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Green technologies for cyanobacteria and natural organic matter water treatment using natural based products

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ABSTRACT

Green technologies are environmentally friendly operations that limit the negative impacts of traditional industrial activities and can contribute to solving the challenge of sustainable water management. This work aims to develop a green water treatment sequence using a natural coagulant (*Moringa oleifera*) and a natural activated carbon (vegetable coconut palm) to remove *Microcystis aeruginosa* and natural organic matter in an integrated process. The proposed sequence, coagulation/flocculation/dissolved air flotation and activated carbon, is an eco-friendly and innovative option for water treatment managers, since it includes environmentally friendly products for the contaminant removal from water, replacing the traditional inorganic/synthetic coagulants and adsorbents. Results demonstrated that waters with concentrations as high as 150–200 μg/L of chlorophyll *a* and natural organic matter between 5 and 16 mg C/L presented removals higher than 80% for turbidity and chlorophyll *a*, 70–80% for dissolved organic matter and 80–90% for UV_{254nm}. *Moringa oleifera* natural coagulant removed almost 80% of *Microcystis aeruginosa* cells in coagulation/flocculation/dissolved air flotation processes. The coconut palm activated carbon showed a high capacity to adsorb DOC from water with a maximum adsorbed capacity of 51.8 mg/g at equilibrium. The values obtained in treated water for the parameters measured are very encouraging, so this innovative integrated sequence may be used for improving water quality, benefiting human health and wellbeing.

1. Introduction

Sustainable water management has received great attention over recent years because of its substantial benefits to the environment, society and economy. According with Ngo et al. (2016), the challenge of sustainable water management is to develop environmentally friendly, economically viable, and energy-efficient processes for treating and preserving the water resources. Thus, successful approaches will provide high removal efficiencies of pollutants and nutrient recovery while also reducing the carbon footprint, minimizing waste, and protecting human health and environment (Ngo et al., 2016).

In this line, the search for sustainable and eco-friendly water treatment processes, through the use of natural coagulants and natural adsorbents have increased during last years. Inorganic and synthetic coagulants, usually used in water treatment processes, are efficient and cost-effective, but need pH and alkalinity adjustments, generate high volumes of sludge, and their residuals in treated water (like aluminium) are linked with neurodegenerative diseases such as Alzheimer, and neurotoxic and carcinogenic effects (Rondeu et al., 2000). In addition, aluminium is not biodegradable and can cause environmental problems during the treatment and disposal of the sludge (Keeley et al., 2014; Xu et al., 2009). On the contrary, natural coagulants like plant extracts are available in abundance, safe to human health, and in general toxic free. They are also non-corrosive which eliminates the concerns of pipe-erosions and their application decrease up to five times the production of sludge (which have a higher nutritional value), reducing the operation and handling costs of the potable water production and improving the sustainability of water purification process (Abidin et al., 2011; Choy et al., 2014). Furthermore, the raw plants

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extracts used in natural coagulants are often available locally and their application provide an extra cost savings, and hence a low cost and more sustainable option (Asrafuzzaman et al., 2011; Natumanya and Okot-Okumu, 2015). Natural materials like *Moringa oleifera* (MO) seeds have been applied in water clarification especially for high turbid waters (>100 NTU) (Ghebremichael et al., 2005; Madrona et al., 2011; Ndabigengesere and Narasiah, 1998; Nkurunziza et al., 2009) and cyanobacterial removal (Lürling and Beekman, 2010; Moretti et al., 2016; Oladoja and Pan, 2015). Other natural materials used in water treatment with success were *Plantago ovata* seeds extracts to remove 95.6% of the initial water turbidity (50–300 NTU) (Ramavandi, 2014); and seeds of Horse chestnut and acorns (*Fagaceae* family) to removed 70–80% of the initial turbidity (Šciban et al., 2009). Muthuraman and Sasikala (2014) used *Strychnos potatorum* and *Phaseolus vulgaris* with similar results for high turbid waters. However, the use of MO seeds and other natural materials as coagulant contributes to the increase of organic matter in treated water (Ndabigengesere and Narasiah, 1998; Okuda et al., 2001).

Vegetable coconut palm has been used in water treatment to remove metals (Okafor et al., 2012; Shen et al., 2010; Sousa et al., 2010), iron (Beenakumari, 2009), phosphate (Anirudhan et al., 2009), sulfate (Namasivayam and Sangeetha, 2008) and organic matter in wastewater effluent from a brewery industry (Olafadehan et al., 2012). All these studies demonstrated that coconut shell can remove the target contaminants and can be used in wastewater treatment. Coconut shells activated carbon was also applied in drinking water treatment to remove fluoride (Said and Machunda, 2014). Results showed 68.2% and 65.9% removal efficiency for field and synthetic waters respectively, when particle size less than 150 μm of coconut shells activated carbon were used. Other natural adsorbents have been used in water treatment for humic acid removal, like rice husk pre-treated by HCl and thermally treated at 700 °C and chemically modified by aminopropylation (Imyima and Prapalimrungsi, 2010), and rice husk and sugarcane bagasse impregnated with $ZnCl_2$ and carbonized at 700 °C (Kalderis et al., 2008). Pine wood and pine bark chars (Mohan et al., 2011) and *Acacia etbaica* (Gebrekidan et al., 2015) were tested in the removal of fluoride and organochlorine pesticides from water, respectively. Bhatnagara and Sillanpaa (2010) presented a review on the use of agro-industrial and municipal waste materials as potential adsorbents for water treatment.

Cyanobacteria are found in freshwater phytoplankton in surface waters and have already posed a serious worldwide environmental issue due to their ability to release toxic metabolites, which have serious health implications (Sivonen and Jones, 1999; Falconer and Humpage, 2005). Natural organic matter (NOM) generally is present in surface freshwaters as a result from the interactions between the hydrologic cycle and the biosphere and geosphere (Mika Sillanpää, 2015). NOM affects water quality, like taste, odor, and color and comprises drinking water production (AWWA, 2000; EPA, 2011).

This work aims to study the use of two natural products (*Moringa oleifera* and vegetable coconut palm) to remove cyanobacteria and natural organic matter (NOM) from waters in an integrated treatment process - coagulation/flocculation/dissolved air flotation (C/F/DAF) and adsorption to natural activated carbon. *Moringa oleifera* is tested as natural coagulant and, since it increases the organic content of the water, coconut palm is used as adsorbent. In Camacho et al. (2017) it was demonstrated that *Moringa oleifera* can be used as a natural coagulant in coagulation/flocculation/sedimentation water treatment process, but it increased the organic matter content of the treated water. The present work proposes to solve this question by integrating an adsorption step and increasing the removal of cyanobacterial cells. Thus, the novelty of this work is the integration of the two treatment processes (C/F/DAF + adsorption on activated carbon), both using natural products. *Moringa oleifera* and vegetable coconut palm activated carbon were selected due to the availability and low-cost. The proposed sequence for water treatment is an eco-friendly option for water treatment managers.

2. Materials and methods

2.1. M. oleifera seeds and coconut palm adsorbent preparation

Moringa oleifera (MO) seeds used in this study were obtained in Aracaju city, Sergipe State, Brazil. Dry seeds were stored at room temperature until use. Before each experiment, the husk covering the MO seeds was manually peeled, and good quality seeds were selected and grounded in a regular blender during 3 min. After this process, MO active coagulant compound was extracted by mixing 5 g of dried MO powder with 500 mL of NaCl (1 M) for 30 min using a magnetic stirrer. The resulting solution was filtered using a 0.5 μm pore fiberglass membrane. More details about the preparation of this coagulant can be found in Camacho et al. (2017).

Vegetable coconut palm activated carbon was provided by Bahiacarbon Agroindustrial, Brazil. The particle average size of the coconut palm activated carbon varied between 0.25 and 0.30 mm, measured using a Mastersizer Micro (Malvern Instruments). The characteristics of this carbon were already presented in Silva et al. (2012). Briefly, surface area is 715.5 m^2/g, micropore area 677.8 m^2/g, total pore volume 0.386 cm^3/g and average pore diameter 20 Å (Silva et al., 2012). The coconut palm activated carbon was grounded to particles sizes below 100 μm (powder activated carbon, PAC).

2.2. Model water samples

Several model waters with different NOM and cyanobacteria concentrations were used to simulate surface waters. To prepare these waters, tap water was mixed with 3.5 ± 0.5 g of organic natural soil during 24 h, after which was filtered to remove the suspended solids. Based on turbidity, this solution was used to prepare the model waters where a stabilised concentration of *M. aeruginosa* cells was added. *Microcystis aeruginosa* cultures (Pasteur Culture Collection, PCC 7820) used in the experiments were grown in laboratory in BG11 medium, at controlled temperature of 24 °C with a 12 h light-12 h dark photo period. This culture produces toxins, namely microcystins-LR. *Microcystis aeruginosa* was analysed by chlorophyll *a* determination. After the preparation of the model water, all the parameters where measured and the experiments started.

2.3. Experimental design

This study was carried out in two distinct phases. In the first phase, adsorption experiments were made and in the second phase the integrated water treatment sequence C/F/DAF + PAC was tested.

2.3.1. Adsorption experiments

A batch system was used to study the adsorption kinetics and equilibrium of dissolved organic carbon (DOC) in the aqueous samples onto the coconut palm PAC. For the batch experiments different concentrations (0–250 mg/L) of coconut palm PAC were placed in glass-stopped flasks along with 100 mL of the model waters and 50 mg/L of MO prepared as previously described. In these experiments, two model waters with different DOC and turbidities, namely low turbidity (Lt) and medium turbidity (Mt), respectively ~10 NTU and ~60 NTU, were used (Table 1). The flasks