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Role of three different plants on simultaneous salt and nutrient reduction from saline synthetic wastewater in lab-scale constructed wetlands

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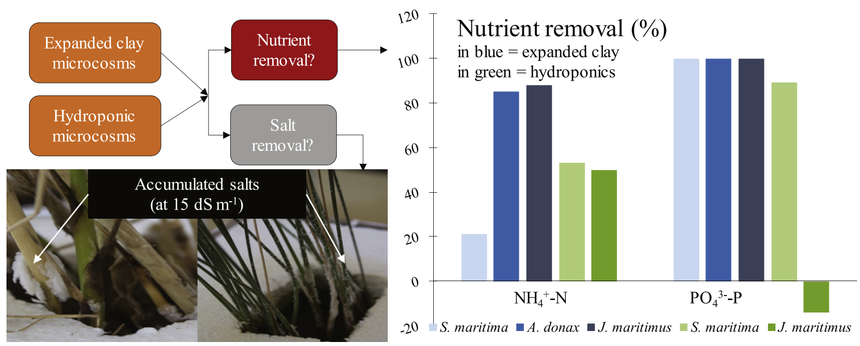
HIGHLIGHTS

- Simultaneous removal of salt and nutrients was tested under hypersaline conditions.
- Planted microcosms have higher ammonia and phosphate removal than unplanted ones.
- The tested plants have limited salt removal capacity for reasonable HRT values.
- Saline wastewater treatment in constructed wetlands is feasible with tested plants.

GRAPHICAL ABSTRACT



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ABSTRACT

Constructed Wetlands (CWs) can be a valuable technology to treat high salinity wastewaters but it is not known their potential for removal of both nutrients and salt, and the type of plants to use. This study evaluated the effect of three plants on salt reduction and simultaneous nutrient removal in CWs microcosms with expanded clay and in hydroponic conditions. Initial values of the synthetic wastewater tested were EC = 15 dS m^{-1}, SAR = 151; NH_4^+-N = 24 mg L^{-1}; PO_4^{3-}-P = 30 mg L^{-1} and NO_3^--N = 34 mg L^{-1}.
With expanded clay CW removal efficiency for NH_4^+-N was 21, 88 and 85%, while for NO_3^--N, it was 4, 56 and 68% for *Spartina maritima*, *Juncus maritimus* and *Arundo donax*, respectively. PO_4^{3-}-P was adsorbed completely in the expanded clay. However, in hydroponic system, removal efficiencies for NH_4^+-N were 53 and 50%, while PO_4^{3-}-P removal was 89 and −14% for *Spartina maritima* and *Juncus maritimus*, respectively. Nutrient removal in planted microcosms was statistically higher than unplanted controls for NH_4^+-N and PO_4^{3-}-P.
However, salt removal was apparent in the hydroponic system only after 23 days of HRT, despite clear salt excretion visible in both *Spartina maritima* and *Juncus maritimus*.
This study demonstrates the potential of two halophytic plants for saline wastewater treatment. However, salt removal in such a scenario could not be well documented and might prove to be impractical in future work.

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1. Introduction

Several industrial processes produce high strength wastewater with variable salinity and nutrient loads. These industries include conventional and unconventional oil and gas exploitation (Lutz et al., 2013), aquaculture (Jesus et al., 2013), tannery (Calheiros et al., 2012), winery production (Ioannou et al., 2015), and agricultural drainage waters (Qadir et al., 2003), among others.

The treatment of these wastewaters is necessary to reduce pollution and eutrophication risk in the discharge area due to organic matter and excess nutrients. However, the presence of dissolved salts can severely hinder the ability to reduce contaminants by conventional wastewater treatment plants due to microbial inhibition (Khengaoui et al., 2015). Furthermore, these treatment plants have limited to no capacity to remove the dissolved salts, which precludes discharge into sensitive surface waters without significant negative impacts on these ecosystems.

Additionally, considering existing and future freshwater scarcity, significant efforts have to be made for alternative water resources, particularly for agricultural irrigation in semi-arid regions. Reuse of treated wastewater is a potential solution but its use may be limited by salinity. Dissolved salts present in irrigation water can directly affect plant growth, accumulate in the soil and lead to soil structure degradation and water logging (US Salinity Laboratory, 1954; Ahmad et al., 2013; Wang et al., 2014). Several guidelines have been proposed for a threshold value of salinity that could be applied to soil that fall within a broad range between 0.5 dS m^{-1} to up to 2 dS m^{-1} (US Salinity Laboratory, 1954) depending on the plant culture in question, as well as the accompanying sodicity hazard (measured by the sodium adsorption ratio (SAR) or residual sodium carbonate (RSC). Nevertheless, moderately saline wastewaters can have variable EC values ranging from 1.27–8.12 dS m^{-1} from hydroponic systems (Chondraki et al., 2012) to 11–14 dS m^{-1} from cheese manufacturing (Prazeres et al., 2016). A treatment system that could reduce excess nutrients and salt simultaneously would be ideal, enabling either discharge to surface waters or reuse for irrigation or other purposes.

One type of wastewater treatment system that could potentially be able to perform such simultaneous treatment is constructed wetlands (CWs) (Shelef et al., 2012). CWs using halotolerant or halophytic plants are capable of reducing nutrient and organic loads in these high salinity wastewaters (Buhmann and Papenbrock, 2012; Jesus et al., 2013). Although, it is still unclear whether or not salt absorption by the plants is an effective method for salt removal within a CW (Shelef et al., 2012), since most studies using saline wastewater and CW treatment focus mostly or exclusively on plant salt tolerance rather than on salt removal.

However, the possibility of phytoremediation for salt removal in soils is already well established (Qadir and Oster, 2002; Qadir et al., 2006), although the removal mechanisms are still under debate. Therefore, this technology could be adapted to constructed wetlands if salt uptake is an important mechanism (Rabhi et al., 2010).

Plants have different salt tolerance mechanisms which might impact treatment efficiency (Yensen and Biel, 2006). This includes exclusion (plants that prevent salt from entering their tissues), accumulation (plants adapted to accumulation of salts within their tissues through various means) and excretion (plants with salt glands or trichomes capable of excreting excess salts). Although, plants with demonstrably high salt tolerance and biomass productivity might also be used for biosaline agriculture for forage or as a biofuel feedstock (Abideen et al., 2011). This is true even for those species exhibiting low salt removal.

Still another aspect not properly explored in the literature is the impact of salinity, or specific salts, on nutrient adsorption by CWs substrate. In particular, adsorption of phosphate to the substrate is considered the main removal mechanism of this nutrient in a CW, and several articles attempt to find the best performing substrate (Vohla et al., 2011). Nevertheless, increases in salinity might lead to desorption of nitrogen ions and improved phosphate removal by precipitation (Jun et al., 2013), or alternatively, decrease phosphate adsorption by competition for adsorption sites (Zhang and Huang, 2011).

Studies testing constructed wetland performance under high salinity conditions are not abundant in the literature and more so regarding the potential for salt removal. Furthermore, in the works investigating the high salinity CW issue, like the ones of Shelef et al. (2012) and Gao et al. (2015) for example, the objectives are focused either on salt or on nutrient reduction, but not on both aspects. Also, the adequate plant type and role under such conditions is still under debate. Therefore, it is not yet certain the feasibility of salt removal in CW. Additionally, studies are needed to analyze whether or not nutrient and salt can be removed simultaneously, and what is the role played by substrate and/or plants in such a scenario.

The objective of this study is to evaluate the impact of three different plants on the simultaneous removal or reduction of salt (EC, Na^+, Cl^-, SAR) and nutrients (NO_3^--N, NH_4^+-N and PO_4^{3-}-P) using CW microcosms containing expanded clay, and also under hydroponic conditions. The plants were chosen based on known or expected different responses to salinity. The first one is *Arundo donax*, which is a halotolerant, high biomass productive plant that is very easy to propagate (Ceotto and Di Candilo, 2010). The second plant is *Spartina maritima*, which is an obligatory halophyte and tolerant to flooding and submersion (Duarte et al., 2013). It also has low biomass productivity and is capable of salt excretion via salt glands. Finally, the last plant is *Juncus maritimus*, which is tolerant to flooding (Álvarez Rogel et al., 2001) and also a halophyte with intermediate biomass productivity.

These plants have rarely (*Arundo donax*) or never (*Spartina maritima* and *Juncus maritimus*) been tested under constructed wetlands scenarios, despite their potential applications in other phytotechnologies such as soil remediation (Conesa et al., 2011; Mesa et al., 2015).

For a more complete picture of nutrient removal in constructed wetlands treating highly saline wastewater, tests with expanded clay as a substrate and tests in hydroponics were performed under planted and unplanted conditions. This was done to pinpoint the direct plant contribution on the removal of nutrients and salts.

2. Materials and methods

2.1. Synthetic saline wastewater preparation

A high salinity synthetic wastewater was prepared with NaCl (9.6 g L^{-1}) at 15 dS m^{-1} (resulting in Na^+ concentration of 3806 mg L^{-1} and Cl^- of 5867 mg L^{-1}). Final nutrient concentrations were 30 mg L^{-1} of PO_4^{3-}-P (prepared from analytical grade, $NaH_2PO_4 \cdot H_2O$); 24 mg L^{-1} NH_4^+-N (prepared from analytical grade NH_4Cl) and/or (depending on the experiment), 34 mg L^{-1} of NO_3^--N (prepared from analytical grade $Ca(NO_3)_2$ and resulted in 2.43 meq L^{-1} of Ca^{2+} in the final solution). This solution had a resulting SAR of 151 and pH close to 7. Micronutrients were also added at 50% of Hoagland solution (Hoagland and Arnon, 1950) strength, resulting in final concentrations of 0.25 mg L^{-1} Bo, 0.25 mg L^{-} Mn; 0.025 mg L^{-1} Zn; 0.01 mg L^{-1} Cu; 0.005 mg L^{-1} Mo and 0.25 mg L^{-1} Fe. As no organic carbon source was added, BOD_5 level was assumed to be close to zero as previously tested in other studies with hydroponic wastewaters (Gagnon et al., 2010).

The generic saline wastewater formulation used was chosen so that the results obtained could be of value to a multitude of applications in hypersaline wastewater treatment. Selected nutrient concentrations match the range of several different wastewater types, but more closely resemble values found in hydroponic wastewaters (Park et al., 2008; Gagnon et al., 2010; Park et al., 2015), while also having low or zero BOD_5.

2.2. Greenhouse microclimate characterization

The location chosen for the tests was the window gallery of the building of the Department of Biology, University of Porto, with South/