and every experimental unit to investigate the effect of RO reject on surrounding soil. Collected samples were packed and labeled separately.

2.6 Sample Analysis

Water quality parameters of influent and effluent such as pH, Electrical Conductivity (EC), Total Dissolved Solid (TDS), phosphate phosphorous (P), nitrate-N, ammonium-N, selected trace element content (Cd, As, Pb) and concentration of Na^{+}, K^{+}, Ca^{2+}, Mg^{2+} were assessed using standard methods. Soil pH, Electrical Conductivity (EC), available phosphorous and available nitrogen content were also analyzed. The analysis was done at the soil and water science laboratory in Faculty of Agriculture, Rajarata University of Sri Lanka using following methods (Table 1).

Table 1: Methods of soil and water analysis

Parameter	**Method of Analysis**
pH, EC, TDS	Multi-parameter analyzer (Hach Sension 156) Soil samples; 1:2.5 and 1:5 soil water suspension
Phosphates (PO_4^{3-})	Olsen method (Olsen et al. 1954)
nitrate-N (NO_3^--N)	Sodium salicylate method (Yang et al. 1998)
ammonium-N (NH_4^+-N)	Phenate method (EPA 1993)
Trace elements (As, Cd, Pb), Na^+, K^+, Ca^{2+}, Mg^{2+}	Spectrophotometry (ICP-OES, iCAP 7000 series) Soil samples; spectrophotometry after acid digestion of soil samples (Martin et al. 1994)

2.7 Data Analysis

Collected data were analyzed by the Analyses of variance (ANOVA) using Statistical Analysis System (SAS) to check whether the means are significantly different from each other. Data were graphically represented using Microsoft Excel according to the mean values.

Removal efficiency of different types of pollutants was determined by using following equation.

Ex:

$$\text{P removal efficiency} = \frac{\text{Inlet [P]} - \text{Outlet [P]}}{\text{Inlet [P]}} X\ 100\%$$

3. Results and Discussion

3.1 Characterization of RO Concentrate

According to the analysis of RO concentrate as shown in table 02, average pH of the RO concentrate was 7.95 and all the recorded values were in the permissible range (6.0 – 8.5) to be released into the environment followed by National Environmental Act, (No.47 of 1980) 2008. The average EC value was 0.76 dS/m According to the irrigation standards recommended by Ayers and Wescot (1985) EC values of RO concentrate is within the usual range of irrigation water. Also the PO_4^{3-}-P, NO_3^--N and NH_4^+-N concentrations of RO concentrate were not exceeding the FAO irrigation water quality standards.

Table 2: Characterization of RO concentrate

Parameter	Average ± SD	Minimum	Maximum	Usual range in irrigation water *
pH	7.95 ± 0.1964	7.80	8.32	6.0 – 8.5
EC (dS/cm)	0.76 ± 0.037	0.707	0.789	0 – 3
TDS (mg/L)	379.3 ± 26.4	342	414	0 – 2000
PO_4^--P (mg/L)	0.04467± 0.020	0.01695	0.07128	0 – 2
NO_3^--N (mg/L)	4.143 ± 1.433	3.369	6.413	0 – 10
NH_4^+-N (mg/L)	0.0409 ± 0.01183	0.02104	0.05043	0 - 5

*Source- Ayers and Wecot (1985)

3.2 Electrical Conductivity (EC) and Total Dissolves Solids (TDS)

According to Fig.2(a), there is a considerable decrease of electrical conductivity between inlet and outlet of each treatment unit during the monitoring period. This reduction can be attributed to the plant uptake of ions as nutrients.

The results revealed that there is a significant difference ($p < 0.05$) of reduction of EC values with five treatments. All the treatments showed higher reduction of EC at 12 weeks after planting. However, the reduction of EC was significantly higher ($p < 0.05$) in Cattail and Cannas while the control (bare soil) showed the lowest reduction.

According to the Fig.2(b), there is a substantial decrease of TDS between inlets and outlets of each treatment unit. The decrease in TDS in the effluent samples might be due to the improvement of filtration ability by root system of the growing plants and biodegradation in the root network (Udom, I.J. et al. 2018).

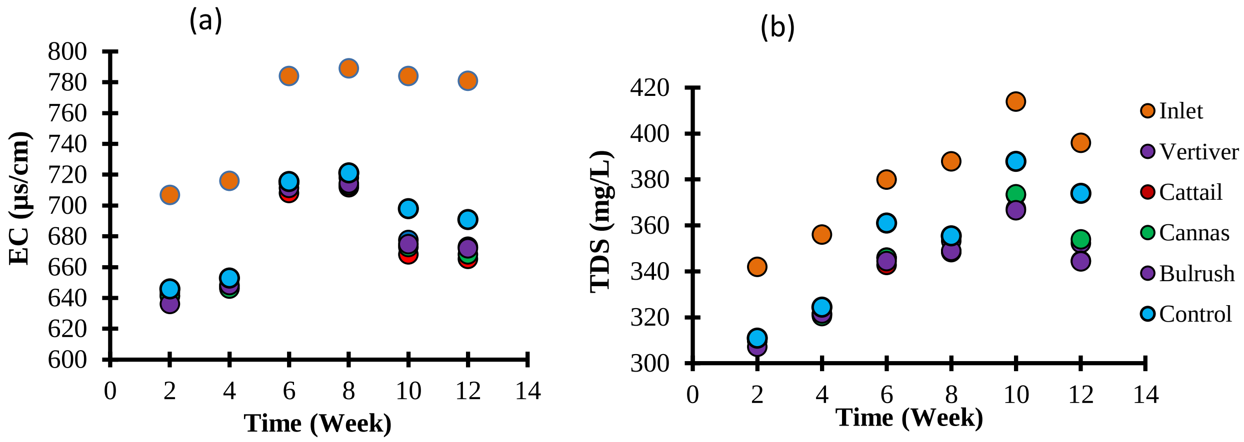

Figure 2: (a) Variation of EC (µS/cm); (b) Variation of TDS (mg/L) in the influents and effluents of each treatment.

All the treatments showed a considerable reduction of EC (11-14%) and TDS (10-13%) at 12 weeks after the planting except control experiment with bare soil. However, at the end, the reduction of both parameters were significantly higher in Cattail and Bulrush while the control showed lowest drop.

3.3 Phosphate Phosphorous

Phosphate phosphorous concentration of effluent samples was diminishing after the treatment as shown in Fig.3(a). The results revealed that there is a significant difference ($p < 0.05$) of reduction of phosphate - P values within five treatments. The highest removal efficiency of phosphate phosphorous over the monitoring period was 45%, 41%, 36% by Cattail, Bulrush and Cannas respectively. Plant uptake and retention in soil are major explanations for the phosphorous removal in wastewater in wetlands. Soil characteristics such as particle size distribution and structure significantly influencing on the magnitude of P removal (USEPA 1993).

Phosphorous is a major plant nutrient and plants tends to uptake it in the form of PO_4^{3-} ions. Phosphorous requirement is increasing with the growth of plants and the highest requirement can be experienced during the growing stage (Grant 2001). Therefore, phosphorous concentration of effluent is diminishing, and the rate of decrease is increasing with time until the plant become mature. The mechanisms for phosphorus removal in constructed wetlands are adsorption, complexation and precipitation, storage, plant uptake and biotic assimilation (UN-HABITAT 2008).

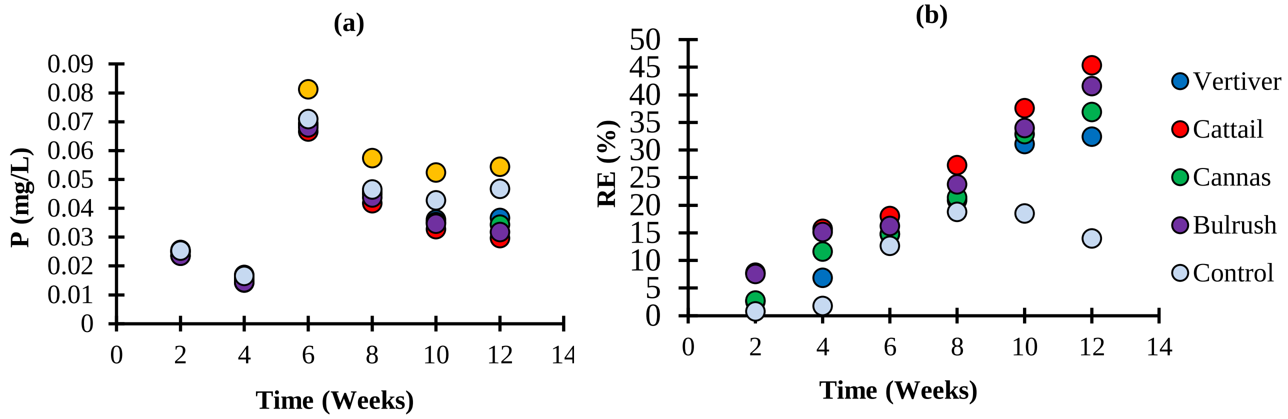

Figure 3: (a) Phosphate phosphorous concentrations (mg/L) of influents and effluents (mg/L); and; (b) Removal efficiencies (%) of Phosphate phosphorous with time

3.4 Nitrogen

The average measured inlet nitrate nitrogen (NO_3^--N) concentration was 4.1 mg/L. However, the influent NO_3^--N concentration varied in each week mainly due to the variation of quality of raw water in RO plant. The NO_3^--N concentration in the final effluent was lower than inlet concentration (Fig.4 a). According to the Fig. 4 (b) NO_3^--N removal efficiency is increased with time. Finally, 31%, 28%, 27% and 23% of removal efficiencies have been achieved by Cattail, Bulrush, Cannas and Vertiver respectively. De-nitrification, adsorption and plant uptake process support to the removal of nitrate-N from wastewater. The rate of reduction was found to increase as plants grow and develop in the wetland units. Cattail reported the highest nitrate-N removal which can be explained by its ability of enhancing nitrification of ammonia to nitrate, then promoting denitrification of nitrate to dinitrogen gas (Gebremariam and Beutel 2008).

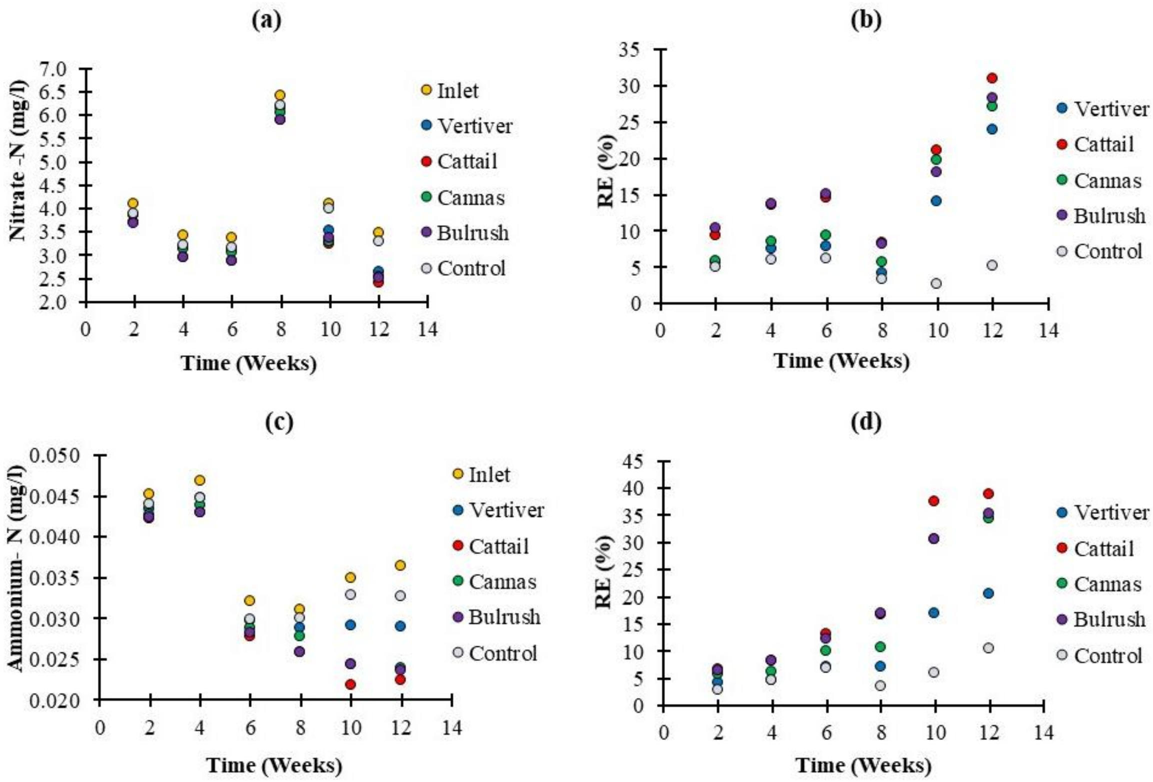

Figure 4: (a) Nitrate- N concentrations (mg/L) of influents and effluents; (b) Removal efficiencies (%) of nitrate- N with time; (c) Ammonium-N concentration (mg/L) in influents and effluents (mg/L); and (d) Removal efficiencies (%) of ammonium-N with time

The higher removal of ammonium nitrogen (NH_4^+-N) during the treatment process could be attributed to direct uptake of plant as a major nutrient and nitrification. According to the results, the efficiency of removing NH_4^+-N from the RO concentrate was increased with time. Finally, the highest removal efficiency of 39% observed for Cattail. Control (bare soil) showed the lowest removal efficiency for ammonium-N (Fig.4 d).

Volatilization, ammonification, nitrification/ de-nitrification, plant uptake and matrix adsorption are main nitrogen removal mechanisms in constructed wetlands (Lee 2009). Ammonium ions are oxidized into nitrate ions by nitrifying bacteria in aerobic zones. Nitrates are converted into di-nitrogen gas by denitrifying bacteria in anoxic and anaerobic zones (UN-HABITAT 2008).

3.5 Trace Element Content

According to the results, cadmium, lead and arsenic content of effluents samples were reduced than the influent samples after the treatment process as shown in Fig. 5. There were no any trace elements detected in the raw water samples obtained during 2nd, 4th and 10th weeks. However, heavy metal concentrations of the influent and effluent samples were within permissible range of irrigation water.

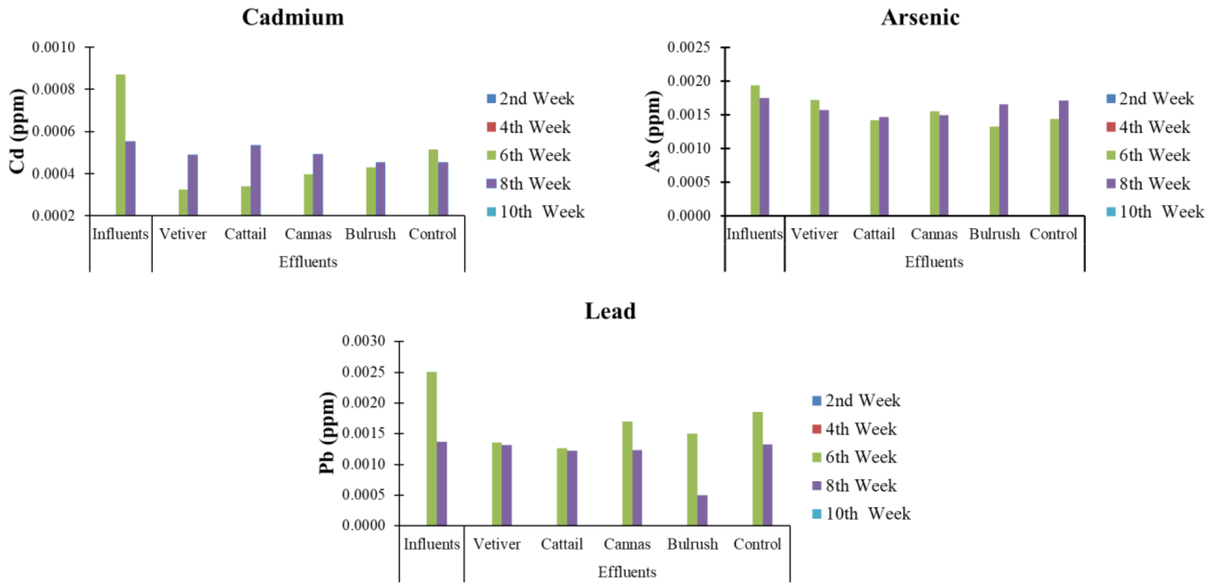

Figure 5. Heavy metals concentration

Heavy metals are usually removed from wastewater and mine drainage in constructed wetlands by the filtration and sedimentation of suspended particles, adsorption, uptake into the plant materials and precipitation by biogeochemical (microbial) processes (Stottemeister et al. 2003, Stottemeister et al. 2006)

3.6 Sodium Adsorption Ratio (SAR)

SAR was comparatively higher in inlets than the outlets of each treatment units (Table 3). SAR contents of all inlets and outlets were within the usual range (0-15) of irrigation water (Ayers and Wescot 1985).

Table 3: Average SAR values for influents and effluents

SAR Influent	SAR Effluents				
	Vetiver	cattail	Cannas	Bulrush	Control
7.19	6.75	6.21	6.42	6.28	6.82

3.7 Soil Chemical Characteristics

The measured pH, EC and Phosphate phosphorous concentration two months after the planting were higher than the initial soil measurements. However, the soil NO_3^- ion concentration was higher in initial measurement. This reduction was comparatively higher in wetlands units planted with Cattail, Bulrush, Vertiver and Cannas than the bare soil. NO_3^- N ions in soil is usually removed by volatilization, up taking by plants and microorganisms etc. Soil chemical characteristics are shown in Table 4.

Table 4: Soil chemical characteristics

Treatment	pH		EC (µS/cm)		Phosphate – P (mg/L)		Nitrate -N (mg/L)	
	Before Treatment	After experiment	Before Treatment	After experiment	Before Treatment	After experiment	Before Treatment	After experiment
Vetiver	8.02	8.06	62.60	132.90	20.25	26.66	39.96	3.72
Cattail	8.02	8.11	62.60	121.87	20.25	22.74	39.96	7.66
Cannas	8.02	8.23	62.60	141.13	20.25	33.65	39.96	4.51
Bulrush	8.02	8.16	62.60	135.67	20.25	23.85	39.96	6.97
Bare soil	8.02	8.21	62.60	126.40	20.25	25.87	39.96	30.98

4. Conclusion

Removal efficiencies (RE) of all measured water quality parameters were increasing with time proving that the four plant species that are used in this study have capability of removing pollutants from RO concentrate. Cattail plants showed the highest removal efficiency for PO_4^{3-}-P, NO_3^--N and NH_4^+-N by 45%, 30% and 39% respectively. Similarly, Cattail plants showed the highest EC reduction (15%) at 12 weeks. Highest TDS reduction (13%) was showed by Cattail and Bulrush plants. Sodium Adsorption Ratio of all treatment plants were under the low sodium (0-10) water quality class. Therefore, it can be concluded that the quality of RO concentrate can be improved using phytoremediation techniques.

However, further studies adjusting different Hydraulic retention time (HRT) to get the maximum performance in wetland units are vital to identify the most effective plant species for treating RO concentrate. This study can be further extended by mixing RO concentrate with kitchen wastewater to improve the microbial processes within the wetland system.

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