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IMPROVE HEAVY METALS AND POLLUTANTS REMOVAL FROM THE PHARMACEUTICALS WASTEWATER USING WASHINGTONIA ROBUSTA: NEW EXTRACTION PROCESS

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Abstract

Jordan is one of the most water-scarce countries in the world. The country's renewable water supply currently only meets about half of the population's water demands. The aim of the present study was to investigate the changes in the level of heavy metals found in pharmaceutical wastewater from Al-Hikma Drugs Industry using palm fronds - Washingtonia Robusta (PF-WR) as organic adsorbents. Removal of heavy metals, turbidity, chemical, and biological oxygen demand, and suspended solids from industrial water using bio-adsorbent materials was studied. Increasing the concentration of PF-WR increased the removal efficiency. The optimum reductions of Fe, Ni, Cu, Cr, Pb, Co, Al, Na, COD, BOD, and turbidity were 64.29 %, 90.35 %, 61.75 %, 62.6 %, 60.60 %, 34.09 %, 10.09 %, 85.26 %, 87.41 %, and 78 %, respectively, were achieved by increasing the PF-WR concentration to 100 %. The removal efficiency was found to be concentration-dependent of PF-WR. The lowest reduction was achieved by sodium and pH.

Keywords:

Adsorption;
Heavy metals;
Physiochemical;
Palm fronds - Washingtonia Robusta.

1 Introduction

Water scarcity continues to be a key worry in developing nations such as Jordan, where accessible water resources are expected to fall by just 91 cubic meters per capita per year for all purposes by 2025 [1]. Furthermore, due to population increase and limited water resources, the gap between supply and demand for water in dry areas is fast expanding. Several studies have proposed methods of supplying water in tandem with wastewater management to address the scarcity of water suitable for consumption or agricultural irrigation [2, 3].

Under these conditions, a water management plan involving the reuse of non-conventional water resources should be undertaken [2, 4]. Non-conventional water reuse has the lowest marginal cost in Jordan and will thus remain the primary component of Jordan's water budget. Non-conventional water from diverse businesses nearly often contains hazardous contaminants such as heavy metals [4, 5]. In addition, plants' growth and development are slowed. Heavy metals (HMs) are necessary nutrients for plant growth and development since they are components of several physiological and biochemical processes [4]. Increased amounts of HMs in the soil and water, on the other hand, can cause poisoning symptoms in humans and growth inhibition in most plants [5, 6]. HMs produced by non-conventional water contributes significantly to environmental degradation and must

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be eliminated [6]. HMs present in excess of acceptable limits, or even at relatively low concentrations, have a negative impact on aquatic life, soil, plants, and human health, [5]. HMs can harm the body's immunological and neural systems, causing diarrhea, stomach aches, severe vomiting, fractures, and sterilization, as well as psychiatric problems, DNA damage, and cancer [6]. HMs has been discovered to be potent enzyme inhibitors [7], causing harm to plant physiological and biochemical processes.

Toxicity can result from HMs binding to enzymes due to their affinity for sulfur bonds, causing protein structure disruption and immobilization of enzymes, or from the displacement of an essential element, causing deficiency effects in these elements [8]. They also bind to the cell membrane, influencing transport activities via the cell wall. Irrigation with industrial water impairs root and shoot growth [9] by decreasing cell division, decreasing antioxidative enzyme activity in plants, and increasing oxidative stress in cells [10]. HMs may be removed from industrial effluent in a variety of ways. These procedures are classified as conventional or non-conventional. Precipitation, membrane filtration, ion exchange, co-precipitation, and adsorption are examples of conventional methods. High energy requirements, production of oxidation by-products, regeneration requirements during ion exchange, bulk toxic sludge generation in flocculation/coagulation methods, short half-life in ozonation, and membrane fouling during the filtration process are all problems caused by using these methods [10]. Adsorbents that are not conventional can be classed as inorganic adsorbents, organic adsorbents, or bio sorbents [5]. Adsorption was recognized as a cost-effective and technically feasible way of removing heavy metals from aqueous solutions [11]. Agricultural by-products are used in biosorption technology to remove heavy metals from aqueous media, making it one of the most promising non-conventional adsorbent techniques for HMs removal from water. In previous studies, a variety of organic wastes such as dead leaves of sawdust [12], rice husk [13], coconut shell, waste tea powder, coconut husk [14], and sugar cane bagasse [5] were employed to remove HMs and test their adsorption characteristics. It was a low-cost, successful technology for removing hazardous compounds from the waste stream [15].

There is no previous review that provides an overview of the adsorption capabilities of utilizing PF-WR as an organic adsorbent for the adsorption of various pollutants. The use of utilizing PF-WR as organic adsorbents as biomass-based adsorbents in the adsorption of different pollutants is demonstrated in this work. The aim of the present study was to investigate the changes in the level of HMs found in pharmaceuticals wastewater from Al-Hikma Drugs Industry using PF-WR as organic adsorbents.

2 Material and methods

2.1 Industrial water samples

The industrial water used in this experiment was obtained from Al-Hikma Drugs Industry. Samples were taken from the sediment tank. pH was measured by using the pH meter. The probe was inserted into the water sample, and the appropriate pH reading was taken.

The industrial water used in this experiment was obtained from al-Hikma Drugs Industry. Samples were taken from the sediment tank according to UNI 5667-13/2000 [2]. The COLORIMETER method was used to determine the parameters PH (hydrogen ion concentration in the wastewater and gives an indication of how acid or alkaline the wastewater), BOD (Biochemical oxygen demand in [mg/l]), COD (Chemical Oxygen Demand in [mg/l]), TDS (Determination of Total Dissolved Solids in [mg/l]), and TSS (Determination of Total Suspended Solids in [mg/l]) as reported by standard methods [16]. Turbidity, Fe, Ni, Cu, Cr, Pb, Co, Al, and Na concentrations were measured by a colorimeter method (Colorimeter DR890, Hach Lange, Germany) [17] according to the manufacturer’s procedures. The composition of industrial water from al-Hikma Drugs Industry, as well as drinking water guidelines set by the World Health Organization (WHO) and irrigation water set by Ayers and Westcot (1994), are summarized in Table 1 [18].

Table 1: Chemical composition of industrial water from al-Hikma Drugs Industry and recommended composition of drinking and irrigation water.

Parameters	Industrial water	Irrigation water (Ayers and Westcot, 1994) [18]
Ph	7.630	< 7.00
BOD [mg/l]	2300.00	< 10.00
COD [mg/l]	3154.00	< 100.00
TDS [mg/l]	2520.00	< 450.00
TSS [mg/l]	692.00	< 50.00
Turbidity (NTU)	371.00	< 2.000
Fe [mg/l]	0.280	5.000
Ni [mg/l]	1.140	0.200
Cu [mg/l]	0.200	0.200
Cr [mg/l]	0.075	0.100
Pb [mg/l]	0.018	5.000
Co [mg/l]	0.030	0.050
Al [mg/l]	0.066	5.000
Na [mg/l]	109.00	< 70.00

2.2 Adsorbent preparation

In this study, PF-WRs were used for the removal of HMs found in industrial water by liquid extraction. The palm leaves were washed with water to remove soluble contaminants from the leaves. The PF-WRs were dried in an electric oven at 100 - 105 °C for 24 hours. Dry leaves were then crushed in the laboratory by a jaw crusher and grinded with an agate mortar (Type BB1A, Masch.Nr.4323, Germany) and sieved to ≤ 1 mm in size. Afterward, two homogeneously powdered samples were taken from palm leaf powder, with a weight of 20 g for each sample.

The extraction of the palm leaves was obtained by a methodology based on [19]. The first sample was mixed with one liter of distilled water, and the second sample was mixed with one liter of ethanol 70 %. The two samples were centrifuged using Hettich universal, at 3000 rpm for 10 minutes, and then the leachate was transferred into sterile bottles and stored in the refrigerator until use. Concentration (10, 20, 30, 40, 50, 60, 70, 80, 90, 100 ml/l) of each extract was prepared by taking an appropriate amount of leachate (100, 200, 300, 400, 500, 600, 700, 800, 900, 1000 ml) and dissolving it in 1000 ml of distilled water.

3 Results and discussion

3.1 Evaluation of industrial water

The suitability of Al-Hikma drugs industry's industrial water discharge for agricultural and domestic purposes was assessed by comparing the values of various water quality parameters with those of irrigation water [20] and drinking water guidelines values, Table 1. The sample analysis reveals that the industrial water from al-Hikma Drugs Industry is not entirely fit for drinking with respect to BOD, COD, TDS, TSS, Turbidity and Ni because the concentrations of these parameters exceed the allowable limits of WHO. Based on HMs parameters except for nickel and sodium, industrial water samples can be utilized for irrigation purposes, but due to extreme levels of pH, BOD, COD, TDS, TSS, and turbidity over the guidelines for irrigation, decreasing these parameters is essential for utilizing this water for irrigation.

3.2 Solvent extractions

Statistical results of the use of PF-WR as adsorbent for the removal of heavy metals in industrial water are presented in Table 2.

Table 2: Analyses of variance of pH, BOD, COD, TDS, TSS,Turbidity, Fe, Ni, Cu, Cr, Pb, Co, Al and Na for industrial water in response to PF-WR as adsorbent.

	Concentration (C)	Solution (S)	Replications	C*S
PH	0.5*	0.3*	0.1*	0.01 ns
BOD [mg/l]	3135309*	333985*	123*	55359*
COD [mg/l]	6191485*	444440*	20*	27611*
TDS [mg/l]	1164496*	1258560*	182*	67253*
TSS [mg/l]	181178*	18001.5*	47 ns	1744*
Turbidity (NTU)	62912*	20705*	0.28 ns	988*
Fe [mg/l]	0.0249*	0.0025*	0.0002 ns	0.0004 ns
Ni [mg/l]	0.9021*	0.034 ns	0.008 ns	0.005 ns
Cu [mg/l]	0.011*	0.0008 ns	0.0003 ns	0.0004 ns
Cr [mg/l]	0.0014*	0.00005 ns	0.00002 ns	0.00003 ns
Pb [mg/l]	0.0001*	0.000009 ns	0.000001 ns	0.000004 ns
Co [mg/l]	0.00028*	0.00005 ns	0.000002 ns	0.000005 ns
Al [mg/L]	0.0003*	0.00002 ns	0.0002*	0.000006 ns
Na [mg/l]	90.1*	32.7 ns	84.2*	2.3*

* Significant difference at 5 % probability, ns - no significant difference between treatment, C*S Concentration (C) Solution (S).

3.3 pH value

The results showed that the pH value of industrial water was significantly affected by the PF-WR adsorbent concentration. The pH value behavior during the treatment process is illustrated in Fig. 1a, where the pH value decreased as the adsorbent concentration increased.

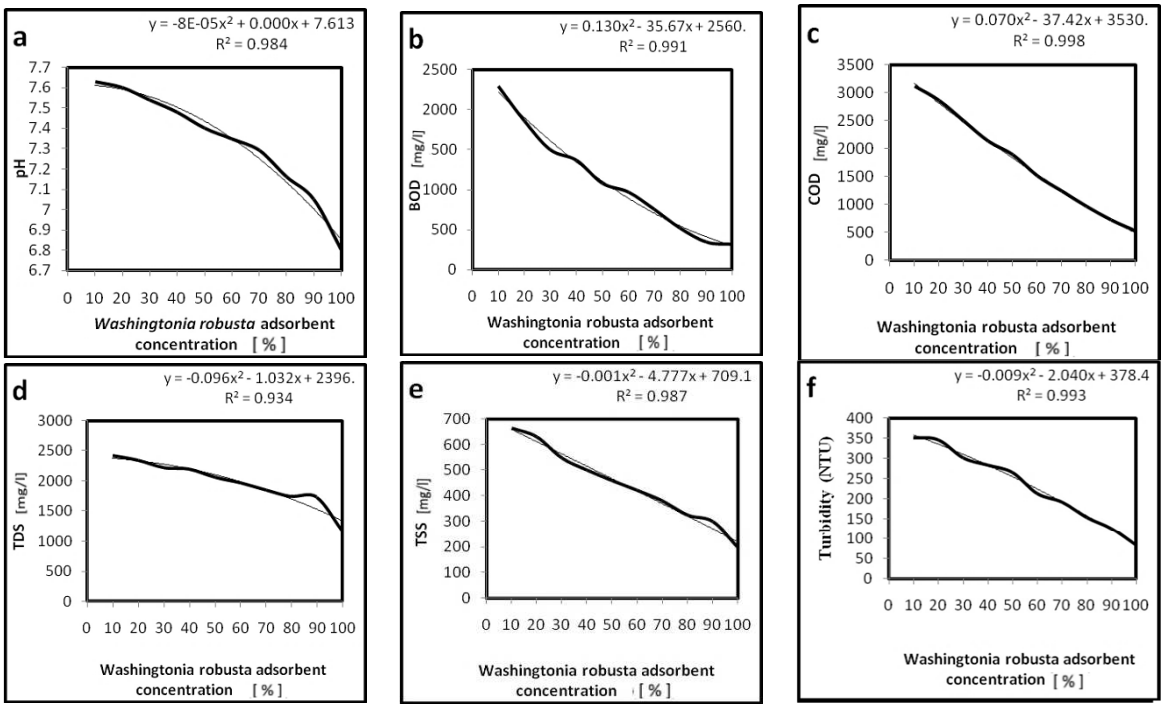


Fig. 1: Physiochemical parameters of industrial water as a result of adsorption using PF-WR extracts at various doses.

The value started to decrease significantly from the concentration of 40 %. The reduction in pH reached 11.4 % as a result of increasing the adsorbent concentration to 100 %. At the highest PF-WR concentration, the pH value returned to 6.8 where the initial value was 7.6.

3.4 BOD, COD, and turbidity removal efficiency

The effect of the adsorbent concentration on the removal of BOD and COD is depicted in Figs. 1b and 1c. The BOD removal percentage using PF-WR increased from 2.91 % to 85.26 % while the removal percentage of COD increased from 2.09 % to 87.41 % with the increase in the amount of absorbent concentration. This increment in removal percentage was statistically significant starting from an adsorbent dose of 10 %. Total dissolved solids and total suspended solids decreased sharply as a result of increasing adsorbent solution concentration, Fig. 1d. The reduction of total dissolved solids and total suspended solids in industrial water was found to increase with the increase of adsorbent dose from 10 % to 90 %, Fig. 1e. The turbidity varied as adsorbent concentrations were increased from 10 % – 100 %, Fig. 1f. The percentage removal of turbidity by PF-WR increased from 4 % to 78 %. The COD and BOD reduction was 2.91 % to 85.26 % and 2.09 % to 87.41 %, respectively. This decrease can be attributed to the concentration gradient between the sorbent and the sorbate; metal adsorption efficiency was increased with an increase in adsorbent dose. Optimum removal capacities were obtained for TSS, turbidity, COD, and BOD using the same adsorbent. Turbidity and suspended solids removal from high-strength wastewater using a high surface area adsorbent was observed by Amosa et al. [20].

3.5 Heavy metals

The findings of this investigation indicated that PF-WR powder has the potential to remove HMs present in industrial water. PF-WR clearly demonstrates strong potential to bio-remove heavy metals from industrial wastewater. Fig. 2 and Fig. 3 illustrate the effect of adsorbent dosages on HMs elimination.

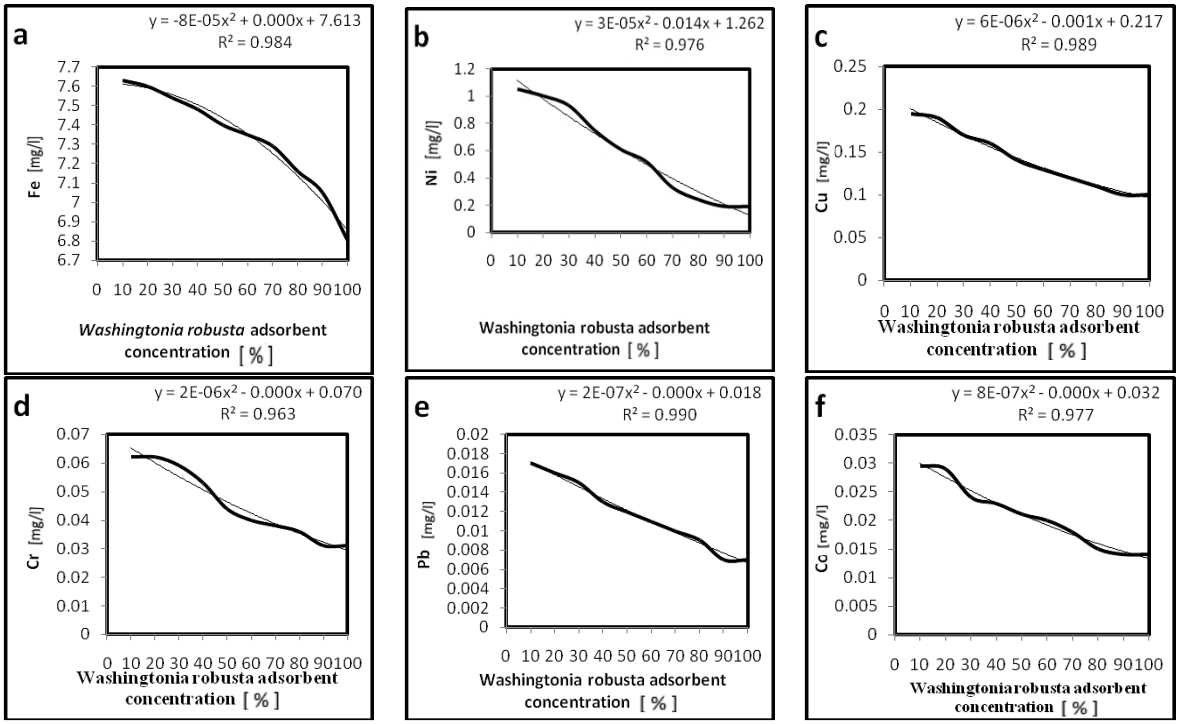


Fig. 2: Chemical composition of industrial water as a result of adsorption using Washingtonia robusta extracts at various doses.

The results show that increasing the adsorbent dosages significantly improves the effectiveness of HMs removal by PF-WR. The percentage removal of iron ions by PF-WR improved from 5.36 % to 64.29 % as adsorbent dosages were raised from 10 % to 100 %. When the adsorbent dose was elevated from 10 % to 100 % mg/l, the percentage of Ni ions recovered by PF-WR increased from 6.68 % to 90.35 %. The impact of adsorbent concentration on another HM adsorption was also investigated. The percentage removal of other ions by the adsorbent increased sharply when the adsorbent dose increased. The maximum percentage removal of Cu, Cr, Pb, Co, Al, and Na was 61.75 %, 62.6 %, 75 %, 60 %, 34.09 %, and 10.09 %, respectively, using the higher adsorbent

dosage. The results indicated that there is an insignificant difference between the removal percentage of some heavy metals like iron, chromium, cobalt, aluminium, and sodium from 80 % to 100 % adsorbent percentage. At 80 % mg/l adsorbent dose, the adsorbent surface is saturated with these metals, and after that, the amount of adsorption reduces [20].

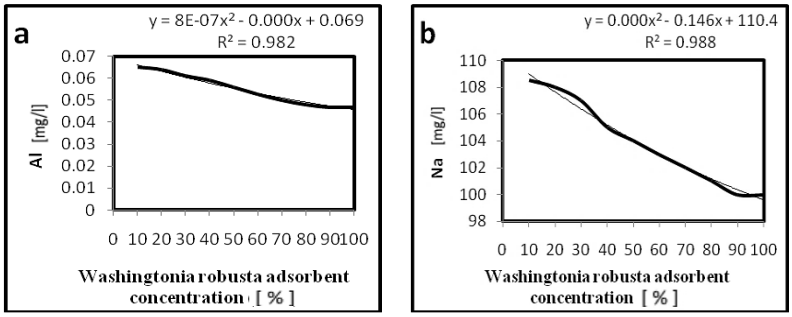


Fig. 3: Aluminium and sodium concentration of industrial water using PF-WR extracts as an adsorbate at various doses.

Previous studies indicated that palm family plants are the best biosorbents for the removal of HMs from industrial wastewater due to the chemical composition of their leaves [13, 22]. The fibers of date pam are lingo-cellulosic materials, which consist of three vital components: cellulose 40 - 50 %, hemicellulose 20 - 35 %, and lignin 15 - 35 %. The high carbon percentage of lignin gives the palm family plants a greater potential for adsorption [21]. The adsorption process is suitable even when the heavy metals are present in concentrations of less than 1 mg/l [26]. The adsorbent dose is important in the adsorption process. The efficiency of PF-WR to remove HMs increased with increasing the adsorbent dosage.

The rate of decrease in HMs due to the use of plant extract was greater for nickel than for sodium, which had the lowest elemental decrease. Looking at physiochemical parameters, the highest reduction was achieved by BOD, while pH was the lowest parameter that decreased due to plant extract, Fig. 4 a) - d).

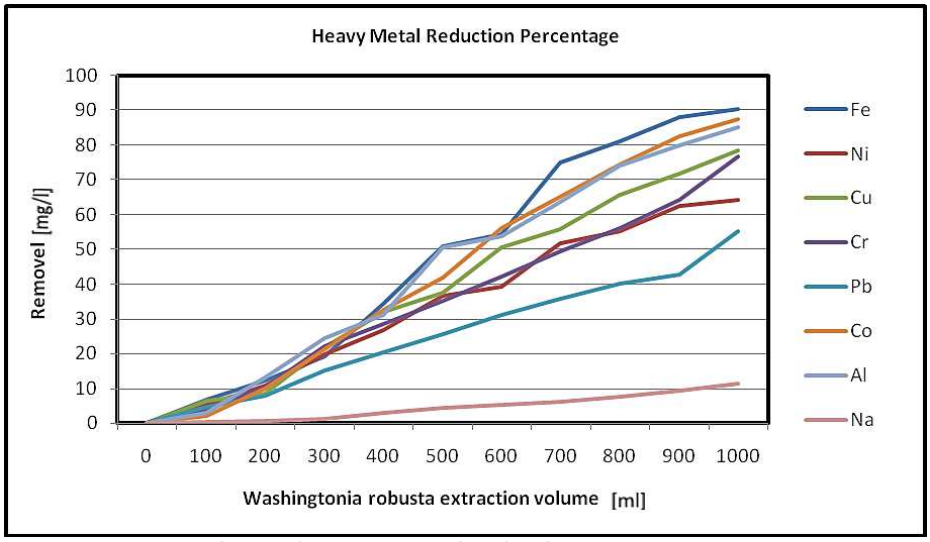


Fig. 4: a) Heavy metal reduction percentage.

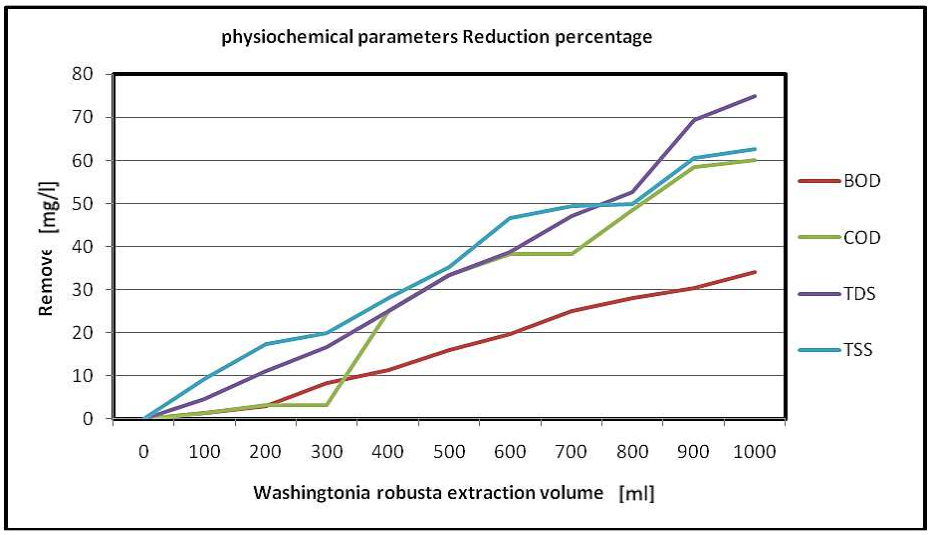


Fig. 4: b) Physiochemical parameters reduction percentage.

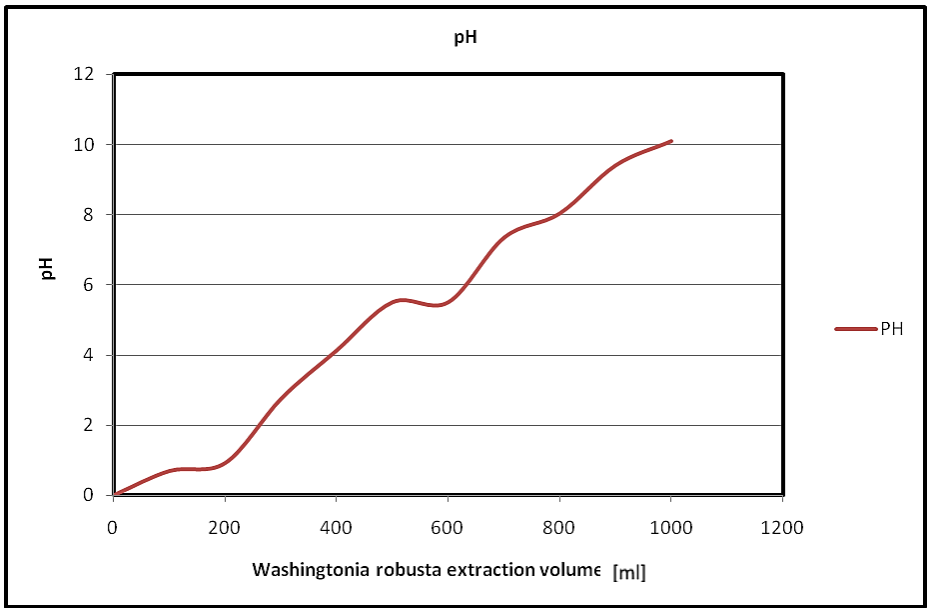


Fig. 4: c). pH changes.

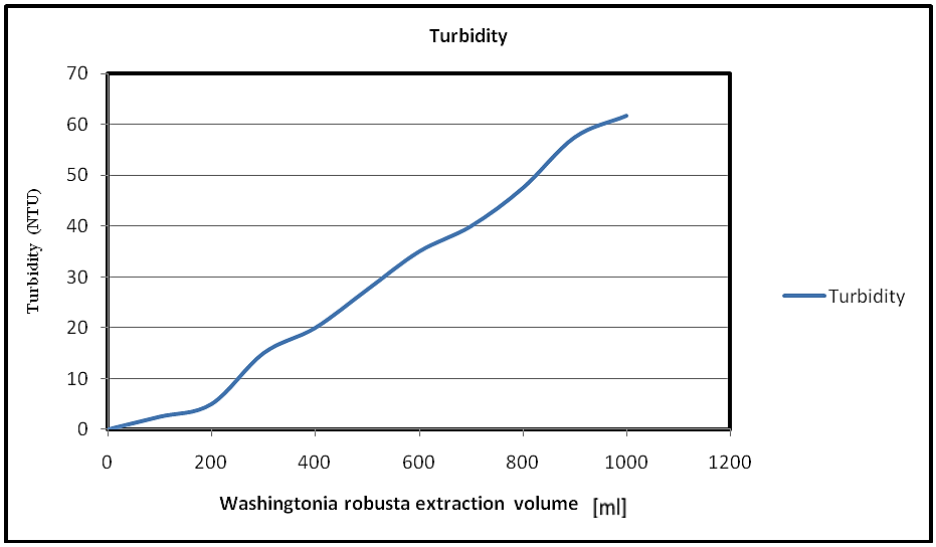


Fig. 4: d) Turbidity changes.

3.6 Difference between the initial and final concentration curve

The difference between the initial concentration curve (industrial water curve) and the final concentration curve (industrial water after mixing with adsorbent) was determined and shown in Fig. 5 for each dosage of adsorbent.

The difference between the two graphs shows the changes in industrial water caused by PF-WR. The area between the two curves grows as the PF-WR effectiveness in purifying industrial water increases. As the adsorbent dose increases from 10 % to 100 %, the percent difference between the initial concentration curve and the final concentration curve increases linearly from 3.06 % to 76.4 %.

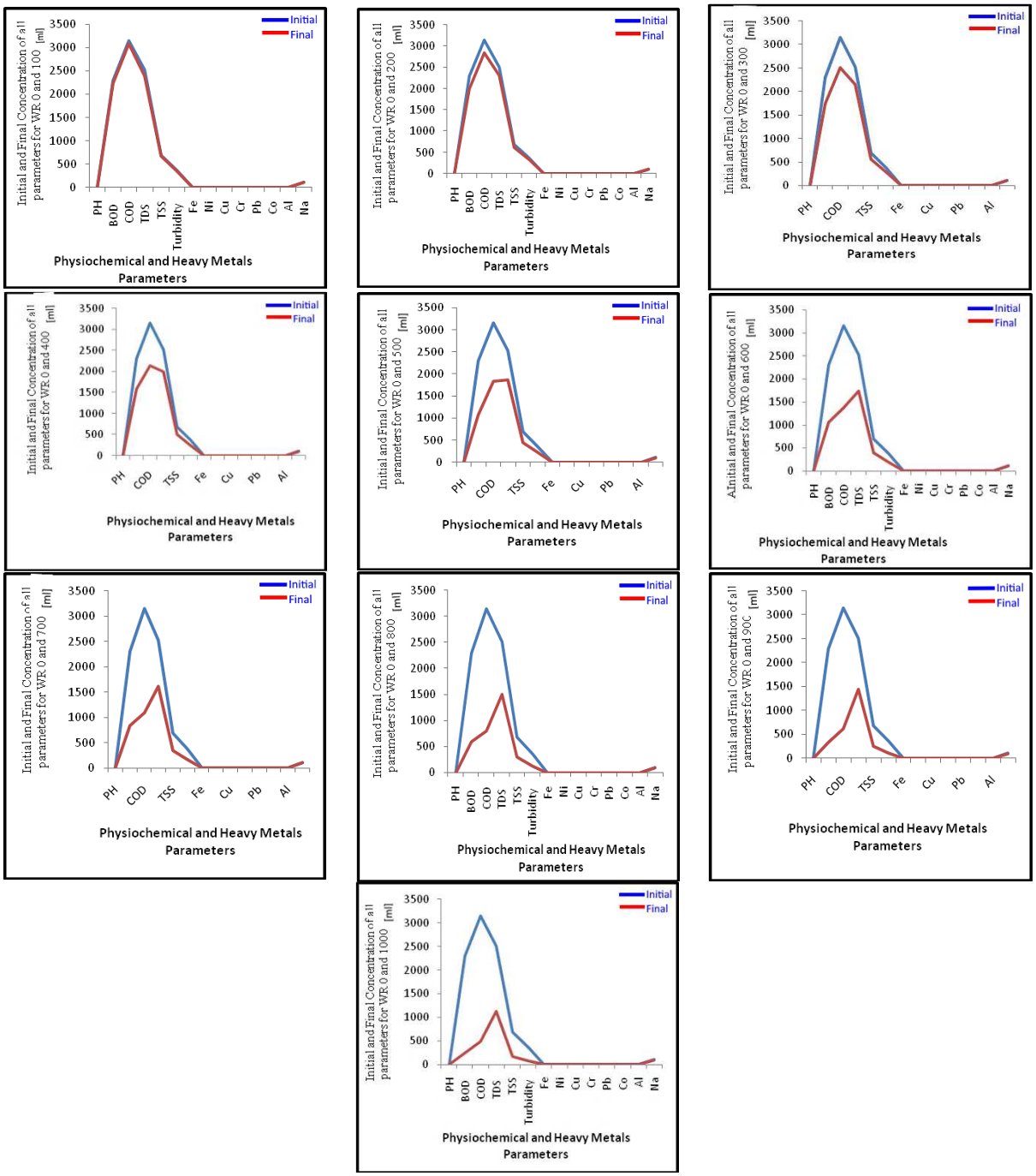


Fig. 5: Difference between initial and final concentration curve taking into consideration all parameters measured in industrial water using PF-WR extracts as adsorbent at various doses.

According to the results, PF-WR has the ability to reduce heavy metals as well as the physicochemical parameter. This result is agreed with different studies: The dependence of the percentage adsorption on pH was reported by El Nemr et al. [22], and they concluded that the

percentage removal of chromium ions increased as the pH value decreased and the optimum pH value was 1. Also, Anah and Astrini [23] investigated the effect of pH on the Cr(VI) adsorption, they performed that the optimum pH for adsorption of Cr(VI) ranged from 1 to 3.

The adsorbent dose has a significant influence on the percentage of adsorption. Results can be explained by the fact that more surface area is available for adsorption to occur, and the number of available active adsorption sites increases by increasing the adsorbent dose. When the adsorption dosage increases, the amount of active surface sites available for adsorption also increases [24]. The same results were obtained by Baby et al [25]. They employed the palm oil kernel shell as an adsorbent and investigated several parameters of adsorption. One of these parameters was the adsorbent dosage. Their investigation study revealed that as the adsorbent dosage increased, the heavy metals and percentage of adsorption were increased.

The number of sites available for adsorption of ions is determined by the concentration of adsorbent used. In this study, the effectiveness of the adsorbent to remove heavy metals increased due to an increase in the active surface suitable for adsorption at higher adsorbent concentrations [19, 25].

4 Conclusion

Palm fronds - Washingtonia Robusta exhibited good adsorption potential for removing heavy metals and physicochemical parameters at low concentrations from the pharmaceutical wastewater. This method was able to get rid of the heavy metals found in industrial water within the limit allowed for agriculture in order to reach the standard of water for drinking purposes. As adsorbent doses were increased from 10 % to 100 %, the removal efficiency of Fe and Ni ions by PF-WR increased from 5.36 % to 64.29 % and from 6.68 % to 90.35 %, respectively. When the adsorbent dosage was raised, the % removal of other ions by the adsorbent rose dramatically. while the other HMs. Using the increased adsorbent dose, the highest removal efficiency of Cu, Cr, Pb, Co, Al, and Na was 61.75 %, 62.6 %, 75 %, 60.60 %, 34.09 %, and 10.09 %, respectively. The COD and BOD, turbidity reduction was 2.91 % to 85.26 %, 2.09 % to 87.41 %, and 4 % to 78 %, respectively. With the increase of the amount of absorbent concentration. These results reveal that removing HMs content using PF-WR would be highly important in environmental applications, and they may be regarded as a very suitable safe usage of PF-WR in agricultural and soil amendment.

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