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ORIGINAL PAPER

Biochar-based constructed wetlands to treat reverse osmosis rejected concentrates in chronic kidney disease endemic areas in Sri Lanka

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Abstract The objectives were to investigate the potential remedial measures for reverse osmosis (RO) rejected water through constructed wetlands (CWs) with low-cost materials in the media established in chronic kidney disease of unknown etiology (CKDu) prevalent area in Sri Lanka. A pilot-scale surface and subsurface water CWs were established at the Medawachchiya community-based RO water supply unit. Locally available soil, calicut tile and biochar were used in proportions of 81, 16.5 and 2.5% (w/w), respectively, as filter materials in the subsurface. *Vetiver grass* and *Scirpus grossus* were selected for subsurface wetland while water lettuce and water hyacinth were chosen for free water surface CWs. Results showed that the CKDu sensitive parameters; total dissolved solids, hardness, total alkalinity and fluoride were reduced considerably (20–85%) and most met desirable levels of stipulated ambient standards. Biochar seemed to play a major role in removing fluoride from the system which may be due to the existing and adsorbed K^{+}, Ca^{+2}, Mg^{+2}, etc. on the biochar surface via chemisorption. The least reduction was observed for alkalinity. This study indicated potential purification of aforesaid ions in water which are considerably present in RO rejection. Therefore, the invented bio-geo constructed wetland can be considered as a sustainable, economical and effective option for reducing high concentrations of CKDu sensitive parameters in RO rejected water before discharging into the inland waters.

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Introduction

Chronic kidney disease of unknown etiology (CKDu) in Sri Lanka was identified in early 1990s and progressively increased to epidemic levels in rural farming communities, specifically in the North Central Province (NCP) of Sri Lanka (Dharma-wardana et al. 2015; Rango et al. 2015; Wimalawansa 2014). The exact prevalence and geographic scope of the problem are unclear, whereas prevalence estimates range from 2 to 15% (Chandrajith et al. 2011; Jayatilake et al. 2013). Estimates for affected population are 400,000 people with a death toll of around 20,000 people (Ranasinghe and Ranasinghe 2015). Though several hypotheses have been advanced concerning the causative factor for CKDu epidemic in Sri Lanka, drinking water is often considered to be a major source of nephrotoxic contaminants that cause CKDu, and the contaminants of concern often come from natural (e.g., local geological materials) sources. Those include fluoride, cadmium and hardness (Wasana et al. 2017). Increased ionicity of drinking water due to irrigation, recharge differences, redox processes in the soil, and features of 'tank' cascades and aquifers and the consequent chronic exposure to high ionicity in drinking water is proposed to debilitate the kidney via a Hofmeister-type (i.e., protein denaturing) mechanism (Dharma-wardana et al. 2015). It was also found that fluoride content of well water in all the areas undergo with CKDu exceeded the WHO recommended level of 1 mg/L. Water in NCP was alkaline which could facilitate mobilization of fluoride from minerals indicating a fluoride mediated mechanism for renal damage (Wanigasuriya et al. 2011). Hence, as an immediate response to the CKDu epidemiology many different water treatment options are delivered to the community as there is no coverage of pipe borne water for the NCP. Reverse osmosis (RO) plants are one of the frequently established systems (>180 for the moment) to provide safe drinking water to the general public.

Although there can be disadvantages of using RO permeate for drinking in the long run for the CKDu endemic areas, it can be easily eliminated either by selective membrane for essential ions or addition of minerals afterward. The RO reject (concentrate water rich in dissolved salts), wastewater from the RO treatment plant, needs to be treated before its discharge. However, in Sri Lanka, the RO concentrate is directly released to the environment for irrigation use or dumped on the soil. As the direct disposal of RO rejects may results adverse effect on the environment, there was a necessity for proper treatment before discharge. The production and disposal of rejected brine (RO concentrate) are an integral part of an overall RO process. Several disposal techniques of the RO rejected water are practiced worldwide. These include direct surface water discharge, discharge to a sewage treatment plant, deep well disposal, land application, evaporation ponds, rejected water (brine/concentrators) and mixing with the cooling water or sewage treatment effluents before surface discharge (Shreesadh et al. 2013). However, these are beyond Sri Lankan context due to the cost and skilled labor involvement. Similarly, land application/irrigation was deemed infeasible due to the discharge limitations in surface water bodies in Sri Lanka.

When water is passing through the semipermeable membrane, it removes most of the minerals and components from inlet water to about 90–95% with an average of 40–50% of inlet water. The high TDS, BOD, COD and other impurities in RO rejected depends upon the intake water quality and can make it prohibitive for discharge into the environment as it can contaminate water bodies directly and soils. However, neither the government nor regulatory authorities focused their attention on RO rejected water in Sri Lanka although Sri Lankan Standards for discharging wastewater is governed by the extraordinary gazette called National Environmental (Protection & Quality) Regulations of 2008. Anyway, if the RO rejected water continued to be dumped on the ground or discharged to irrigation canals, it will subsequently damage the inland waters, aquifers and soils in the long run. Hence, we hypothesized that the low-cost phytoremediation through constructed wetlands (CW) would be an option for the treatment of RO rejected water in the NCP.

Constructed wetlands or engineered wetlands (EWs) are man-made land plots same as natural wetlands which are used to purify soil and wastewaters depending upon the requirement. Wetlands are considered as proven natural treatment technology for the purification of wastewater and soil. Phytoremediation in EWs is a hopeful alternative to treat wastewaters and an increasingly recognized pathway to advance the treatment capacity of wetland systems (Zhang et al. 2010). Various contaminants can be minimized using water hyacinth, water lettuce and Vetiver grass such as total suspended solids (TSS), total dissolved solids (TDS), electrical conductivity, hardness, biochemical oxygen demand (BOD), chemical oxygen demand (COD), dissolved oxygen (DO), nitrogen, phosphorous, heavy metals and other contaminants (Herath and Vithanage 2015; Otte and Jacob 2006). It was found that 70% of TDS reduction can be done by water lettuce (Awuah et al. 2004) and up to 56% by Vetiver grass (Gupta et al. 2012). Also, water hyacinth was the most effective for the removal of cadmium, chloride, iron, copper, manganese, lead, fluoride, sulfate, nitrate, phosphate and potassium (Sanmuga Priya and Senthamil Selvan 2014; Jamuna and Noorjahan 2009; Rezania et al. 2015).

A constructed wetland system needs a media for the growth of macrophytes except for a free water surface (FWS) constructed wetland system. Hence, the growth media is an important factor when designing the wetland system for subsurface flow (SSF) (Rizwan and Athapattu 2014). The SSF wetland is typically constructed as a bed or channel containing appropriate media with coarse rock, gravel, sands and other soils. In SSF and FWS wetlands, where the plants are growing in soil under the water logged conditions, roots, rhizomes and stems physically penetrate the soil layer and provide effective fluid movement by increasing hydraulic conductivity. Hence, the growth media is a crucial factor to be considered. Biochar, which is produced by thermal decomposition of biomass under oxygen-limited conditions (pyrolysis), has been found to be efficient in removing different ions including heavy metals, nitrate, phosphate, pharmaceuticals, pesticides and other ions such as calcium and magnesium (Herath et al. 2015a, b). At the same time, clay tiles are being recognized as a promising material for fluoride removal from water (Vithanage et al. 2012; Yadav et al. 2006).

Hence, this research was carried out to investigate the remedial measures to rejected effluent of the RO plants in CKDu prevalence areas, using a pilot-scale constructed wetland incorporating biochar, calicut tiles and native soil as media together with plants as potential phytoremediators to treat RO rejected water and improve its quality to comply with the Central Environmental Authority (CEA) Standards in Sri Lanka.

Materials and methods

Since the Medawachchiya District Secretariat (DS) Division is one of the areas recorded with largest number of CKDu patients, it has the highest number of RO plants in NCP. Among all RO plants the selected plant for this study is with highest capacity and situated at Sangilikanadarawa Grama Niladhari Division (GND, the smallest administrative sector) in which is 20 cubic meters per day (20,000 L/day). The water source of the plant is a covered shallow well with a diameter of 9 m and the depth of 8 m. According to the historical data, the well provides water throughout the year even in the dry season, whereas many wells are seasonal. The current beneficiary of the RO plant is about 2300 personnel including the surrounded GNDs. About 5100 L/day is being provided only for drinking and cooking purposes.

Design of constructed wetlands

In order to purify the contaminants in RO rejected water at Sangilikanadarawa, water lettuce and water hyacinth were selected for FWS constructed wetland system while *Vetiver grass* and *Scirpus grossus* were selected for SSF constructed wetland system in field experiment. Free floating macrophyte-based free water surface (FWS) and horizontal flow subsurface flow (SSF) were selected for the treatment. The schematic diagram of the proposed system is shown in Fig. 1. The legend gives the details of the system. Size of each CW is 1 × 3.25 m. Two controls were selected to measure the efficiency of each type of constructed wetland and the bio- and geo-materials used in this field experiment.

Rejected water connection was extended up to the site and pipeline network of wetland system was laid. The G1000 PVC membrane was laid in the bottom of the wetlands for all types of wetlands. Then the selected three types of materials; calicut titles, native

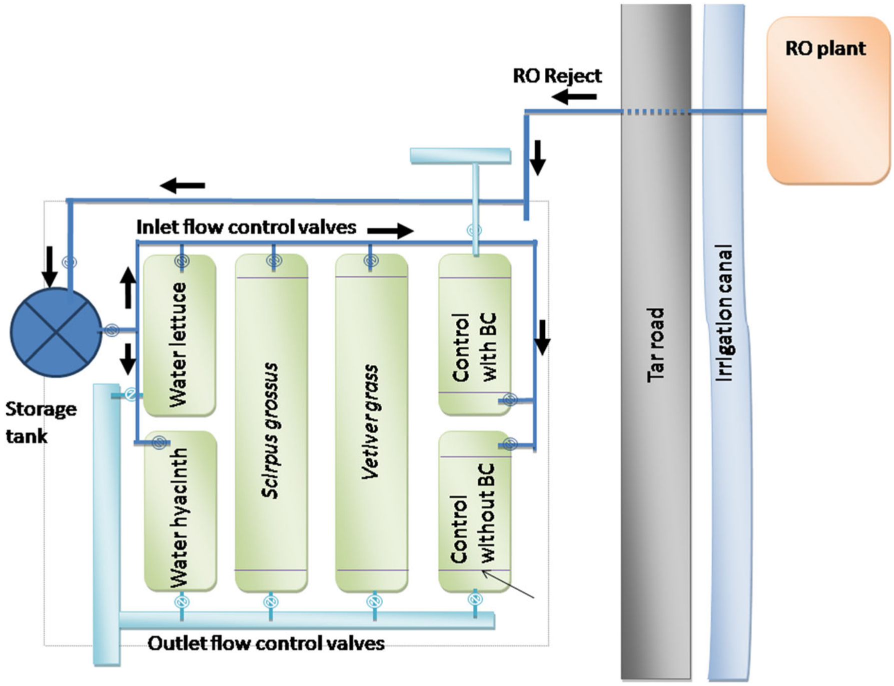

Fig. 1 Schematic diagram of the CW established at Sangilikanadarawa, Medawachchiya

soil and biochar were mixed to the given proportion and placed media for subsurface flow wetlands.

Operational conditions for field experiment

Water lettuce, water hyacinth, *Vetiver grass* and *Scirpus grossus* were obtained from the nearby sites and transplanted to FWS constructed wetland system and SSF constructed wetland system, respectively, for the field experiment. Two systems were left plant-free as controls. RO rejected water was continuously added at one end of the system and flowed out from the opposite end as shown in Fig. 1. The system was maintained for three months.

Sampling and analysis of bio- and geo-materials

The volume of wastewater inflow was measured by integration flow meter. The evaporation was measured using pan evaporator. The rain gauge was installed at the site and daily rainfall was measured if any rainfall occurred. The water quality was analyzed intermittently. Water samples were collected periodically from inlet and outlets of respective wetland systems and analyzed using methods given in Table 1. Physical observations of the systems were done by measuring the heights color of leaves and plant density periodically. Removal rate was calculated monthly as the difference between the rates of respective parameters in the RO rejected water entering and leaving the systems divided by area of the wetland. After one month, leaves and fresh roots were cut into 2-cm pieces and air-dried at 80 °C in the laboratory. The removal rate per biomass was calculated as the removal rate divided by the plant dry weight. The air-dried solid materials were blended, and samples were analyzed to obtain the rate of removal.

Sampling of analysis water quality parameters

Laboratory experiments for water quality parameters such as pH, TDS, conductivity, temperature, salinity BOD and COD were carried out using water samples taken from the stream. Water samples were collected for clean 500-ml bottles from the raw water, RO rejected and RO permeates of the selected RO plant at Sangilikanadarawa, Medawachchiya, and tested for phosphate, nitrate nitrogen, ammonia nitrogen, COD, BOD, pH, total dissolved solids (TDS), salinity and conductivity.

Table 1 Methods of analysis for water and analytical data

Constituents	Method	Permissible levels	Analytical data	Unit
Total dissolved solids	Conductivity meter	500	563	mg/L
Hardness	Titrimetric method	250	306	mg/L
Alkalinity	Titrimetric method	200	336	mg/L
Fluoride	Ion-selective electrode	1.0	0.97	mg/L
Phosphate	Ascorbic acid method	2.0	0.68	mg/L
Nitrate	Cd reduction method	10	0.88	mg/L
Ammonia N	Ion-selective electrode	0.06	0.02	mg/L
pH	pH electrode	6.5	8.5	
Iron	Atomic absorption	0.3	0.02	mg/L
Chromium	-do-	0.05	ND	mg/L
Lead	-do-	0.01	ND	mg/L
Cadmium	-do-	0.003	ND	mg/L
Arsenic	-do-	0.01	ND	mg/L
Mercury	-do-	0.001	ND	mg/L
Cyanide	CN analyzer	0.05	ND	mg/L

Results and discussion

Rejected effluent of RO plant

The rejected water (concentrate) was about 50% of raw water supplied to RO plant at Sangilikanadarawa and is directly discharged into irrigation canal. Some chemical parameters such as total dissolved solids (TDS), total alkalinity (as $CaCO_3$), total hardness (as $CaCO_3$) and fluoride (as F^-) of RO rejected water have exceeded (500, 200, 250 and 1.0 mg/L, respectively) the maximum required permissible level according to the drinking water standard of SLS 614:22013/(UDC663.6); however, no parameter has exceeded the parameters given in National Environmental Regulations (NER) [1:2008] (Table 1). Even though there is no issue at present compared to the NER standards, it may become high when the dose per day is considered which can cause an environmental issue in future, especially with hardness and fluoride which have been considered as a sensitive groundwater quality parameter in CKDu prevalence areas.

Hydraulic retention time

It was recorded that the hydraulic retention time (HRT) for water hyacinth varied between 3 and 9 days and varied with respect to the inlet water to the system which corroborates with the HRT of other studies (Vaidyanathan et al. 1985; Giraldo and Garzon 2002). The HRT for FWS was selected as 3 days while the SSF wetland unit was designed to have 4 days of retention time based on the calculations. The bed media for SSF were selected carefully and operated by controlling the inlet flow starting from designed continuous flow with a retention time of 4 days.

Bio-geo media for wetland beds

Most commonly used bed materials in SSF constructed wetlands were ranging from medium gravel to coarse rock for BOD, COD and nitrogen removal of wastewaters in the USA, and sand to gravel (Arceivala and Asolekar 2007). The locally available soil, calicut tiles and biochar were selected as the bed bio-geo media. Biochar was produced by pyrolyzing *Gliricidia* biomass at >700 °C in a closed reactor as a by-product of generating energy. Locally available soil was used with calicut tiles with the range of 18–25 mm particle size average value of 15–17.5% by mass. According to the literature, the selected biochar had a capacity of removing Cr, Ni and Mn and the plant growth was highly favorable with 2.5% percent by mass of the soil (Bandara et al. 2016; Herath et al. 2015a, b). Also the selected biochar had an increased total surface area of 714 m^2/g which led the capability of increasing adsorption along its lifetime. At the same time, the particular biochar was a by-product of the dendro power plant at Labunoruwa, Anuradhapura, and reuse of the particular material would help in the reduction of waste from the environment. However, since biochar is light, it needs a base to be mixed in order to make it as a supporting media for constructed wetlands.

Reduction of total dissolved solid (TDS)

The values of TDS reduced with respect to the TDS value of the inlet which is RO reject. During the initial stage (after 4 days), both wetland effluents indicated higher reduction percentage of TDS than later. After 20 days, both reached its minimum value but again started increasing with slight increment. After 30 days, it started to reduce again which may be due to the change in the RO rejected concentrate. The SSF control wetland in the wetland No-1 (control with biochar) indicated higher reduction than wetland No-2 (control without biochar). At the initial stage, the percentage difference of wetland No-1 and wetland No-2 is 18.4. Later, both the wetlands gave the lower performance than the initial, but still, the wetland with biochar showed the higher reduction percentage.

Testing of wetlands with *Vetiver grass* and *Scirpus grossus* was started after 16 days of planting and it is also represent a considerable reduction percentage than wetland No-5 and 6, but less amount with respect to the control with biochar. Then the reduction capacity started to increase and, however, then reduced after two weeks.

Total alkalinity (as $CaCO_3$)

The percentage reduction of alkalinity compared to RO rejected water is shown in Fig. 2b. At the initial stage, discharge effluent indicated high reduction in the total alkalinity than later on in the FWS wetland system. Wetland with water lettuce reached its minimum reduction value within 20 days where as wetland with water hyacinth reached its minimum in 25 days. Control wetland with biochar represented the highest reduction percentage of total alkalinity in the initial stage which is more than 65%, whereas control without biochar indicated >50%. Nevertheless, both reduction percentages are slightly decreased with the time period. The reduction percentages of *Vetiver Grass* and *Scirpus grossus* wetlands showed a slight increase >40% within the same time frame.

Total hardness (as $CaCO_3$)

According to Fig. 2c, water lettuce and water hyacinth applied wetland reached its minimum reduction value within 16–20 days. Control wetland with biochar represented the highest reduction percentage of hardness in the initial stage which is more than 45%. The reduction capacity of hardness in *Vetiver grass* and *Scirpus grossus* wetlands is considerably high compared to FWS wetlands.

Fluoride (as F^-)

The reduction percentage of fluoride with respect to the RO rejected water is graphically represented in Fig. 2d. At the initial stage, FWS wetland systems discharged effluent indicated no significant change with the later values. According to Fig. 2d, the wetland which contained water lettuce reached its minimum reduction value within 20 days. At SSF, control wetland with biochar and without biochar represented the highest reduction percentage of fluoride removal in initial stage which is more than 80%. Thereafter the reduction capacity decreased. However, the wetlands with *Vetiver grass* and *Scirpus grossus* had a higher initial reduction of fluoride. *Scirpus grossus* reached its highest value in 30 days, but the *Vetiver grass* exhibited a continuous increase in removal capacity of fluoride.

A large variation in raw water quality was observed during the period of study (Table 2). However, compared to FWS, the SSF has exhibited significant reduction in the considered parameters. Among 4 different plant types, the best types are *Vetiver grass* and *Scirpus grossus*. It is clear that the biochar's role in treating the RO rejected water by considering the two types of controls. Control with biochar has demonstrated better results in terms of parameters assessed than the control without biochar. Therefore, waste by-product biochar seemed to have strong influence on the treatment of RO rejected water with a beneficial of reducing pollutant on the other hand. Therefore, these SSF systems can be used as sustainable solutions to treat RO rejected water in NCP.

Wetland system performance

Figure 3 shows the wetland system performances with fluoride versus hardness. Accordingly, the best

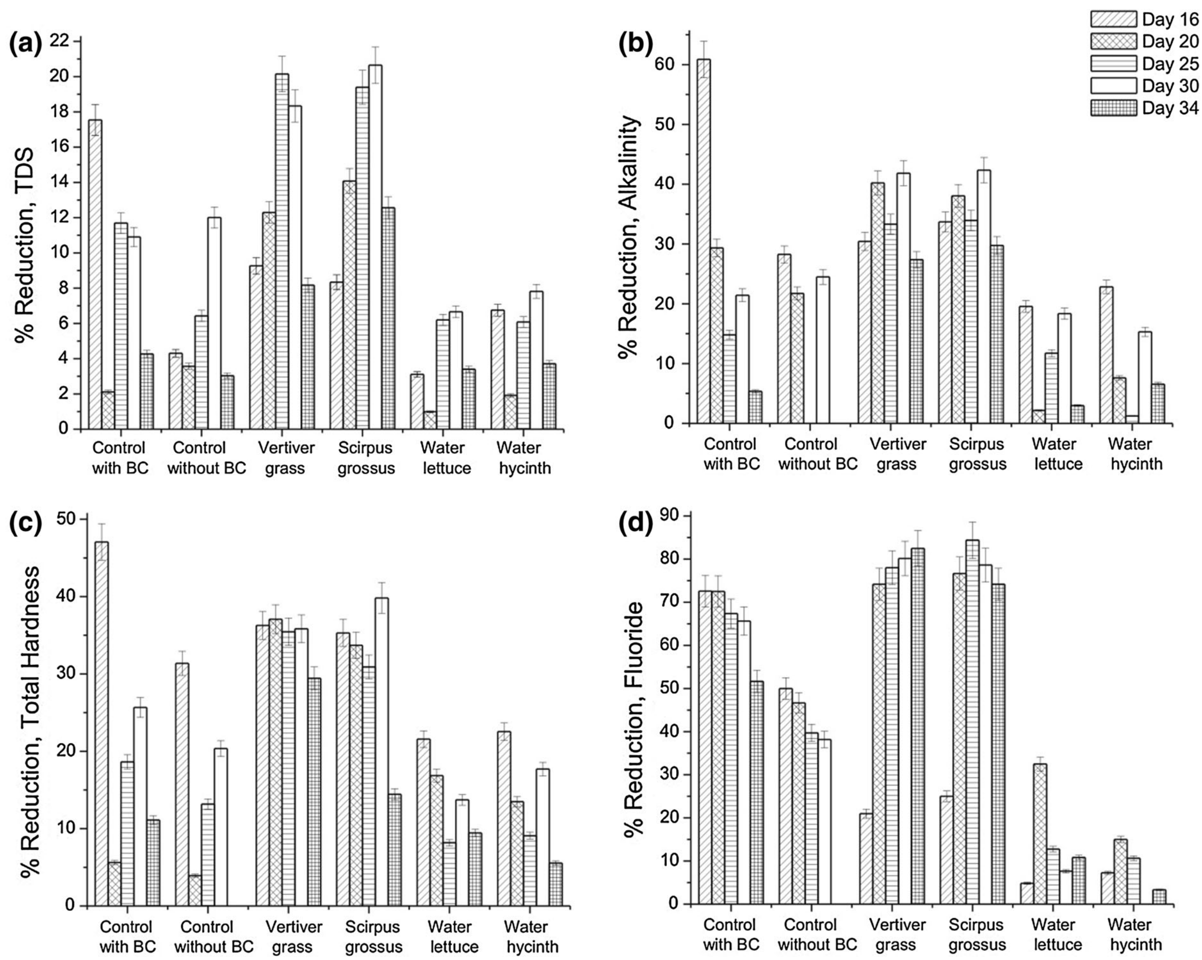

Fig. 2 Percentage reduction in TDS (**a**), total alkalinity (**b**), total hardness (**c**) and fluoride (**d**) in different constructed wetland systems in different days for the RO rejected water

performance is given for fluoride removal by the SSF wetlands with both *Vetiver grass* and *Scirpus grossus* with respect to the drinking water standard (SLS 614: 2013). The control set up with biochar also demonstrated excellent performance while the control without biochar is given fair enough performance compared to two other types of FWS wetland systems. Even though the hardness removal has not reached the level of drinking water standard of SLS 614: 2013, there is a considerable reduction with both SSF *Vetiver grass* and *Scirpus grossus* CWs and also by the SSF control with biochar.

Figure 4 shows the wetland system performances with alkalinity versus TDS. Neither SSF wetlands nor FWS wetlands could reduce the alkalinity up to the drinking water standard (SLS 614: 2013) as a whole, but still demonstrated a considerable reduction stand alone with respected RO rejected water quality. Similarly, the best performances for both alkalinity and TDS are exhibited by the two SSF CWs, *Vetiver grass* and *Scirpus grossus*.

Mechanisms of remediation

The efficiency of remediation achieved by constructed wetland systems may depend upon different factors including the type of media used (i.e., biochar, Calicut titles and native soil), selectivity of plants and availability of microorganisms. It can be suggested that the contaminant removal is manly governed by various biological and physicochemical factors including microbial activity, uptake by plant species,

Table 2 Maximum and minimum % reduction in different wetland types

Wetland type	TDS % reduction		Alkalinity % reduction		Hardness % reduction		Fluoride % reduction	
	Min	Max	Min	Max	Min	Max	Min	Max
Control with BC (wetland No-1)	2.1	17.5	5.4	60.9	5.6	47	51.6	72.6
Control No BC (wetland No-2)	3	12	0	28.3	0	31.3	0	50
Vetiver grass (wetland No-3)	8.2	20.2	27.4	41.8	29.4	37	20.9	82.5
Scirpus grossus (wetland No-4)	8.3	20.6	29.7	42.3	14.4	39.8	25	84.3
Water lettuce (wetland No-5)	1	6.6	2.1	19.6	8.2	22.5	4.8	32.5
Water hyacinth (wetland No-6)	1.2	22.8	1.3	22.8	5.5	21.5	0	15

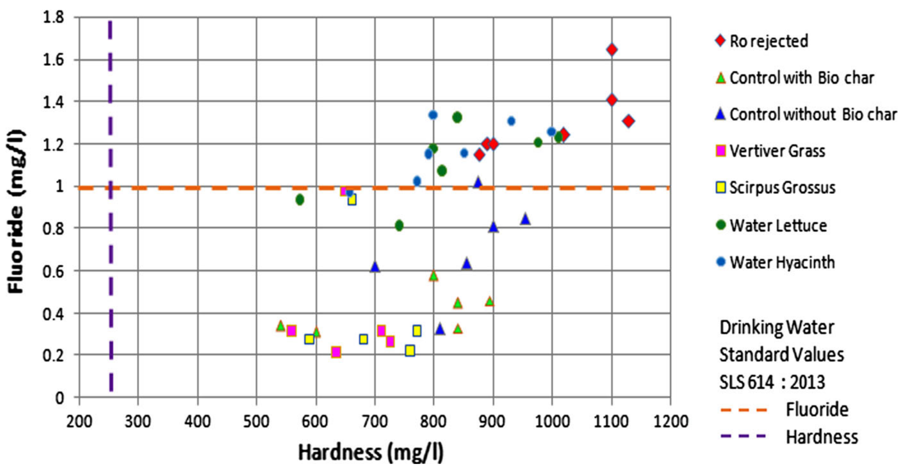

Fig. 3 Wetland system performances with fluoride versus hardness

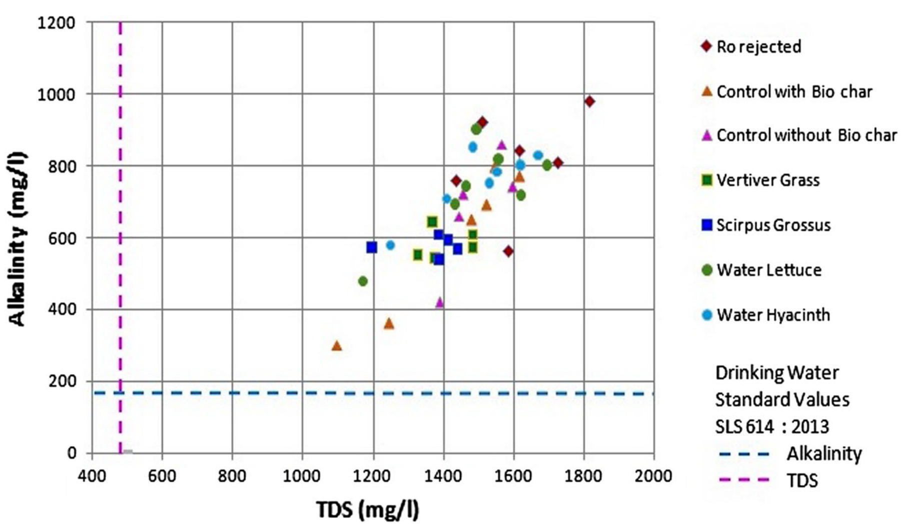

Fig. 4 Wetland system performance with alkalinity versus TDS

sedimentation, flocculation, precipitation, adsorption, complexation, oxidation and reduction and cation and anion exchange (Ladislas et al. 2015).

Particle size and pore size distribution in the medium in constructed wetland systems has gained a significant attention among the soil physical parameters (Bruch et al. 2014). Results revealed that these soil physical parameters had an influence on purification capacity. For example, soil disturbance involves compaction and the vertical integration of soil horizons and regolith, which leads to new soil textures and change in bulk density. Additionally, loss of macrostructure and addition of biochar may change the way water is held within the soil matrix.

Biochar contains a variety of oxygen-containing functional groups (i.e., –COH, –C=O and –COOH) (Herath et al. 2015a, b). Oxygenated functional groups contain long pairs of electrons and protonated functions may facilitate to adsorb hydrated fluoride ions (Mohan et al. 2014). In addition, with water swelling, H_3O^+ F^- hydrated ion pairs may diffuse into biochar surface (Mohan et al. 2012, 2014). This process might have involved in fluoride removal from the RO rejected water (Fig. 5). Another possible way of fluoride adsorption involves the role of ash that is available in the biochar. It is well known that metal-fluoride bonds are strong. Therefore, it can be suggested that Ca^{2+} and Mg^{2+} salts in the biochar may form insoluble fluorides via chemisorption process (Halder et al. 2016; Mohan et al. 2012, 2014; Oh et al. 2012). Further, Al, Ba, Mn and Fe fluorides have slight water solubilities and most of the biochars contain those elements. Hence, there is a possibility of producing biochar from the plants itself grown in the constructed wetland. Furthermore, biochar provide habitats for microorganisms that are responsible for decreasing total dissolved solids, alkalinity and hardness of the RO rejected water.

Moreover, wetland plant species are capable of growing in environments where their root system is submerged. Additional characteristics including ease of rooting, fast establishment, quick growth, extensive rates of photosynthesis and elevated usage of water make them successful in constructed wetland systems (Williams et al. 2010). As a result, it can be suggested that the selected wetland plant species may uptake high concentration of plant macro- and micronutrients including Ca, Mg in order to gain an excessive growth rate. In addition, these plants transport air toward the root system by diffusive and/or convective mechanisms via specialized gas channel tissues called aerenchyma (Williams et al. 2010). At the same time, preparation of biochar by the plants grown in the CWs after harvesting might be another possibility since the retention of metals such as Ca and Mg in plants may increase the fluoride immobilization more and more inducing water purification (Zeng et al. 2013).

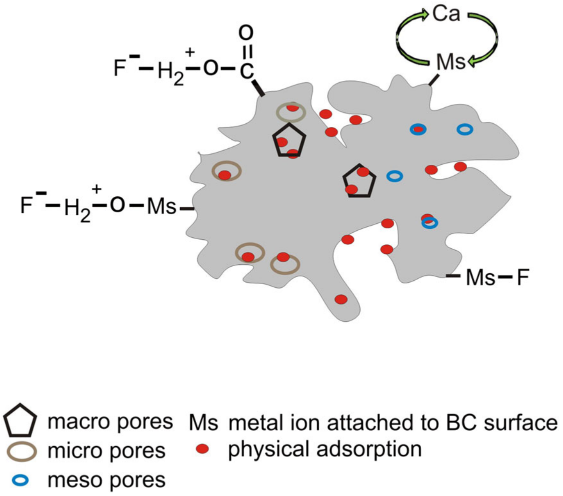

Fig. 5 Schematic representation of removal of different ions by biochar in the media

Conclusions

Four different types of constructed wetlands; two free water surface (FWS) and two subsurface flow (SSF) bio-geo CWs were established and operated for 30 days to understand its influence to treat the RO rejected concentrate. The results showed that total dissolved solids, hardness, total alkalinity and fluoride were reduced considerably with the SSF constructed wetlands than the FWS constructed wetlands. About 20–85% removal was observed from the pilot-scale constructed wetland units incorporated with biochar. The efficient role of biochar was confirmed from the two control units indicating higher removal of ions in biochar-embedded control unit including Ca and F via chemisorption mechanism. Despite the influent water quality variations, the decline pattern of removal efficiency of constructed wetland systems over time could be attributed to filtration-bed clogging. For instance, sufficient mechanical pretreatment is required to remove excessive amount of suspended solids in subsurface flow system, since they may cause filtration-bed clogging and consequent surface flow. The cost incurred to construct this system was approximately 400USD with locally available materials which is minimal. Hence, the system seemed to be sustainable and cost-effective to a community participation project to achieve and promising in reducing CKDu sensitive water quality parameters.

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