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Coupling Constructed Wetland and Electrochemical Reactor to Remediate Contaminated Water by Heavy metal

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ABSTRACT

Clean wetlands-based and electrochemical technologies have become viable alternatives to the remediation of potentially toxic metal (MPT) contaminated environments. We developed a wetland pilot system (CW) for treating contaminated water. Therefore, CW has as plant the *Alocasia macrorhiza* as well as an electrochemical reactor (ER) with electrodes of Fe and Al. The ability to remediate water by CW was evaluated with the contaminated water from Igarapé do Quarenta. This rive is known to be extremely polluted by MPT. We carry out three batches of experiments: i) 120 h of contaminated water permanence in the CW, and ii) 1.5 h of electrochemical process at a current of 2 A in the RE. We determined the MPT Cu, Zn, Cr, and Pb concentration in water samples at interval of 24 h from the CW by flame Atomic Absorption Spectroscopy (FAAS) in the three batches of experiments carried out. On the other hand, we also determined the concentration of Cu, Zn, Cr, and Pb by FAAS in the biomass of *Alocasia macrorhiza*. The results showed that CW was able to remove Cr to the level recommended by CONAMA. In the case of Cu and Pb, the reduction caused by CW was very close to the levels recommended by CONAMA, except Zn. *Alocasia Macrorhiza* presented the following sequence for removing MPT: Zn > Pb > Cu > Cr. It shows *Alocasia Macrorhiza* as to be promising for the phytoremediation for MPT. The RE batch showed the following sequence of MPT remediation: Zn, Cr > Cu > Pb. In addition, RE was able to reduce Zn and Cu concentrations to the levels recommended by CONAMA. Therefore, the CW remediates contaminated water, however, it must be improved to remove Pb to the level recommended by CONAMA.

Keywords: Water Quality, Alocasia macrorrhiza, Fe/Al electrodes

Resumo

Tecnologias eletroquímicas e baseadas em áreas úmidas limpas se tornaram alternativas viáveis para a remediação de ambientes contaminados por metais potencialmente tóxicos (MPT). Desenvolvemos um sistema piloto de wetland (CW) para o tratamento de água contaminada. Portanto, a CW tem como planta a macrorriza de Alocasia e um reator eletroquímico (ER) com eletrodos de Fe e Al. Esta rive é conhecida por ser extremamente poluída pelo MPT. Realizamos três lotes de experimentos: i) 120 h de permanência de água contaminada no CW, e ii) 1,5 h de processo eletroquímico a uma corrente de 2 A no RE. Determinamos a concentração de MPT Cu, Zn, Cr e Pb em amostras de água a um intervalo de 24 h do CW por Espectroscopia de Absorção Atômica de Chama (FAAS) nos três lotes de

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experimentos realizados. Por outro lado, também determinamos a concentração de Cu, Zn, Cr e Pb por FAAS na biomassa de Alocasia macrorhiza. Os resultados mostraram que o CW foi capaz de remover o Cr até o nível recomendado pelo CONAMA. No caso do Cu e Pb, a redução ocasionada pelo CW foi muito próxima aos níveis recomendados pelo CONAMA, exceto Zn. Alocasia Macrorhiza apresentou a seguinte seqüência para remoção do TMF: Zn> Pb> Cu> Cr. Mostra Alocasia Macrorhiza como promissora para a fitorremediação para MPT. O lote RE apresentou a seguinte sequência de remediação do MPT: Zn, Cr> Cu> Pb. Além disso, RE foi capaz de reduzir as concentrações de Zn e Cu aos níveis recomendados pelo CONAMA. Portanto, o CW remedia a água contaminada, porém, deve ser melhorado para retirar o Pb ao nível recomendado pelo CONAMA.

Palavras-chave: Qualidade da Água, Alocasia macrorrhiza, eletrodos Fe / Al

1. Introduction

The accelerating industrialization in developing countries with an enormous and increasing demand for natural resources has caused high emissions of the heavy metal into the biosphere. For example, Manaus city has increased number of inhabitants from 1940 (284,118 inhabitants) to 2014 (2,020,301) with starting an industrial plant in 1969 [(Santana and Barroncas 2007),(IBGE 2014)]. As a result, the higher anthropogenic emissions have considerably increased into the Educandos hydrographic basin, which is an important hydrological system from Manaus Industrial District. Educandos basin is a central area of Manaus city, harboring 250,000 inhabitants, and an industrial district formed by over 400 high throughput state-of-art companies. The electric-electronic sector represents 55% of the production of the Manaus Industrial District. Educandos basin has an area of 46.14 km^2 with a perimeter of 48.11 km corresponding 10.22% of Manaus urban perimeter. Its drainage area involves 22 neighborhoods. This region harboring more than 500 industrial enterprises showing a high technological index. Urban and industrial district are responsible for high sewage discharges into Educandos floodplain and Negro River [3,4]. As a result, high contents of heavy metal are found in aquatic ecosystem [(Pio, Souza, and Santana 2014),(Santana 2015)], soil [(Souza and Santana 2014)], and fish [(Santana 2016)].

Heavy metal pollution in water bodies (both surface and groundwater) is a serious environmental problem. Because of the high solubility in the aquatic environments, heavy metal has absorbed by living organisms and entered the food chain. Large concentrations of heavy metal may accumulate in the human body and can cause serious health disorders [(Cheng et al. 2002)]. Heavy metals could also induce impairment and dysfunction in blood, cardiovascular, endocrine, immune, reproductive and urinary system. Pb, for example, is known at elevated level, to affect intellectual performance in children and impairment of cognitive development in adults [(Majolagbe, Kasali, and Ghaniyu 2011)]. Therefore, it is necessary to treat metal-contaminated wastewater prior to its discharge to the environment. Conventional treatment processes such as chemical precipitation, ion exchange, and electrochemical removal can achieve heavy metal removal from inorganic effluent. These processes have significant disadvantages, which are, for instance, incomplete removal, high-energy requirements, and production of toxic sludge [(Barakat 2011)].

Among the several technologies used for wastewater treatment, two deserve highlights: Constructed wetland (CW) and electrochemical reactors (ER). The ER offers an efficient means of pollution control through redox reactions, promoting the removal and/or destruction of pollutants, directly or indirectly, in cells or reactors, with almost total independence of adding Chemicals [12]. ER is a method widely used to remove heavy metal in wastewater is the electrodeposition. This method has ability of removing heavy metal high concentrations. However, the heavy metal

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removing process is very expensive because of the electrical energy consumption [13]. On the other hand, phytoremediation that use the aquatic and/or terrestrial vegetation to remove, detoxify or stabilize persistent pollutants such as heavy metal is an alternative very cheap for cleaning soil and water [6]. Particularly, phytoremediation methods with aquatic plants, known as CW, has shown efficiency, low-cost and low-maintenance treatment systems for removing heavy metal from municipal or domestic wastewaters [7]. The long time for cleaning the contaminated water is the principal problem attributed to CW use. The CW integration is the best management practice concept are a sustainable means of treating storm water and more economical (e.g., construction and maintenance) and energy-efficient than traditional centralized treatment systems [(Lee, Scholz, and Horn 2006),(Pio, Antony, and Santana 2013)].

The linking of the ER and CW has some advantages such as reducing of cost associated with increasing of efficiency of water cleaning. This work we show finding of pilot system build using to CW of *Alocasia Macrorrhiza*, phytoremediator of heavy metal [(Lin et al. 2009),(Truong and Barbara 2001)], coupled with ER containing iron/aluminum as electrodes.

ER Optimization

For made ER, we essayed plate electrodes of graphite, iron, aluminum and copper with a dimension at 500mmx50mmx6.5 mm. According to AKbal and Camci [(Akbal and Camci 2011)], different materials could be used as electrodes for electrocoagulation. However, these authors claim iron and aluminum are efficient for cleaning metal-contaminated wastewaters. We assayed twelve plate electrodes formed by combinations of graphite, iron, aluminum and copper using a oscilloscopic DPO3034 and C0105339 Tektronix generator (Figure 1).The experiment was conducted with wave amplitude 3.00 Vpp and frequency 106 kHz by the function generator, oscilloscope recorded the attenuation voltage electrochemical cell, by Ohm's Law, was possible to calculate the pair higher current generator. The cathode/anode area ideal proportion was tested with the pair of lower resistivity electrode under the following conditions: voltage 1 V, current 1 A, 30 min and 1 cm distance between electrodes (Table 1). This initial configuration on average has widespread recommended in the literature [4,5]. The best combination of variables was chosen using full factorial design 24 [5].

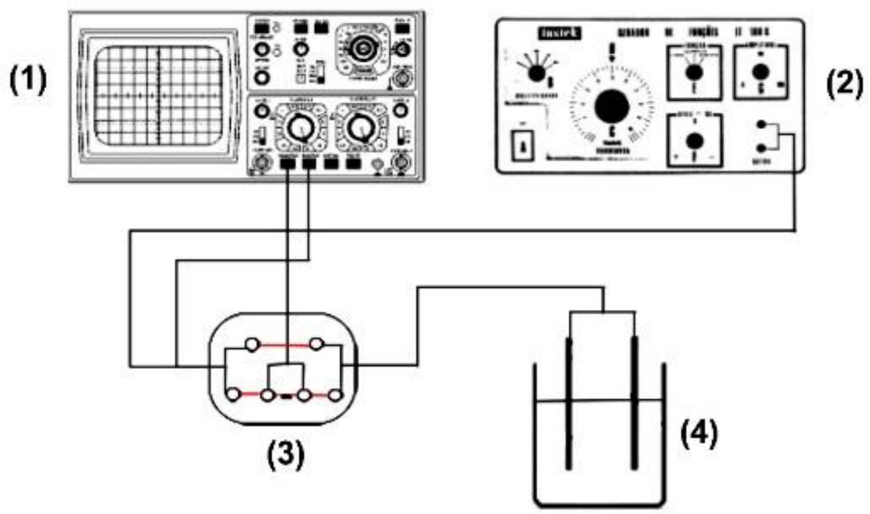

Figure 1 – (1) digital oscilloscope, (2) arbitrary function generator, (3) a circuit with known resistance, and (4) a pair of electrodes.

The plate electrodes performed with an arbitrary function generator (C010539), oscilloscope (DPO3034) with a known resistance and electrical contacts

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established with crocodile clips. The electrodes ran in a bipolar mode, so only the power supply connected outer electrodes. In addition, to remove the oxide and/or passivation layer from the electrodes, the electrode surfaces were ground with sandpaper before each experiment. Preliminary, we evaluated the electrode efficiencies for removing MPT by modifying the size relationship between cathode and anode, choosing pairs of electrodes with lower resistivity and an ideal range under conditions: 1.0 V voltage, 1.0 chain, 30 min and distance from the electrodes of 1 cm. For getting the ideal electrical conductivity as well as the conversion rate of the operation potentiostat system, we arranged the plate electrodes in a disposal parallel. The performance of electrocoagulation with plate electrodes of Al, Fe, Cu, G point to the coefficient of variation of 4.6% with low values of electrical resistivity ($\Delta\rho = \rho_{maximum} - \rho_{minor} = 0.052\ \Omega m$). By having the lowest resistivity as well as high reduction of heavy metal, we choose the combination Al/ Fe (anode/cathode). The combination Al/Fe advantage is the operational simplicity to produce the heavy metal electrocoagulation, mainly chromium. For choosing the best combination of variables, we used factorial design analysis 2^4 (Table 1) using to R program [(Barros Neto, Scarminio, and Bruns 2003),(Team 2012)]. Table1 shows that factorial design suggested the following experiments as the best heavy metal reduction: Cu (assay 7), Zn (assay 10), and Cr (assay 16). However, the major reduction for all MPT occurred in the assay 9.

Table 1 - Results of factorial design 2^4

Assay	A	B	C	D	Cu	Zn	Pb	Cr	Total Remotion %
					---------- mg L^{-1} ----------				
1	1	1	1	90	7.75	35.31	4.56	7.54	80.03
2	2	1	1	90	13.02	23.84	12.24	2.08	81.48
3	1	2	1	90	7.98	27.68	2.78	8.79	82.90
4	2	2	1	90	17.89	30.46	3.01	1.74	80.78
5	1	1	2	90	5.59	24.42	1.91	5.77	86.35
6	2	1	2	90	10.49	22.77	2.00	2.41	86.37
7	1	2	2	90	2.71	25.74	2.27	5.17	87.01
8	2	2	2	90	16.31	30.21	10.24	1.50	78.91
9	1	1	1	150	3.91	20.71	4.06	1.30	89.15
10	2	1	1	150	7.53	19.41	9.73	1.41	86.22
11	1	2	1	150	8.51	24.07	7.68	0.39	85.28
12	2	2	1	150	9.17	22.33	5.35	0.32	86.54
13	1	1	2	150	2.64	24.36	4.19	1.73	88.08
14	2	1	2	150	11.26	22.67	13.99	1.63	82.06
15	1	2	2	150	3.91	21.39	2.66	0.99	89.52
16	2	2	2	150	10.15	22.34	9.53	0.55	84.59

A = current (1 and 2 A), B = voltage (1 and 2 V), C = distance of the electrodes (1 and 2 cm) and D = time of electrolysis (90 and 150 min.)

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The high current values used in this causes an increasing of pH values of 3.90 to 7.50. The elevation of pH value carries out two processes: metal precipitation as hydroxides and/or metallic ion adsorption on the surface plate electrodes. Additionally, Table 2 shows the high solubility reduction of Pb, Zn, Cu and Cr with the variation of the pH value.

Table 2 – Solubility value in the pH interval from 3.93 to 7.58

Ion metallic	Kps	pH	
		3,93	7,58
Pb^{2+}	$1,43x10^{-20}$	2,26	$1,43x10^{-7}$
Zn^{2+}	$3,0x10^{-17}$	$4,75x10^{3}$	$3,0x10^{-4}$
Cu^{2+}	$4,8x10^{-20}$	7,60	$4,8x10^{-7}$
Cr^{3+}	$6,3x10^{-31}$	$9,98x10^{-11}$	$6,3x10^{-18}$

CW-reactor pilot system

The CW consisted of two layers: pebble (depth 15 cm) and white sand (depth 10 cm), added in polyvinyl chloride tank (PVC) at 150 L and sterilized with HNO_3 10% (Figure 2). We planted the *Alocasia macrorrhiza,* collected in preserving the area from the Federal University of Amazonas, in white sand at intervals of 10.0 cm. During 10 days, the *Alocasia macrorrhiza* irrigates with distilled water. Finally, we connected CW and RE systems. Another CW+ER system also was made but without the *Alocasia macrorrhiza*.

The ER has a built-in tank (8 L), with electrodes fixed by threaded galvanized iron bars. Its hydraulic system is of the continuous type, with electrodes arranged in parallel, the chosen electrical connection was the monopolar type with potentiostat operation. As the power supply was used the model DC Power Supply FA - 3005, with a capacity to supply 30 V and 5 A. To check the current variation was used multimeter Nippon America ST-5505 model. The reactor has an upper inlet to collect the water-treated CW and a bottom outlet tap of a 3/8 device connected to the outside of the tank.

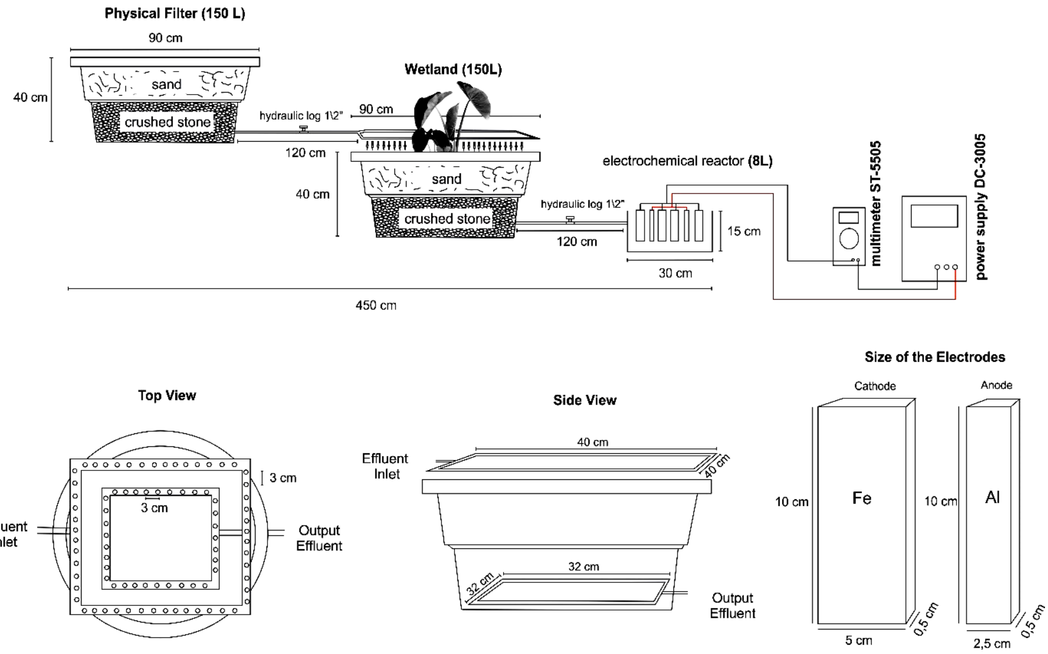

Figure 2 – CW system Scheme/ER to remediate water contaminated by MPT. T1 tank (filter) - filled with pebble and sand, T2 tank - CW - sand, pebble and plant species, T3 - ER.

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Contaminated water treatment

We checked the system efficiency with water samples, which was collected in the Igarapé do Quarenta (IG40) stream, located Educandos basin (Manaus, AM). The IG40 has received a high discharge of wastewater contained a high heavy metal concentration from Manaus Industrial District [(Pio, Souza, and Santana 2014),(Souza and Santana 2014),(Santana 2016)]. Initially, the *Alocasia macrorrhiza* stayed in contact with contaminated water for 48 h in CW. After, aliquot of 10 mL of water was collected from CW in intervals 24 h until 192 h. Thus, we submitted water to ER for 1.5 h. We executed this batch experiment in triplicate.

Heavy metal analysis

For plant analysis, we rinsed with deionized water and separated the roots, leaves, and stems, oven-dried at 60 °C until constant mass. The part plant was ground a fine powder in a high-speed Tekater mil. The protocol used in tissue digestion consisted of 15 mL of concentrated HNO_3 in a 50 mL digestion tube, mixed gently by swirling and allowed to overnight. We placed the digestion tubes in a heating block set at 150 °C for 1 h. We sampled the upper part for Zn, Pb, Cr, and Cu concentration determination with a GBC atomic absorption spectrometer AS932. Water sample analysis consisted in the use of an aliquot of 100 ml of water before filtered through a 0.45 μM Millipore membrane pore been pre-concentrated with 10 mL of concentrated HNO_3 at 100 °C until 10 mL and final volume complete to 25 mL with deionized water [(Santana and Barroncas 2007)].

Efficiency of the CW+ER system

According to batch experiment, the contaminated water from IQ40 showed varied concentrations of Cu, Pb, Zn, and Cr (Table 3). Commonly, metal concentrations have a concentration above the level permitted by Brazilian Environmental protection law, number 357/2005, Conselho Nacional do Meio Ambiente - CONAMA [("Conama 257/2005" 2014)] (Table 3) indicating low water quality. The metal-contaminated IQ40 water is a result of the uncontrolled urbanization process accompanied by contaminated wastewater release throughout the Manaus city has increased last years.

Table 3 – Metal concentration obtained in CW/RE batch experiments

	Batch experiment	Cu	Dp	Zn	Dp	Pb	Dp	Cr	Dp
IG40 water	1	0.13	0.09	1.83	0.03	0.19	0.04	0.04	0.01
	2	0.19	0.22	1.47	0.06	0.29	0.02	ND	ND
	3	0.27	0.41	1.83	0.09	0.79	0.01	ND	ND
CW	1	0.06	0.02	0.38	0.07	0.15	0.03	ND	ND
	2	0.08	0.02	0.29	0.09	0.20	0.05	ND	ND
	3	0.22	0.06	0.30	0.48	0.29	0.08	ND	ND
Alocasia Macrorrhiza	1	0.28	0.41	2.13	0.01	0.23	0.05	0.01	0.22
	2	0.23	0.02	2.35	0.07	0.32	0.06	0.016	0.02
	3	0.22	0.06	1.30	0.04	0.42	0.02	ND	ND
ER	1	0.02	0.07	ND	ND	0.13	0.03	ND	ND
	2	0.03	0.03	ND	ND	0.15	0.01	ND	ND
	3	0.01	0.01	ND	ND	0.14	0.02	ND	ND
CONAMA("Conama 257/2005" 2014)	Classe 1	0.009		0.18		0.01		0.05	
	Classe 3	0.013		5		0.033		0.05	

ND = non-detected, DP = standard deviation

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The water treatment by CW reduces the metal concentrations according the following order: Zn (81.0%) > Cu (43.4%) > Pb (38.5%) (Figure 3). The CW performance permitted removal a reasonable amount of Zn, Cu, and Pb during batch experiments. Despite the metal reduction, the Zn, Cu, and Pb concentrations are major than recommended by Brazilian law yet. The Zn, Cu, and Pb concentrations show that the *Alocasia Macrorrhiza* is tolerant to metal-contaminated water IQ40. Truong and Hart [(Truong and Barbara 2001)], Liu et al. [(Liu et al. 2010)], Qi-Tang et al. [(Qi-tang et al. 2002)], and Zaman et al. [(Zaman and Mangkoedihardjo 2014)] also related the heavy metal tolerance of the *Alocasia Macrorrhiza*. However, the metal concentration values into the *Alocasia Macrorrhiza* do not show the potential phytoremediator for Zn, Cu, and Pb. The ER show the following sequence of heavy metal reduction: Zn e Cr (~100%) > Cu (95.5%) > Pb (51.7%). It observes a heavy metal reduction according to CONAMA, except Pb. The Figure 3 shows the heavy metal reducing process in pilot system.

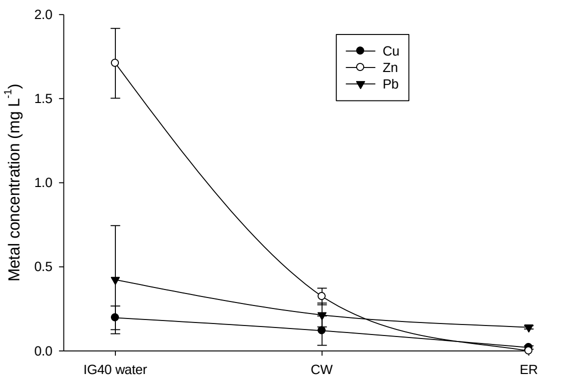

Figure 3. Heavy metal reducing in CW+ER pilot system.

4. FINAL CONSIDERATIONS

The results showed that CW-ER system reduces the heavy metal concentration into contaminated water from IQ40. The heavy metal concentration obey the following sequence of reduction in CW: i) Pb showed the major reduction rate; ii) Cu has the minor reduction concentration rate, and iii) Cr had concentration in level recommended by CONAMA [20]. The *Alocasia Macrorhiza* reduces the heavy metal concentration according to the sequence Zn > Pb > Cu > Cr. ER showed the following reduction heavy metal concentration: Zn e Cr (~100%) > Cu (95.5%) > Pb (51.7%). Our findings suggest ending cleaning heavy metal level according to the CONAMA recommendation [20], except Pb. The Pb cleaning could improve with elevation of time in CW, high the plant number or use of new plant species.

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