

Phytoremediation in Removing Selected Heavy Metals from Aqueous Solutions

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(Received: 8 March 2017 - Accepted: 6 November 2017)

Abstract - Bioaccumulation of *Ulothrix zonata* was compared for cadmium, copper, nickel, and zinc removal at various concentrations and contact time. The results of the present investigation showed that the lowest amount of Cd, Cu, Ni and Zn was adsorbed when the initial heavy metal concentration was 10 ppm whereas Cd showed highest concentration at 20 ppm and Cu exhibited greatest removal at 30 ppm. The metals Ni and Zn exhibited highest removal at 40 ppm. The value of Freundlich model constant ($1/n$) for different metals ranged from 0.334 to 0.721 and the values of Langmuir separation factor values (R_L) varied between 0.111 and 0.722 which indicated favorable bio-sorption by the biomass of the algae. The order of metals uptake was found to be Ni>Zn>Cu>Cd. The finding of the study showed that *U. zonata* has much potential as a biosorbent for the sorption of Cd, Cu, Ni and Zn and indicated that the metal uptake was concentration-independent for Cd and Zn whereas for Cu and Ni an increase in initial metal concentration resulted in higher metal uptake. This present study leads the way for the future studies on the use of algae as a cheap bio-adsorbent for heavy metals removal in Iraq which is suffering from huge inland waste water.

Key Words: Heavy Metals, Wastewater, *Ulothrix zonata* and Bio-sorbent.

Introduction

The contamination by heavy metals from industrial waste water to the water resources effect life significantly, because heavy metal ions can accumulate in the environment and into food chains (Chervona *et al.*, 2012). Therefore, the process of heavy metal removal is very important and industries should be aware of it. Many researchers around the world tried various technologies to remove the toxic heavy metals from aqueous solutions and the techniques includes adsorption, biological methods, electro coagulation, electro dialysis and various membrane separation techniques. Among the various techniques, Biological methods are very important area of research with huge potential for research and applicability for removal of heavy metals. Various biological methods which are recommended in the current scenario includes: trickling filter, bio-sorption, activated sludge process and various anaerobic processes.

The old technologies of heavy metal removal have several disadvantages such as high cost, chemical uses and large volume residual sludge. The phytoremediation is a new alternative method for heavy metal removal process because of low cost, low biological sludge, high efficiency and environmental friendly (Farooq *et al.*, 2010; Srivastava and Majumder, 2008; Abdel-Aty *et al.*, 2013).

The bioaccumulation process is known as an active mode of metal accumulation by living cells which depends on the metabolic activity of the cell (Volesky, 1990; Wase and Forster, 1997). Bio-sorption is a term that describes the removal of heavy metals by the passive binding to non-living biomass from an aqueous solution (Davis *et al.*, 2003).

Algae are important agents and are already been used by many wastewater facilities. The contamination of Shatt Al-Arab, southern Iraq by heavy metals was reported by many researchers (DouAbul *et al.*, 1987; Al-Saad *et al.*, 1996; Al-Saad *et al.*, 1997) and it was attributed to agricultural, industrial, anthropogenic activities around the river beside the ground water effects.

The indiscriminate discharge of chemical toxins especially Cd, Cu, Ni and Zn etc into the environment ensure their transfer into plants, animals and man, and it was also reported that high concentrations of heavy metals in irrigation waters could effects plants and crops growth, interfere with uptake of other essential nutrients or form dust deposit on fruits and render the edible portion of plants toxic to humans and grazing animals (Dan'Azumi and Bichi, 2010).

Algae have been proven efficient biological vectors for heavy metal uptake. Bio-sorption potential of two strains *Spirogyra* sp. and *Spirulina* sp. has been studied under different initial of metal concentrations (Mane and Bhosle, 2012). *Spirulina* sp. treated with different metal ions has been employed to understand the sorption for management of industrial wastewater (Doshi *et al.*, 2007). Deng *et al.* (2007) reported that the green algae *Cladophora fascicularis* is highly efficient for the bio-sorption of copper from aqueous solution.

Results showing high sorption of Pb⁺² from solution by biomass of commonly available filamentous green algae *Spirogyra* species (Gupta and Rastogi, 2008). Khalaf (2008) observed bio-sorption of textile dye from textile water by no viable biomass of *Aspergillus niger* and *Spirogyra* species. Brahmhatt *et al.* (2012) use the filamentous algae *Pithophora* species for the removal of cadmium, chromium and lead from industrial waste water. Gao and Yan (2012) observed the response of *Chara globularis* and *Hydrodictyon reliculatum* to lead pollution. Sheng *et al.* (2004) used the locally harvested brown marine algae *Sargassum soecies* and *Padina* species for the removal of cation of Cd⁺² and Cr⁺³ and anion (Cr⁺²) from diluted aqueous solutions.

The high biodegradation and bio-sorption capacity of some potential cyanobacterial species: *Oscillatoria* sp., *Synechococcus* sp., *Nodularia* sp., *Nostoc* sp. and *Cyanothece* sp. dominated the effluents and mixed cultures showed varying sensitivity. Contaminants were removed by all the species either as individuals or in mixtures (Dubey *et al.*, 2011). Lee and Chang (2011) observed the bio-sorption capacity from aqueous solutions of the green algae species, *Spirogyra* and *Cladophora*, for lead (Pb (II)) and copper (Cu (II)). In comparing the analysis of the Langmuir and Freundlich isotherm models, the adsorption of Pb (II) and Cu (II) by these two types of bio-sorbents showed a better fit with the Langmuir isotherm. In Saudi Arabia, the accumulation of heavy metals by the green algae *Chaetomorpha aerea*, *Enteromorpha clathrata* and *Ulva lactuca* were carried out to measure the level of iron, nickel, cobalt, zinc, cadmium and lead in three site of the Saudi coast of the Arabian Gulf (Al-Homaidan, 2007). Al-Mayaly (2011) studied the use of filamentous algae *Mougoutia* sp. to remove lead from contaminated water under laboratory conditions. It was found that the algae were able to remove this metal with high efficiency.

It was found that the filamentous green algae are more efficient in removing heavy metals than micro algae like *Chlorella vulgaris*. This may relate to the increasing of active sites according to the multiply of cell numbers in each filament (Akthar *et al.*, 1996). Furthermore, the chemical nature and polarity of the adsorbent surface can influence the attractive forces between the adsorbent and adsorbate (Babel and Kurniawan, 2003). Patrón-Prado *et al.* (2010) observed the rate of bio-sorption of cadmium and copper ions by non-living biomass of the brown macroalga *Sargassum sinicola* under saline conditions. They concluded that presence of salt did not significantly affect the rate of bio-sorption and there is antagonistic effect on bio-sorption when both these metals are present in the solution. Michael *et al.* (2015) studied the lower and upper tolerance levels of *Cladophora* to abiotic conditions (pH, salinity and nutrients concentrations) and found that *Cladophora* could be a possible candidate a new West Water Treatment tool in these experimental conditions.

Materials and Methods

U. zonate was collected from waste water of the polluted Shatt Al-Arab channels, southern of Iraq. Algae was washed under running tap water and double distilled water to remove other algae and any epiphytes and adhering foreign particles like sand and debris. The washed biomass was first air dried for 24 hrs. and then in an oven at 80°C to constant weight. The dried biomass was then ground in an analytical mill and then sieved through 2 mm mesh size sieve and stored in polyethylene bottles. Experiments were performed at room temperature in 250 ml Erlenmeyer glass flasks containing aqueous solution of Cd, Cu, Ni and Zn of known concentrations, i.e., 10, 20, 30, 40 ppm which was found to be sub-lethal for *U. zonata* in these contact times. Analytical grade cadmium nitrate [Cd(NO₃)₂], copper chloride (CuCl₂), nickel chloride (NiCl₂), and zinc chloride (ZnCl₂) were used to prepare the controls. An amount of 250 mg portion of biomass was added to each flask and the mixtures were agitated in a rotary shaker at 180 rpm. The contact times were 60, 120, 180, 240 minutes.

Aliquots of the powdered samples were analysed for heavy metals using the methods described by Kureishy (1991). Heavy metals content were quantified using Inductively Coupled Plasma-optical emission spectroscopy (ICP-OES; Perkin Elmer Optima-3300 RL). The amount of metal sorbed at equilibrium, q (mg.g⁻¹), which represents the heavy metal uptake was calculated from the difference in metal concentration in the aqueous phase before and after adsorption according to Basha *et al.* (2006). Adsorption from aqueous solutions at equilibrium is usually correlated by Freundlich and Langmuir isotherm (Freundlich, 1907).

$$q = K Ceq^{1/n}$$

In this model, K (l.g⁻¹) and $1/n$ are the constants to be determined from the data while q is the amount adsorbed and C is the equilibrium concentration in the solution, For a good adsorbent, $0.2 < 1/n < 0.8$ and a smaller value of $1/n$ indicates better adsorption and formation of rather strong bond between the adsorbate and adsorbent. Langmuir equation (Langmuir, 1916 & 1917) was expressed as:

$$q = q_{max} \frac{bCeq}{1+bCeq}$$

Where q_{max} is the amount of adsorption corresponding to complete monolayer coverage, i.e., the maximum adsorption capacity and b (l.mg⁻¹) is the Langmuir constant.

Results

Bioaccumulation capacities of the green algae *U. zonata* were studied for cadmium, copper, nickel and zinc removal at various concentrations (10 ppm, 20 ppm, 30 ppm, and 40 ppm) at different intervals of times (Figs. 1, 2, 3 and 4). Cadmium showed lowest metal uptake at 10 ppm ($q = 1.5$) in 60 min and highest at 20 ppm ($q = 8.23$) with contact time of 180 min. The minimum metal uptake for Cu was recorded at 10 ppm ($q = 2.5$) in the contact time of 60 min which increased into 7.21 (q) at 30 ppm in 180 min. For Ni and Zn the metal uptake increased with increasing initial concentration of the metal, with lowest uptake at 10 ppm ($q = 6.11$) and ($q = 2.45$) at 60 min contact time and highest at 40 ppm ($q = 16.17$) and ($q = 9.22$) at 120 min. The order of metal uptake for the dried biomass was found to be $Zn > Ni > Cd > Cu$.

The metal uptake capacity of the biomass was evaluated using Freundlich and Langmuir isotherms. The Langmuir isotherm (Langmuir, 1916 & 1917) represents the equilibrium distribution of metal ions between the solid and liquid phases. The q_{max} and b determined from the slope and y -intercept of the plot. Highest q_{max} value was observed for Ni (30.998 $mg.g^{-1}$). The linear form of Freundlich adsorption isotherm was used to evaluate the sorption data. The values of $1/n$ ranged between 0.334 and 0.701 which indicates good adsorption. The separation factor values (R_L) indicated (Table 1). For Zn, Ni and Cu Freundlich model fitted satisfactorily as depicted by high values of correlation coefficients R^2 .

The results of the study indicated that the metal uptake was concentration-independent for Cd and Zn whereas for Cu and Ni an increase in initial metal concentration resulted in higher metal uptake (Figs. 1, 2, 3 and 4).

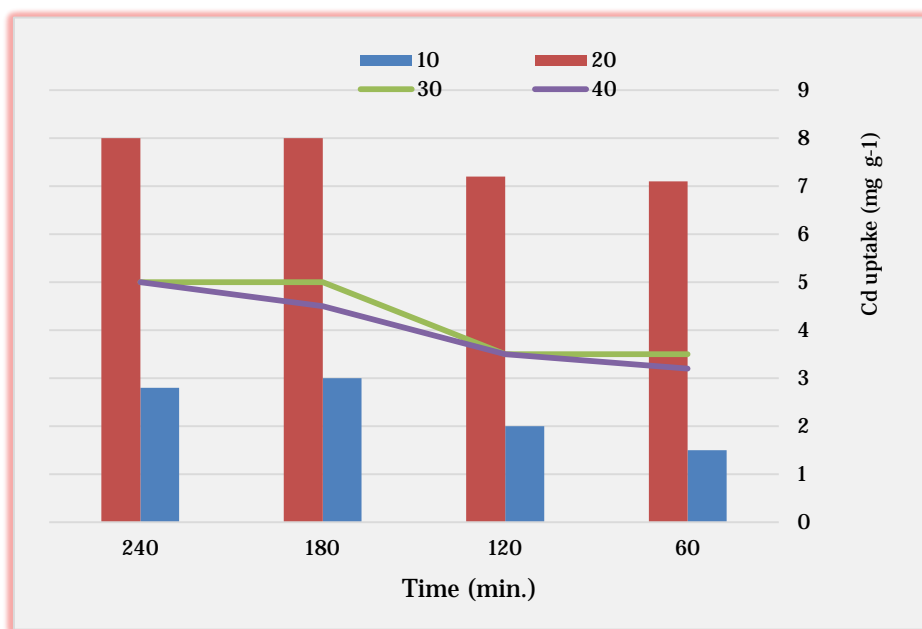


Figure 1. Cadmium uptake by *U. zonata*.

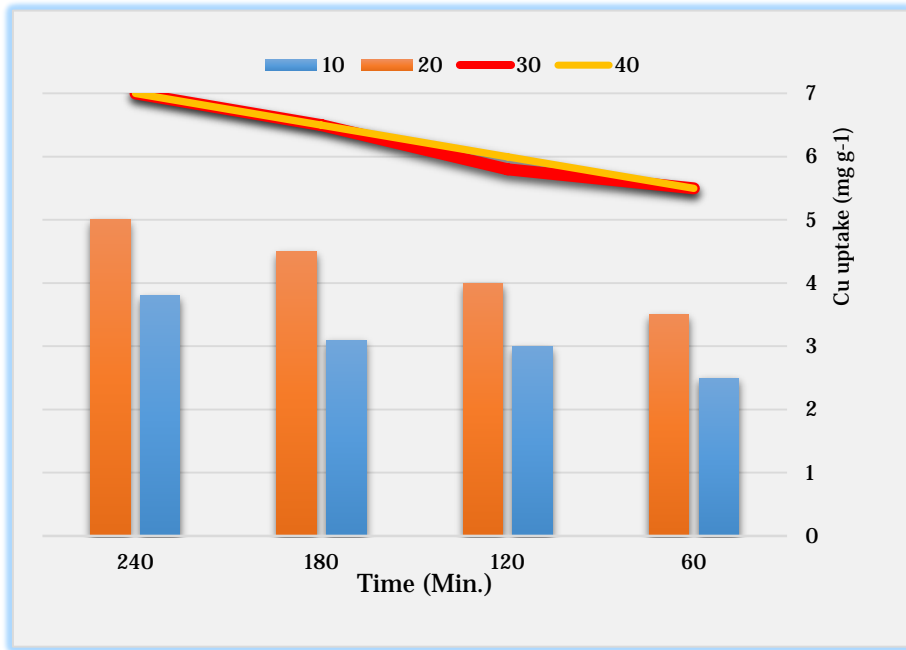


Figure 2. Copper uptake by *U. zonata*.

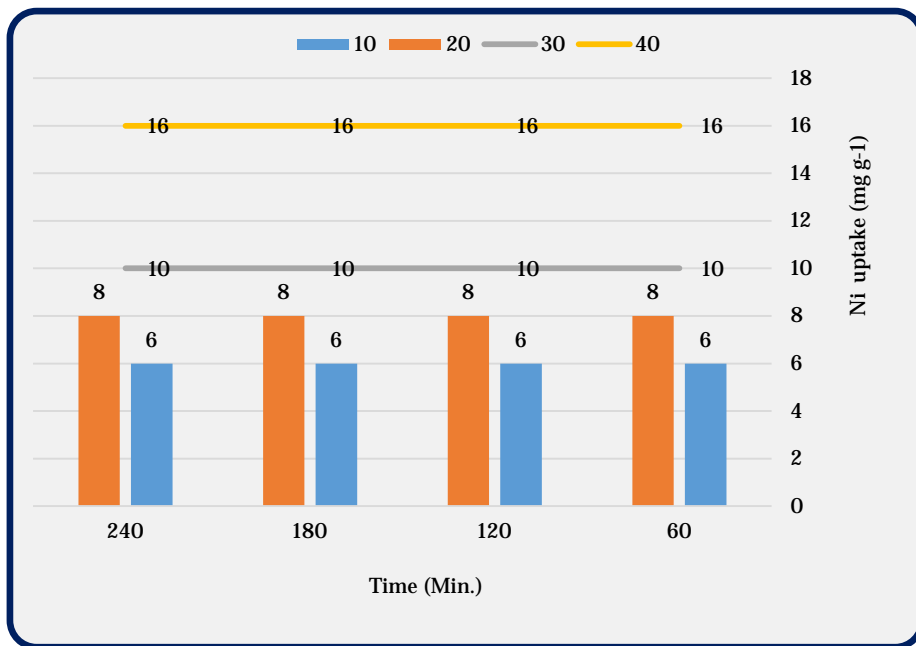


Figure 3. Nickel uptake by *U. zonata*.

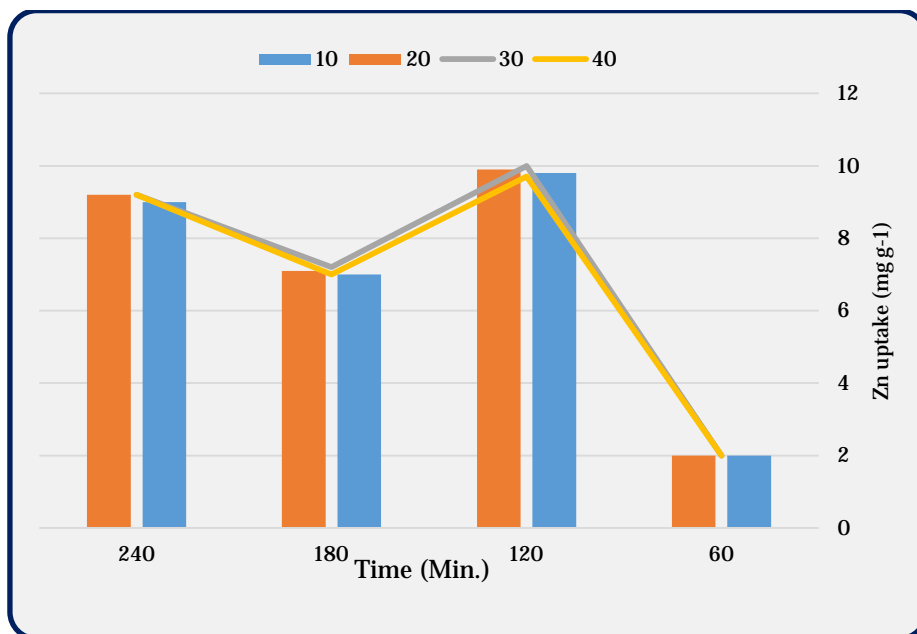


Figure 4. Zinc uptake by *U. zonata*.

Table 1. Isotherm constant for (Cd), (Cu), (Ni) and (Zn) bio-absorption.

Isotherm constants	Cd	Cu	Ni	Zn
q_{\max} (mg.g ⁻¹)	4.799	12.811	30.998	4.808
b (l.mg ⁻¹)	0.122	0.061	0.098	0.059
R_L	0.246±0.110	0.518±0.299	0.477±0.288	0.722±0.022
R^2	0.397	0.454	0.877	0.922
Freundlich				
$1/n$	0.334	0.565	0.682	0.701
R^2	0.082	0.639	0.966	0.991

Discussion

Evident from the results, for all the metals studied, the least metal uptake was recorded at 10 ppm while initial concentration for highest uptake varied in different metals. Among all the metals studied, Cd recorded decrease in the metal uptake at higher concentrations after attaining the maximum values. This appears to be the same results obtained by Ahalya *et al.* (2005) for the same metal and explained due to the increase in the number of ions competing for the available binding sites in the biomass and also due to the lack of binding sites for the complexation of these ions at higher concentration levels.

At lower concentrations, all metal ions present in the solution would interact with the binding sites and thus facilitate maximum adsorption. At higher

concentrations, more ions are left unabsorbed in solution due to saturation of binding sites (Ahalya *et al.*, 2005). Also, contact time for minimum uptake was found to be 60 min for all the metals. However, contact time for maximum uptake also differed for different metals. The separation factor values (R_L) indicated (Table 1) that metal sorption onto the biomass was favorable (Hall *et al.*, 1966). According to Kadirvelu and Namasivayam (2000), n values between 1 and 10 represent beneficial adsorption. The results values of $1/n$ ranged between 0.334 and 0.701 which indicates good adsorption (Table 1). The magnitude of K and n shows easy separation of heavy metal ion from wastewater and high adsorption capacity (Ahalya *et al.*, 2005). The value of n , which was related to the distribution of bonded ions on the sorbent surface, was found to be greater than unity indicating that adsorption is favorable. Tien (2002) found that the magnitude of K and n showed easy uptake of surface area and dry weight of algal cells. It was found to be the main factor influencing metal sorption and indicates favorable Huge load of wastes from industries.

Bio-sorption using biomass derived from fresh water algae, marine seaweeds and fungi has recently attracted growing interest of researchers. Many potential binding sites occur in algal cell walls and alginate matrices (Tam *et al.*, 1994; Saitoh *et al.*, 2001). Algal cell surface has several kinds of functional groups with varying affinity for an ionic species. Low and high affinity functional groups are involved in sorption of metal ions at high and low concentrations of metal ions, respectively (Mehta and Gaur, 2001). Cell wall of green algae contains heteropolysaccharides, which offer carboxyl and sulfate groups for sequestration of heavy metal ions. *U.zonata* was found to record high uptake values for Ni, Zn and Cd. Algal cells have showed considerable potential in removal of heavy metal from aqueous solutions. Alpana *et al.* (2007) observed 97 % removal of Pb^{2+} by *Pithophora odeogonia* and 89 % removal by *Spirogyra neglecta* in at 30 min from a solution containing 5 mg l⁻¹ initial concentration of Pb^{2+} by a biomass concentration of 1 g l⁻¹. Further, higher removal of 133.3 mg Cu (II) g⁻¹ of dry weight of Biomass of *Spirogyra* species was observed in 120 min contact period with an algal dose of 20 mg l⁻¹ by Gupta and Rastogi (2008). The results of this study indicated that the metal uptake was concentration-independent for Cd and Zn whereas for Cu and Ni an increase in initial metal concentration resulted in higher metal uptake, which was in agreement with the results obtained by Ahuja *et al.* (1999) for cobalt where, increase in cobalt concentration resulted in the increased uptake of the metal.

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إزالة بعض العناصر الثقيلة من المحاليل بواسطة المعالجة بالطحالب

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المستخلص - لقد تم مقارنة الأستهلاك الحياتي للطحلب *U. zonete* للعناصر الكادميوم والنحاس والنيكل والخرصين في تراكيز مختلفة وفترات مختلفة. أوضحت النتائج بأن أقل كمية تم امتصاصها من العناصر Zn, Ni, Cu and Cd كانت عند تركيز 10 ppm، بينما كان أعلى امتصاص للكادميوم عند تركيز ppm، و كان أعلى امتصاص للنحاس عند تركيز 30 ppm، أما النيكل و الخرصين فكان أعلى امتصاص لهما عند تركيز 40 ppm، وكانت قيمة ثابت موديل Freundlich (1/n) للعناصر المختلفة تتراوح بين 0.334 إلى 0.721، وكذلك قيمة عامل الفصل لـ Langmuir يتراوح بين 0.111 إلى 0.722 موضحاً امتصاصاً لصالح كتلة الطحالب المستخدمة. إن ترتيب أخذ العنصر كان كالآتي $Cd > Cu > Zn > Ni$. إن النتائج أوضحت بأن الطحلب *U. zonata* له قابلية كبيرة للامتصاص الحياتي للعناصر Zn, Ni, Cu and Cd وكان عنصر الكادميوم Cd والخرصين Zn غير متأثرين بالتركيز الأولي لهما في المحلول، بينما عنصر النيكل Ni والنحاس Cu فقد كانا متأثرين بزيادة تركيزهما في المحلول حيث إن أي زيادة في التركيز تبعها زيادة في أخذ العنصر. تعتبر الدراسة فاتحة لدراسة مستقبلية باستخدام الطحالب كمصدر امتصاص حياتي رخيص لأزالة العناصر الثقيلة في نهر شط العرب في البصرة - العراق الذي يشكو من كثرة المياه الملوثة.