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The Treatment of Polluted Water by Heavy Metals in Maysan Governorate by Phytoremediation

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Abstract

The environmental difficulties faced by Iraqi society, particularly in the countryside, worsen over time due to a lack of interest in environmental duties caused by a lack of awareness of the environmental risk. At a time when oil companies sought Iraqi government approval to invest in and develop Iraqi oil fields, particularly in the south of Iraq, heavy metals emerged as one of the most visible pollutants associated with the oil industry as by-products of oil extraction operations. Therefore, the environment is exposed to gradual demolitions that directly affect biodiversity and deteriorate water quality in the region. Consequently, it became necessary to find environmentally friendly treatment methods from the elements of the environment itself. Hence, the phytoremediation method was ideal for solving some pollution problems in the southern regions of Iraq, specifically the Iraqi marshes. This research discusses the impact of the most prominent heavy metals polluting the waters of Umm Al-Na'aj Pond and Al-Hawizeh marshland, proposes two methods for phytoremediation, and shows the efficiency of each of them in the adsorption of heavy metal pollutants.

Keywords: Al-Hawizeh Marshland, Umm Al-Na'aj Pond, Heavy metals, Water Pollution, Phytoremediation,

المعالجة النباتية للمياه الملوثة بالمعادن الثقيلة في محافظة ميسان

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الخلاصة

تتفاقم الصعوبات البيئية التي يواجهها المجتمع العراقي وخاصة في الريف مع مرور الوقت بسبب عدم الاهتمام بالواجبات البيئية الناجمة عن عدم الوعي الكافي بحجم الخطر البيئي. في الوقت الذي تسعى فيه الشركات النفطية إلى الحصول على موافقة الحكومات العراقية من أجل الاستثمار، وتطوير الحقول النفطية العراقية، خاصة في جنوب البلاد. تظهر المعادن الثقيلة كأحد أبرز الملوثات المرتبطة بصناعة النفط باعتبارها منتجات ثانوية لعمليات استخراج النفط. لذلك تتعرض البيئة لعمليات هدم تدريجية تؤثر بشكل مباشر على التنوع البيولوجي وتؤدي إلى تدهور نوعية المياه في المنطقة. لذلك أصبح من الضروري

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إيجاد طرق معالجة صديقة للبيئة ومن عناصر البيئة نفسها، فكانت طريقة المعالجة النباتية مثالية لحل بعض مشاكل التلوث في المناطق الجنوبية من العراق وتحديداً الأهوار العراقية. يناقش هذا البحث تأثير أبرز العناصر الثقيلة الملوثة لمياه بركة أم النعاج وهور الحويزة ويقترح طريقتين للمعالجة النباتية ويبين كفاءة كل منهما في امتزاز العناصر الثقيلة.

1. Introduction

Heavy metals have long improved human well-being but are also dangerous and pose a significant threat to human and aquatic life [1]. Heavy metals, including lead, arsenic, copper, zinc, and cadmium, are very toxic when they enter the biological system [2] [3]. The presence of heavy metals in aquatic ecosystems is becoming an increasing problem of concern about their influence on plant and animal life due to their tendency to remain an environmental pollutant for a long time and can penetrate through the food chain, causing harm to different species [4]. Heavy metals are bound to protein binding sites and induce cell distortion by moving original metals away from their usual binding [5]. Thus, heavy metal remediation from polluted sites is key to recovering site validity [6]; [7]. The presence of these metals in soil and water supplies should be controlled at levels that are compliant with international regular standards. There are several physicochemical methods for removing heavy metals, such as vapour extraction, incineration [8], solidification [9], thermal desorption [10], and soil looseness and the degradation of the heavy metals include several physicochemical methods for removing heavy metals [11]. A broad range of recovery strategies are used, but these methods generally help to contaminate air and groundwater, decrease soil fertility, and make them unsuitable for agriculture [12]. In addition, physicochemical methods are usually costly, limiting their extensive usage, particularly in developing countries, for improving polluted land [13]. During recent decades, bioremediation has become a growing and environmentally sustainable method using the natural potential of living organisms in the rectification of contaminated lands [14]. Usually, bioremediation may be graded as on-site or ex-site. The pollutants are extracted from the ex-situ bioremediation and treated elsewhere [15].

The study area is located in the southern part of Iraq within the Mesopotamian plain in Maysan Governorate, represented by the villages of Abu Khasaf and Al-Ma'ail, which Al-Ma'ail (Al-Zubair) river passes through. The study area is restricted to latitudes (31º 40′–31˚38′ N) and longitudes (47º 18′–47˚30′ E), (Figure 1). Al-Ma'ail River is one of the branches of Al-Kahla'a River from the rivers that flow into Umm Al-Na'aj Pond and Al-Hawizeh Marshland. The lower part of Al-Ma'ail River, represented by Umm Al-Na'aj Pond and Al-Hawizeh Marshland, is a significant habitat for many plant and animal species. On the left side of Al-Ma'ail River (Al-Zubair) lies next to the Halfaya oil field, extending along its length from its branching area from Al-Kahla River to its outlet in Umm Al-Na'aj Pond and Al-Hawizeh Marshland. On the right side of the river, cement and brick factories provide raw materials to the company operating in the Halfaya oil field. They are next to agricultural lands belonging to residents of marsh villages. Therefore, the environmental quality of Al-Ma'ail River has been subjected to great pressure from various human activities.

Figure 1: Study area and sampling sites

Oil exploration, refining, and distribution are closely linked to water supplies and qualities. Since petroleum products supply more than 90% of transport energy in almost every region, there is no question about a risk to water supplies in any petroleum activity. As water supply is under pressure as part of climate change and the growing population, in addition to waste generated by well-drilling activities, the environmental risks associated with oil exploration may also increase [16]. Different kinds of waste are produced by drilling oil and gas wells. Some of these wastes are natural by-products of drilling [17]. Drill cuttings and materials used to drill the well, such as drilling fluid and its related additives. The drill cuttings and the drilling fluid used to lift the cuttings from the well are the key ways drilling activities can affect the environment [18]. In general, all drilling muds have many unwanted components that can potentially harm the environment. The most common of these are heavy metals, salt, and HCs. The concentration of these materials varies significantly. The primary concern arises when the drilling fluid must be disposed of [19].

2. Materials and Methods

2.1 Analysis of heavy metals

The heavy metals that were selected for this study are:
Arsenic (As), Cadmium (Cd), Chromium (Cr), Copper (Cu), Lead (Pb), Nickel (Ni), Zinc (Zn), and Vanadium (V). The values of heavy metal concentrations in water samples are presented in Table 1. The evaluation of the results was made based on international and regional standards.

Table 1: Mean concentrations of heavy metals in water samples of the study area in ppm

Site	As	Cd	Cr	Cu	Ni	Pb	V	Zn
SW 1	0.122	0.126	0.39	0.68	0.73	0.93	0.076	1.15
SW 2	0.102	0.091	0.22	0.31	0.34	0.43	0.066	0.71
SW 3	0.092	0.095	0.25	0.13	0.21	0.28	0.063	0.63
SW 4	0.075	0.083	0.11	0.09	0.16	0.15	0.061	0.61
MEP[St.1], 2002 [20]	0.05	0.005	0.05 (Cr6)	1	-	0.01	-	-
CDWQ[St.2], 2006 [21]	0.01	0.005	0.05	1	-	0.01	-	5
IQS[St.3], 2009 [22]	0.01	0.003	0.05	1	0.02	0.01	-	3
E.U DWD[St.4], 2012 [23]	0.01	0.005	0.05	2	0.02	0.01	-	-
WHO[St.5], 2017 [24]	0.01	0.003	0.05	2	0.07	0.01	0.005-0.025	3
U.S EPA[St.6], 2018 [25]	0.01	0.005	0.1	1.3	0.07	0.015	-	5

St.: Standards
St.1: Ministry of Environmental Protection of People's Republic of China (MEP)
St.2: Canadian Drinking Water Quality (CDWQ)
St.3: Iraqi Quality Standards (IQS)
St.4: European Union Water Framework Directive (E.U DWD)
St.5: World Health Organization (WHO)
St.6: United States Environmental Protection Agency (U.S EPA)

3. Phytoremediation

Phytoremediation is a plant-based technology used to regenerate polluted soil and water supplies of untreated and genetically modified plant species [26]. The key justification for the phytoremediation project is the prospect of inexpensive repair [27]. The presence of heavy metals in soil, sediment, and water is usually the product of the mining, drilling, and warfare operations, in addition to the fact that in urban areas, heavy metals are primarily generated by transport, industrial pollution, wastewater sludge and tanning, electroplating, and the use of batteries [28]. The unparalleled prevalence of heavy metals in the atmosphere, such as arsenic, cadmium, chromium, copper, and lead, affects sociological and environmental health [29]. Oxidative stress is primarily caused by binding heavy metals to the DNA and nuclear proteins in living cells and biological macromolecules (Flora et al., 2008).

3.1 Criteria for Selecting Phytoremediation

Plant selection is based on various factors, including their ability to withstand pollutants, remediation properties, and adaptation to other site-specific variables [30]. Plants' most advantageous characteristics include tolerance to regional climatic conditions, the depth of the plant's root structure, the species' survival capacity as soil, the ability to extract or degrade the contaminants in question to a less toxic shape, a rapid growth rate, quick planting, and the maintenance and absorption of large volumes of water via evapotranspiration [31].

3.2 Phytoremediation in the study area

Natural phytoremediation is the process of introducing plants into the ecosystem and assimilating pollutants naturally through their roots without human or industrial interference.

Heavy metals, pesticides, and xenobiotics have been cleaned up by such processes as organic compounds [33], toxic aromatic pollutants [34], and acid mine drainage [35]. Phytoremediation is an environmentally friendly technology and is a safe and inexpensive way to eliminate pollutants, doing the same job as a group of engineers for one-tenth of the cost in some situations.

3.3 Phytoremediation by Phragmites australis

Many plant species work to adsorb heavy metals from their environment, whether aquatic or land. Among the plants with high adsorption capacity for heavy metals is the Phragmites australis. It is widely spread in the study area, especially in Umm Al-Na'aj Pond and Al-Hawizeh Marsh, as it forms large fields in the region's waters. In addition to the aesthetic that the reed plant gives to the environment of the marshes, it was found to have good efficiency in the adsorption of heavy metals. The process of monitoring and analyzing the adsorption capacity of the reeds for heavy metals was conducted according to the following practical steps:

3.3.1 Fieldwork

Three stations were selected for plant sampling, considering the similarity of climatic and environmental conditions and the difference in plant spread and density between the three stations to infer the ability of Phragmites australis field to adsorb at different plant densities. The three stations were within Umm Al-Na'aj Pond and Al-Hawizeh Marsh, as shown in Figure 1. Phragmites australis samples were collected in plastic bags, and after sampling, they were dried in an oven below 50 °C.

3.3.2 Laboratory work

A wet digestion method was followed to determine heavy metals in plant samples, which was modified by Rashid (1986) [36]. One gram of dry plant material is transferred to a 100ml Pyrex digestion tube, then 10ml 2: 1 nitric acid and perchloric acid mixture is added and left to stand overnight, or until the vigorous reaction phase ends, then a small short stem tube funnel is placed in the mouth of the tubes to reflux acid. After the initial digestion, the tubes are placed in a cold digester. The temperature is raised to 150 ° C for one hour, and then the temperature rises to 235 ° C, where dense white fumes of perchloric acid appear in the tubes. Digestion continues for another 30 minutes. The tubes lifted the rack from digesting mass, allowed cooling for a few minutes and then carefully added a few drops of deionized water (DI) through the funnel. After condensing the vapours, the solution from each tube is thoroughly mixed and left alone for a few hours. It is then evaluated using an atomic absorber.

Phragmites australis was chosen as an example of a naturally occurring plant healing process. The results of the analysis of Phragmites australis samples found that they contain concentrations of heavy metals, as shown in Table 2.

Table 2: Concentrations of heavy metals in Plant samples (Roots of Phragmites australis)

Sample NO.	Zn ppm	Pb ppm	Ni ppm	Cu ppm	Cr ppm	Cd ppm	As ppm	V ppm
SP1	18.09	2.21	0.142	8.612	0.624	0.241	0.427	1.47
SP2	16.28	1.98	0.118	7.415	0.589	0.198	0.34	1.32
SP3	15.78	1.04	0.102	7.098	0.501	0.097	0.301	1.02
(Kabata-pendias, 2001) *[37]*	150-200	-	20-30	15-20	1-2	5-10	-	-

Through the results of the laboratory analyses that were conducted on field water samples, especially for the waters of Al-Hawizeh Marsh, the readings were compared between SW1 sample (the beginning of Al-Hawizeh Marsh where the plant was less dense) with the sample SW4 (the end of Al-Hawizeh marsh where the plant was denser). Heavy metals removal in water samples according to the higher density of the plant in the water, cause the lower concentrations of heavy metals and distance from the sources of pollution (household sanitation wastewater, agricultural drainage water and Halfaya oil field), the concentrations of heavy metals in the water sample decrease. The percentage of heavy metals removal was calculated in the water samples within the botanical modelling areas, Umm Al-Na'aj Pond and Al-Hawizeh Marsh, between SW1 and SW4 according to the following equation:

$$Removal\ rate = \frac{C1 - C2}{C1} * 100\%$$

Where C1 is the first concentration, C2 is the second concentration [38]. Results of applying phytoremediation using roots of Phragmites australis show it was very good for copper element removal, reaching 86.76%, and then comes lead at 83.87%, nickel 78.08% and the lowest removal rate for vanadium element was 19.74% of its concentration at the beginning of Umm Al-Na'aj Pond (SW1), as shown in the Table 3 and Figure 2.

Table 3: Variations in removal % of heavy metals Initial (SW1) and Final (SW4) concentrations by Phragmites australis.

Metals	SW1 (Initial concentration) ppm	SW4 (Final Concentration) ppm	Removal ratio%
As	0.122	0.075	38.52
Cd	0.126	0.083	34.13
Cr	0.39	0.11	71.8
Cu	0.68	0.09	86.76
Ni	0.73	0.16	78.08
Pb	0.93	0.15	83.87
V	0.076	0.061	19.74
Zn	1.15	0.61	46.96

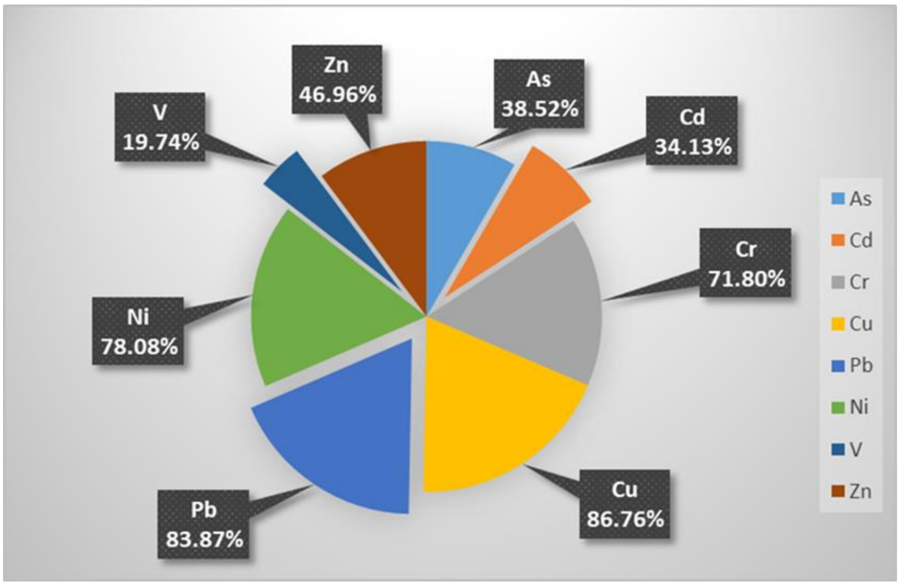

Figure 2: Variations in removal per cent of heavy metals after treatment with Phragmitesaustralis

3.4 Phytoremediation by *Eichhornia crassipes*

The second plant type used as another tool for phytoremediation in the study area is water hyacinth, a free-floating perennial aquatic plant native to tropical and subtropical South America. Water hyacinths rise over the water's surface with long, dense, glossy ovule leaves up to 50 cm high (Plate 1). The leaves are 10-20 cm long on a stem that floats on its base above the water surface with floating bulb-like nodules. Their stalks are long, spongy, and bulbous. Freely hanging roots are purple-black. Because of the effectiveness of Eichhornia crassipes (Water hyacinth) in adsorption of heavy metals from water [39], it was chosen as a model for conducting a laboratory experiment to estimate the percentage of removal of heavy metals by roots of Eichhornia crassipes plant on actual field water samples collected from the study area. The sample SW1 was selected from Al-Hawizeh Marsh water to stabilize the same field and climatic conditions in which Phragmites australis worked in the natural removal process to compare the removal ratios between natural and laboratory methods and suggest the best.

3.4.1 Fieldwork

Water hyacinth (Eichhornia crassipes***)*** was collected from the study area, and the samples were placed in a plastic bag. Samples were brought to the laboratory, and the branches were separated. The roots were washed thoroughly with tap water first and then frequently distilled water. The roots were left for five days to dry under the sun.

Plate 1: Eichhornia crassipes Samples

3.4.2 Laboratory work

The dried Eichhornia crassipes (Water hyacinth) roots were analyzed by digestion using the nitric acid-perchloric acid procedure described by AOAC (1990) [40]. 10 ml of heavy metals are already present in the roots, allowing us to estimate their concentration. 10 ml of concentrated nitric acid was added to 1 g of dried roots in a glass container and gently boiled for 30-45 minutes to oxidize all simple oxidizable materials. 5 ml of perchloric acid was added to the mixture and boiled until a white fume emerged. The mixture was cooled, and 20 ml of distilled water was added and boiled to reduce residual fumes. The water hyacinth root powder was refrigerated until the time of the experiment. After two days, the experiment was conducted. 1 g of dried water hyacinth and a ground root was mixed with 50 ml of each adsorbent solution at room temperature, and the mixture was stirred in a shaker device (technical laboratory) for 30 minutes (plate 2). At 32.3 ° C and pH 8, the water characteristics in the sampling sites by placing the shaker device with the mixture inside an oven after setting the oven to (32.3 ° C) and after a period of shaking, leave the absorbent and adsorbent mixture for 24 hours, without the hassle. The next day, this mixture was filtered with Whatman filter paper and centrifuged at 3000 rpm for 10 minutes [41]. The heavy metal content of the filter was measured using an Atomic Absorption Spectrophotometer (AAS) in the Ministry of Science and Technology's laboratories.

Plate 2: Steps of laboratory work in the laboratories of the Department of Geology

4. Interpretation of the results of the water hyacinth experiment

According to the results, one gram of water hyacinth root showed a remarkable ability to absorb heavy metals at different concentrations. In general, the highest bio-absorption of minerals (represented by the removal rate) was for cadmium (from 0.126 to 0.004 ppm with a removal percent of 96.8%) (Table 4 and Figure 3), while the lowest bio uptake was for zinc but still good removal (from 1.15 to 0.29 ppm with a removal percent of 74.78%). Arsenic (from 0.122 to 0.027 ppm, with a removal percent of 77.87%). After Cd removal comes Pb 89.46%, Cu 88.53%, Cr 84.87%, Ni 83.97% and V 81.58%. The result of this method, with a high percent removal of heavy metals, is considered very encouraging and interesting to apply in different sites suffering from environmental pollution with such metals. Besides, this kind of remediation is low-cost, clean, and eco-friendly. The interaction between the active sites on water hyacinth root powder and the minerals represented by the bio-absorption can be physically explained using van der Waals forces between the positively charged heavy metals in this study and the negatively charged particles in the adsorbents [42]. Adsorption can be attributed to ion exchange. The intrinsic absorption of substances is determined by their surface areas, as demonstrated by the effect of different adsorbent sizes on adsorption capacity [43].

Table 4: Variations in removal of heavy metals in % and their concentrations after treatment by water hyacinth root in (SW1)

Metals	Metals Concentration in plant sample root ppm	SW1 (Initial Concentration) ppm	Concentration after treatment with water hyacinth root ppm	Removal ratio%
As	6.87	0.122	0.027	77.87
Cd	5.18	0.126	0.004	96.82
Cr	3.47	0.39	0.059	84.87
Cu	9.97	0.68	0.078	88.53
Ni	2.54	0.73	0.117	83.97
Pb	8.4	0.93	0.098	89.46
V	3.89	0.076	0.014	81.58
Zn	14.12	1.15	0.29	74.78

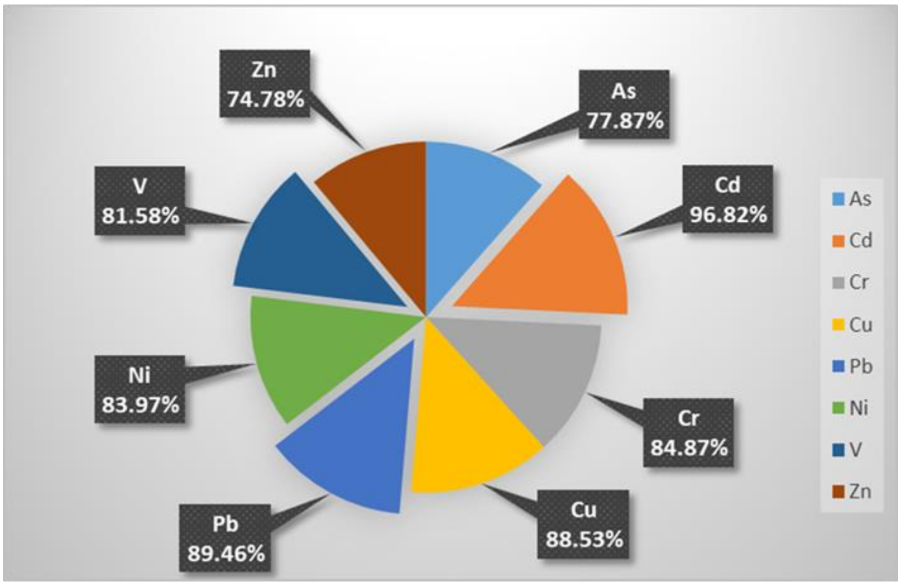

Figure 3: Variations in Removal percent of heavy metals in SW1 after treatment with water hyacinth root

5. Conclusion

The difference exists in the adsorption ability of the cadmium element in particular, as the percentage of its removal was 34.13% by Phragmites australis roots, while the percentage of removal using dried Eichhornia crassipes roots reached 96.82%. At the same time, the removal percent of vanadium, which was a characteristic of the laboratory method, reached 81.58%, while the removal percent in the natural method was 19.74%. The laboratory removal rates for arsenic and zinc were 77.78% and 74.78%, respectively, whereas natural removal was much lower, at 38.52% for arsenic and 46.96% for zinc. As for the rest of the metals, the removal percent was close between the natural and laboratory methods, with the preference for the laboratory method for all elements, as shown in Figure 4. The results of the removal of heavy metals were shown in the first two methods by evaluating the role of the naturally grown (Phragmites australis) plant in the study area by analyzing the SW1 sample before the water passed through the (Phragmites australis) and SW4 fields after the water left the field into a marsh in the middle of Al-Hawizah marshland. Vanadium had the lowest removal rate of 19.74%, while copper had the highest removal rate (86.76%). In the second method, the highest removal percentage was for cadmium 96.82%, and the lowest was for zinc 74.78%. The laboratory method using (Eichhornia crassipes) showed higher efficiency than (Phragmites australis) (plate 3 and plate 4), bearing in mind that both methods are poured into one bucket, which is proof of the efficiency of environmentally friendly natural methods in purifying water contaminated with heavy metals.

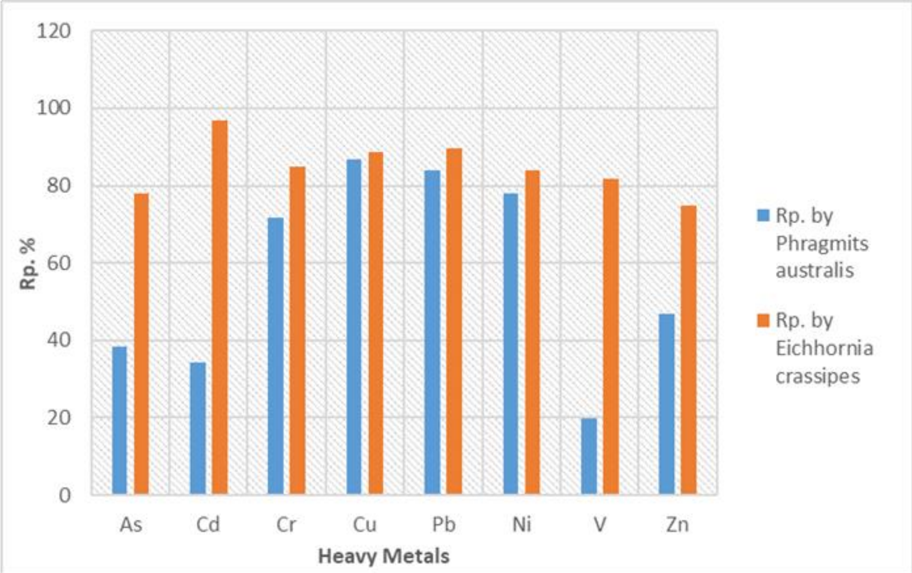

Figure 4: Differences in removal percent between the field and laboratory methods

Plate 3: Root of Phragmites australis sample Plate 4. Root of Eichhornia crassipes sample

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