

Utilization of organic waste as bio-adsorbent for the removal of heavy metals in waste water samples

Kaunain Ayaz^{1*}, Ghazala Yaqub¹, Muhammad¹ and Zishan Ahmad²

¹Department of Environmental Sciences, Kinnaird College for Women, Lahore, Pakistan

²Department of Veterinary Pathology, Faculty of Veterinary and Animal Sciences, PMAS Arid Agriculture University Rawalpindi

Abstract

Heavy metals are elements that are naturally present in the Earth's crust in different concentrations. Anthropogenic activities have disrupted the natural cycle and balance of certain heavy metals causing heavy metal contamination. This research was conducted to study the efficiency of different bioadsorbents viz. amaltas leaves, bar leaves, coconut husk, cow dung, jamun leaves, olive stone, maize cob, neem leaves and date stone for the removal of heavy metals from aqueous solutions as well as industrial effluents prior to optimization of conditions and various parameters. The bio adsorbents were pretreated and chemically activated. The samples were then analyzed using the atomic absorption spectrophotometer version 210VGP to check the efficiency and adsorption. The presence of metals was highest in the paper industry effluent followed by the textile industry and the car wash effluent. The results demonstrated that several low-cost agricultural and industrial waste materials exhibited substantial potential for heavy-metal removal, with almond husk and olive stone achieving consistently high removal efficiencies of 80–90% across the tested metals. Industry-specific adsorbents also showed promising performance, including 90% Cr⁶⁺ removal by maize cob from textile effluent, 85% Zn²⁺ removal by cow dung from paper-industry wastewater, and 76.2% Cr⁶⁺ removal by cow dung from car-wash effluent. Overall, the findings highlight the potential of waste-derived bio-adsorbents as economical and environmentally sustainable alternatives for heavy-metal remediation.

Keywords: Heavy metals; Bioadsorbents; Cow dung; Industrial effluents; Maize cob

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Introduction

Rapid industrialization and the extensive use of metals and chemicals in manufacturing processes have resulted in the discharge of large volumes of wastewater containing high concentrations of toxic heavy metals. These pollutants pose a serious threat to ecosystems and human health due to their persistence, non-biodegradability, and potential for bioaccumulation and biomagnification (Ahluwalia and Goyal, 2007; Wan Ngah and Hanafiah, 2008). Besides industrial activities, processes such as mining, smelting, electroplating, metal extraction, and paint manufacturing are also major contributors to heavy metal contamination. Heavy metals are of significant environmental concern

even at trace levels, as they are toxic, carcinogenic, and mutagenic in nature. Exposure to these metals has been associated with various adverse health outcomes, including cancers, neurological disorders, organ failure, developmental issues, and damage to the circulatory and gastrointestinal systems (Gautam *et al.* 2012; Lee *et al.* 2012; Nagajyoti *et al.* 2010; Jain *et al.* 2016). The contamination of water bodies with heavy metals primarily stems from untreated industrial effluents originating from sectors such as textiles, tanneries, paper and pulp, fertilizers, mining, and battery manufacturing (Gunatilake, 2015; Jain *et al.* 2016). The degree of toxicity

*Corresponding author's e-mail: ghazalayaqub147@gmail.com

and environmental impact of heavy metals depends on several factors, including the metal type, concentration, pH of the effluent, solubility, and wastewater characteristics.

Conventional methods for heavy metal removal include ion exchange, membrane filtration, coagulation, electrochemical treatments, and chemical precipitation. While effective, these methods are often limited by high operational costs, complex designs, the generation of secondary pollutants (such as sludge), and selective removal efficiencies (Tripathi and Ranjan 2015). In contrast, adsorption techniques, particularly those utilizing low-cost and sustainable adsorbents, have gained prominence due to their simplicity, cost-effectiveness, and adaptability to various contaminants. Among various treatment strategies, bio-adsorption has emerged as a promising alternative due to its eco-friendliness and economic viability. This method involves the use of biological materials such as agricultural residues, forestry by-products, algae, and other forms of biomass to adsorb heavy metals from contaminated water. Bio-adsorbents including rice husk, coconut husk, maize cob, sawdust, peat, fly ash, and bentonite have been extensively explored for their metal binding capacities (Nagajyoti *et al.* 2010; Tripathi and Ranjan 2015; Shamim *et al.* 2016; Othman *et al.* 2013). The efficiency of bio-adsorption is influenced by several parameters such as pH, contact time, adsorbent dosage, temperature, and initial metal concentration. Optimization of these variables is essential for maximizing removal efficiency (Lee *et al.* 2012). Recent studies have continued to demonstrate the potential of agricultural wastes as sustainable and low cost adsorbents for removal of heavy metals from contaminated water. A recent comprehensive review reported that agricultural residues such as nutshells, stones, husks, seed coats, and agroforestry wastes can effectively adsorb different heavy metals, including Cu, Pb, Cd, and Ni, while simultaneously contributing to waste valorization and reducing treatment costs. Similarly, recent research has emphasized the development of green sorbents from locally available agricultural wastes, highlighting their adsorption efficiency, environmental compatibility, regeneration potential, and prospects for wastewater treatment applications. These findings support the continued investigation of readily available plant-based wastes as alternative adsorbents, particularly for practical treatment of industrial wastewater. Given the growing demand for cost-effective and environmentally sustainable solutions to

heavy metal contamination, this study aims to investigate the adsorption potential of several low-cost bioadsorbents—including coconut husk, bar leaves, and maize cob—derived from agricultural and plant-based wastes. These materials, often discarded as waste, offer a dual benefit of waste valorization and effective water treatment.

Materials and methods

Collection of data

The first stage of research includes the collection of primary and secondary data. The secondary data collection was carried out through a number of means such as published articles, books, journals, technical reports, and print media including newspapers. Primary data consisted of collecting waste water samples from industrial discharge streams. All relevant material was thoroughly examined to achieve the objectives of this study. All experiments were conducted in triplicate to ensure the reliability and reproducibility of the results.

Sample collection

Various types of organic wastes which are often discarded as waste were collected such as oat husk, olive stones, corn cobs, tree leaves etc. The samples were collected, thoroughly dried and ground.

Sample list

The following were used as bioadsorbents:

- | | |
|----------------|--------------------|
| ➤ Almond husk | ➤ Amaltas leaves |
| ➤ Bar leaves | ➤ Chickoo leaves |
| ➤ Coconut husk | ➤ Coconut shell |
| ➤ Cow dung | ➤ Date stones |
| ➤ Jamun leaves | ➤ Maize (corn) cob |
| ➤ Neem barks | ➤ Neem leaves |
| ➤ Oat husk | ➤ Olive stone |
| ➤ Peanut husk | |

Sample preparation

Fresh organic samples were obtained, washed, sundried, air dried and finally crushed using an electrical grinder. The ground material was then sieved through a mesh size of

110-112 μm to attain fine fibers. They were then stored and labeled in zip lock bags for further processing.

The samples were treated with a solution of 0.3M HNO_3 for 24 hours, followed by washing with deionized water till pH 7.2 was achieved. The samples were then oven dried at 60°C. Once prepared, the samples were stored in a desiccator.

Chemicals used

The following chemicals were used:

HNO_3 , Deionized water, Copper sulfate, Potassium dichromate, Zinc sulfate, Nickel, HCL, NaOH

Preparation of stock solutions

Metal salts were used for the preparation of stock solutions, for each metal 1M solutions were made respective to their molecular weight by dissolving the salt in 1000 ml of distilled water. The standard solution for each metal was prepared in a standard flask. A range of solutions was made with distinct intervals such as 0.1 ppm, 0.2 ppm, 0.4 ppm so on and so forth. The salts used for stock solutions are as follows:

Chromium

Metal salt: Potassium dichromate
Chemical formula: $\text{K}_2\text{Cr}_2\text{O}_7$
Molecular weight: 294.18 g/mol

Zinc

Metal salt: Zinc sulfate heptahydrate
Chemical formula: $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$
Molecular weight: 287.54 g/mol

Copper

Metal salt: Copper(II) sulfate pentahydrate
Chemical formula: $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$
Molecular weight: 249.68 g/mol

Manganese

Metal salt: Manganese(II) sulfate pentahydrate
Chemical formula: $\text{MnSO}_4 \cdot 5\text{H}_2\text{O}$
Molecular weight: 241.08 g/mol

Nickel

Metal Salt: Nickel(II) Chloride Hexahydrate
Chemical Formula: $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$
Molecular Weight: 237.69 g/mol

Formula

$$\frac{X}{Y} \times 1000 = mg$$

X= Molecular weight of salt

Y= Atomic weight of metal

Batch sorption experiments

The sorption studies were completed at room temperature utilizing a funnel shaped flask, containing 50ml of the test arrangement, with known metal ion concentration. Before blending with adsorbent, the pH was changed in accordance with 7.0 with NaOH. A known measure of dried pretreated fiber (0.2 g) called a biomass was added into a conical flask containing metal ion solution and firmly secured with cellophane and shaken completely, permitting adequate time for adsorption balance (likewise to be analyzed). The substance of the flask was shifted on Whatman filter paper of 0.42 μm porosity and the supernatants will be analyzed by AAS.

Optimization of conditions

The effects of adsorbents were analyzed and compared with each other in three categories of optimal conditions:

1. The effect of metal ion concentration
2. The effect of dose of adsorbent
3. The effect of contact time

Determination of effect of adsorbent dose

The adsorbents were used in the quantity of 0.2g/8ml, 0.4g/8ml and 0.8g/8ml. After their application with metals, the 0.2gm quantity was chosen for further adsorption study to remove Cu, Cr, Mn, Ni and Zn because of its sorption capacity.

Determination of effect of contact time

A 50 ml solution of the standard Cr^{+2} solution was poured into six different conical flasks and the contact time was allowed to vary from 15 to 60 min with the interval of 20 minutes for each such as 15, 30, 45, and 60 minutes respectively. Along with that the pH conditions were kept

neutral which is pH 7.0. The same process was repeated for the other standard solutions of the remaining metal ions of Cu^{2+} , Zn^{+2} , Mn^{+2} and Ni^{2+} .

Determination of effect of metal ion concentration

A 50 ml solution of 0.1, 0.2, 0.4, 0.6, 0.8 and 1.0 M Cr^{+2} concentrations were prepared and poured into six conical flasks label A, B, C, D, E and F respectively with the pH maintained at 7.0 for all. Then 0.2 g of the pretreated biomass were added to each of the standard metal ion solution of Cr^{+2} and the flasks tightly covered with cellophane and shaken vigorously for 1 h. The same process was repeated for various standard solutions of the other metal ions of Cu^{2+} , Zn^{+2} , Mn^{+2} and Ni^{2+} .

Determination of effect of pH

All the bioadsorbents were treated to obtain neutral pH conditions which is pH 7.0. Then 0.2 g of the pretreated biomass was added to the standard metal ion solutions of Cr^{+2} and the flasks which were tightly covered with cellophane and then was shaken vigorously for 1 h. The suspensions made will be Cu^{2+} , Zn^{+2} , Mn^{+2} and Ni^{2+} .

Atomic absorption spectrophotometer (AAS)

The presence of metal ions such as Cu^{2+} , Cr^{2+} , Zn^{+2} , Mn^{+2} and Ni^{2+} content in each experiment was determined using an Atomic Absorption Spectrophotometer. Analytical grade standards were used to calibrate the instrument, which did periodical checking throughout the analysis for instrument's response.

Application of adsorbents on industrial waste water

Waste water samples from different industries were allowed to react with adsorbents one by one. The waste water was taken from the industries that produce highly concentrated heavy metal waste water. The industries included car wash, textile and paper. All industries were Lahore based. The heavy metals concentrations were detected first and the adsorption experiment was applied to remove the heavy metals present in the samples. The overall performance of these adsorbents were checked and analyzed and the results were deduced to check the practicability of the adsorbents in industries.

The waste water samples taken from car wash, textile and paper industry were allowed to be treated with the adsorbents under optimized conditions. The samples were digested and then adjusted to specific pH and the adsorbent's doses were set between 0.1g and 1.0 g with the contact time between 15 and 60 minutes. The samples were then analyzed on Atomic

Adsorption Spectrophotometer to check the concentrations of heavy metals before and after the treatment process.

Digestion process of waste water samples

The buffer solutions of H_2O_2 and HNO_3 solutions were made of 50 ml quantity. Each waste water sample was digested one by one. An industrial waste water sample was taken in a round bottom flask in 3 ml quantity and further the buffer solutions of 5ml H_2O_2 (mixture of 30 ml H_2O_2 + 70 ml H_2O) and 5 ml HNO_3 (mixture of 50 ml H_2O + 50 ml HNO_3) were added in 5 ml quantity respectively. This mixture in round bottom flask was adjusted inside a beaker full of silica gel and the flask will be set straight with the help of Styrofoam sheet adjusted around the round bottom flask. Along with this adjustment inside the beaker, the round bottom flask's opening will be set closed with Styrofoam sheet. The mixture will be heated in a microwave oven for 40 seconds with the 10 seconds interval in between the heating process to prevent the explosion of mixture due to heat. The mixture was then filtered in a volumetric flask and the flask was then filled with distilled water.

Chemicals used

➤Distilled water ➤Hydrogen peroxide ➤Nitric acid

Calculation of metal ions i.e. adsorption capacity of adsorbents

The metal uptake was calculated by a simple concentration difference method, while the adsorption capacity that is; amount of metal ions (mg/g) adsorbed per g (dry mass) of biomass was calculated using equation (1).

$$Q = \frac{V(C_i - C_f)}{W \times 1000} \dots\dots\dots (1)$$

Where Q is the mass (g) of metal ion adsorbed per g of sample (biomass), C_i is the initial metal ion concentration mg/l, C_f/C_r is the residual metal ion concentration mg/l at equilibrium, W is the weight of biomass in reaction mixture and V is the volume of metal ions solution used (ml).

Percentage removal of metal ions

Percentage removal of metal ions can also be computed using equation (2).

$$\%R = \frac{C_i - C_f}{C_i} \times 100 \dots\dots\dots (2)$$

Where % R is the Percentage removal, C_i is the Initial metal ion concentration and C_f is the Final metal ion concentration.

Results and discussion

The heavy metals present in water originated from the effluent of several industries. Aqueous water streams have become a major threat to flora and fauna as well as other receiving water bodies due to the presence of heavy metals (Taty *et al.* 2005). Although copper is amongst the essential trace elements for humans, animals and microorganisms, WHO world health organization recommends a maximum concentration of 1.5 mg/L of copper II in drinking water (Tumin *et al.* 2008). A concentration exceeding this limit will cause toxicological effects on humans causing ulcers, hypertension, brain damage and even death (Liu *et al.* 2008). In case of microorganisms, copper toxicity disrupts the integrity of the plasma membrane. Various industries including metal cleaning, paper board mills, wood pulp production and chemical industries are mostly responsible for the release of copper II in the form of industrial effluent leading to contamination of water resources contributing to health effects to humans and microorganisms (Zhang *et al.* 2014). In order to protect the water resources and to recover copper removal techniques need to be adopted such as ion exchange, chemical precipitation, solvent extraction and membrane separation along with several others (Iqbal *et al.* 2010). All of which are highly costly and not as effective whereas on the other hand adsorption has proven to be an effective as well as an active separation and purification technique to be utilized in the industries for the treatment of water and wastewater (Al-Ashesh *et al.* 2000). The benefit of adsorption is the clean and sludge free operation however it has high regeneration

and manufacturing costs (Otero *et al.* 2013). The need is to discover a low cost adsorbent with high efficiency. Over the years much research has been conducted to study the application of rice husk, peanut husk, sawdust and other agricultural waste products due to their abundance, effectiveness and low cost along with the advantages of being environmentally friendly, high adsorption capacity of heavy metals (Ali *et al.* 2016). The conditions were optimized to determine the most suitable conditions for maximum removal with respect to dosage, contact time and pH. Table I shows the conditions of chickoo leaves for all heavy metals for the removal of chromium 83% efficiency was recorded whereas chickoo leaves were most efficient in the removal of copper up to 90% removal however, chickoo leaves proved to be the least efficient in the removal of nickel and manganese at 75% removal. Table XVI shows that 90% removal of copper was achieved at a contact time of 45 mins whereas chromium was removed in at a contact time of only 15 mins. A similar result was recorded by the application of peanut husk in Table II and Table XVI as an adsorbent where it proved least effective in the removal of nickel at only 55% with a contact time of 30 mins, followed by manganese at 67% at a time duration of 15 mins whereas peanut husk proved most effective in the removal of chromium at 90% at a time duration of 60 mins (1 hour).

On the other hand almond husk (Table III and Table XVIII) and olive stone (Table V and Table XX) had an overall high removal rate for chromium, copper, manganese, nickel and zinc ranging from 80-90% after time duration of 60 mins for each metal. The conditions

Table I. Optimal conditions for removal of heavy metals using chickoo leaves

Adsorbent: chickoo leaves			
Conc. before treatment (Ci)	Conc. after treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)
0.6	0.1	Chromium (Cr) 0.0625	83
1	0.1	Copper (Cu) 0.1125	90
0.4	0.1	Manganese (Mn) 0.0375	75
0.4	0.1	Nickel(Ni) 0.0375	75
0.8	0.1	Zinc (Zn) 0.0875	87.5

for date stone are presented in Table IV and Table XIX where chromium was removed with 83% efficiency after a contact time of 60 mins (1 hour). On the contrary, date stone had the lowest efficiency in the removal of copper at only 67% even after 1 hour of contact time and highest for zinc at a contact time of 45 minutes. Making date stone a suitable adsorbent for the removal of zinc contaminated wastewaters. The conditions for smaltas leaves are presented in Table VI and XXI with highest removal rates of copper of 90% at 60 mins and lowest removal rates for manganese despite being in contact for 1 hour.

Textile industry

The increase in population has caused an increase in the demand for clothing and apparel, hence in order to meet the demands of the buyers and the change in fashion

trends the manufactures are producing greater amounts of products for many countries such as Bangladesh, India and Sri Lanka the textile sector is a major contributor towards the national GDP (Gross Domestic Product). Although it may have a positive impact on the economy of the country it also comes with negative environmental impacts such as environmental pollution especially wastewater streams. Textile industry is labeled as the worst pollution offenders and major contributors towards pollution using more than 2000 chemicals and 7000 different types of dyes. Along with releasing great amounts of waste water textile mills are also responsible for heat generation through their processing the effluents released is fully saturated with dyes, detergents, bleaches and several other hazardous chemicals. Moreover, the effluent is discharged into the environment and surrounding water bodies with improper treatment allowing it to seep into the ground

Table II. Optimal conditions for removal of heavy metals using peanut husk

Adsorbent: Peanut husk				
Conc. before treatment (Ci)	Conc. after treatment (Cf)		Adsorption capacity (A.C) (g)	Percentage removal (%)
		Chromium (Cr)		
0.1	0.01		0.01125	90
		Copper (Cu)		
0.6	0.1		0.0625	83
		Manganese (Mn)		
0.6	0.2		0.05	67
		Nickel(Ni)		
0.2	0.09		0.01375	55
		Zinc (Zn)		
0.8	0.1		0.0875	88

Table III. Optimal conditions for removal of heavy metals using almond husk

Adsorbent: Almond husk				
Conc. before treatment (Ci)	Conc. after treatment (Cf)		Adsorption capacity (A.C) (g)	Percentage removal (%)
		Chromium (Cr)		
1	0.2		0.1	80
		Copper (Cu)		
1	0.1		0.1125	90
		Manganese (Mn)		
0.8	0.1		0.0875	88
		Nickel(Ni)		
0.8	0.1		0.0875	88
		Zinc (Zn)		
1	0.1		0.1125	90

Table IV. Optimal conditions for removal of heavy metals using date stone

Adsorbent: Date stone			
Conc. before treatment (Ci)	Conc. after treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)
0.6	0.1	Chromium (Cr) 0.0625	83
0.6	0.2	Copper (Cu) 0.05	67
0.4	0.1	Manganese (Mn) 0.0375	75
0.4	0.1	Nickel(Ni) 0.0375	75
1	0.1	Zinc (Zn) 0.1125	90

Table V. Optimal conditions for removal of heavy metals using olive stone

Adsorbent: Olive stone			
Conc. before treatment (Ci)	Conc. after treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)
1	0.2	Chromium (Cr) 0.1	80
1	0.1	Copper (Cu) 0.1125	90
0.8	0.1	Manganese (Mn) 0.875	87
0.6	0.1	Nickel(Ni) 0.0625	83
1	0.1	Zinc (Zn) 0.1125	90

Table VI. Optimal conditions for removal of heavy metals using amaltas leaves

Adsorbent: Amaltas leaves			
Conc. before treatment (Ci)	Conc. after treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)
0.2	0.1	Chromium (Cr) 0.0125	50
1	0.1	Copper (Cu) 0.1125	90
0.8	0.3	Manganese (Mn) 0.0625	63
0.8	0.1	Nickel(Ni) 0.0875	88
0.2	0.1	Zinc (Zn) 0.0125	50

reaching the water table, polluting rivers, streams, aquifers and other water bodies along with soil as well leading to heavy metal contamination. Heavy metals associated with textile mills include cadmium, copper, chromium and lead which are used in the production of dyes and pigments (Halimoon and Yin 2010).

Treatment of textile effluents is pivotal to ensure the safety of human, living organisms and the environment on a whole. Hence, several agricultural and industrial waste bio adsorbents were applied to textile wastewater to determine the most suitable bioadsorbent for textile industries, the trend observed according to Table XXII for the adsorption of zinc (Zn) in textile effluent was jamun leaves > date stone > amaltas leaves > neem leaves > bar leaves > coconut husk. At a constant

adsorbent dose of 0.2 g and time duration varying from 15 minutes to 60 minutes. the maximum removal was obtained with a dose of 0.2 g jamun leaves and a contact time of 15 minutes to achieve a removal efficiency of 60% where amaltas leaves worked best at 60 minutes and neem leaves at 45 minutes.

Table XXIII shows the adsorption of chromium (Cr) revealing a trend maize cob > cow dung > neem leaves > coconut husk > amaltas leaves > jamun leaves > olive stone > date stone > bar leaves with maize cob being the most efficient in removing chromium with 90% removal efficiency with a dose of only 0.2 g and a contact time of 45 minutes where bar leaves were the least effective with a time duration of 60 minutes and a removal rate of 45%.

Table VII. Optimal Conditions for Removal of Heavy Metals using Bar Leaves

Adsorbent: Bar Leaves			
Conc. Before Treatment (Ci)	Conc. After Treatment (Cf)	Adsorption Capacity (A.C) (g)	Percentage Removal (%)
Chromium (Cr)			
0.4	0.03	0.037	92.5
Copper (Cu)			
0.2	0.005	0.0195	97.5
Manganese (Mn)			
0.1	0	0.1	100
Nickel(Ni)			
0.3	0.01	0.029	96.7
Zinc (Zn)			
0.2	0.1	0.01	50

Table VIII. Optimal Conditions for Removal of Heavy Metals using Coconut Husk

Adsorbent: Coconut Husk			
Conc. Before Treatment (Ci)	Conc. After Treatment (Cf)	Adsorption Capacity (A.C) (g)	Percentage Removal (%)
Chromium (Cr)			
0.6	0.25	0.035	58.3
Copper (Cu)			
0.2	0.115	0.0085	42.5
Manganese (Mn)			
0.4	0.274	0.0126	31.5
Nickel(Ni)			
0.1	0	0.01	100
Zinc (Zn)			
0.3	0.102	0.0198	66

Table IX. Optimal Conditions for Removal of Heavy Metals using Coconut Shell

Adsorbent: Coconut Shell				
Conc. Before Treatment (Ci)	Conc. After Treatment (Cf)	Adsorption Capacity (A.C) (g)	Percentage Removal (%)	
0.1	0.0125	Chromium (Cr) 0.00875	87.5	
0.2	0	Copper (Cu) 0.02	100	
0.6	0.2	Manganese (Mn) 0.04	66.7	
0.1	0.002	Nickel(Ni) 0.0098	98	
0.4	0.1	Zinc (Zn) 0.003	75	

Table X. Optimal Conditions for Removal of Heavy Metals using Cow Dung

Adsorbent: Cow Dung				
Conc. Before Treatment (Ci)	Conc. After Treatment (Cf)	Adsorption Capacity (A.C) (g)	Percentage Removal (%)	
0.3	0	Chromium (Cr) 0.003	100	
0.2	0.001	Copper (Cu) 0.0199	95	
0.4	0.002	Manganese (Mn) 0.0398	99.5	
0.1	0.04	Nickel(Ni) 0.006	60	
0.3	0.1	Zinc (Zn) 0.002	66.7	

Table XI. Optimal Conditions for Removal of Heavy Metals using Jamun Leaves

Adsorbent: Jamun Leaves				
Conc. Before Treatment (Ci)	Conc. After Treatment (Cf)	Adsorption Capacity (A.C) (g)	Percentage Removal (%)	
0.1	0	Chromium (Cr) 0	0	
0.6	0.4	Copper (Cu) 0.02	33.3	
0.2	0.05	Manganese (Mn) 0.015	75	
0.3	0.1	Nickel(Ni) 0.002	66.6	
0.3	0.2	Zinc (Zn) 0.001	33.3	

Paper industry

The rapid increase in industries and literacy had caused an up rise in the demand and usage of paper, wood pulping and paper production generates considerable concentrations of pollutants contributing to high levels of Biological Oxygen Demand (BOD) and Chemical Oxygen Demand (COD), toxicity, Total Suspended Solids and color. The pulp and paper is high on water consumption nearly 20-60,000 gallons for each ton being produced. The discharged waste water leads to the formation of scum, the growth of slime, loss of scenic beauty also leading to an increase in toxic components in the water body resulting in the death of marine and aquatic living organisms (Pokhrel and Viraraghavan 2004).

Table XXIV shows the trend of removal efficiency of zinc as cow dung > coconut husk > maize cob > olive stone >

amaltas leaves > bar leaves > jamun leaves > neem leaves > date stone. Cow dung being the most efficient in the removal of zinc from paper industry wastewater with the removal efficiency of 85% after a contact time of only 15 minutes and a dose of 0.2 g where date stone was seen to have the lowest adsorption and removal rate of only 39% despite a contact time of 45 minutes. However, in the case of chromium according to Table XXV the trend was much different from the trend of zinc, coconut husk > neem leaves > amaltas leaves > jamun leaves > bar leaves > maize cob > cow dung. In the case of removing zinc cow dung was the most efficient with removal rate of 85% for paper industry waste water on the other hand it was the least efficient for the removal of chromium in the sample with a minor removal rate of 16.7% after 30 minutes contact time.

Table XII. Optimal conditions for removal of heavy metals using maize cob (corn)

Adsorbent: Maize cob (corn)			
Conc. before treatment (Ci)	Conc. after treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)
0.4	0.05	Chromium (Cr) 0.0035	87.5
0.1	0.02	Copper (Cu) 0.008	80
0.1	0	Manganese (Mn) 0.01	100
0.3	0.1	Nickel(Ni) 0.002	66.6
0.2	0.073	Zinc (Zn) 0.0127	63.5

Table XIII. Optimal conditions for removal of heavy metals using neem bark

Adsorbent: Neem bark			
Conc. before treatment (Ci)	Conc. after treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)
0.3	0.1	Chromium (Cr) 0.002	66.6
0.1	0	Copper (Cu) 0.01	100
0.2	0.125	Manganese (Mn) 0.0075	37.5
0.2	0.05	Nickel(Ni) 0.015	75
0.1	0.1	Zinc (Zn) 0	0

Carwash industry

Many studies have been conducted on the removal of heavy metals from the waste water of all types of industries including leather, tanning, sugar, dying, textile and many others but very limited research has been conducted on Car wash waste water. Due to which there is lack of awareness and knowledge about the best management practices for car wash industry. Although many processes and treatment methods have been advised such as coagulation, flocculation, filtration, adsorption and chemical oxidation but very few car wash stations are equipped with treatment systems, mostly the effluent is drained into the main sewerage drain. Car wash effluent contains numerous impurities including dirt, sand, oils, grease stones, detergents, and even

organic matter which if disposed of untreated will eventually reach the nearby water bodies including rivers, lakes and oceans (Lau *et al.* 2013).

Table XXVI shows that only bar leaves were successful in the removal of zinc from carwash waste water with a removal efficiency of 52% with an adsorbent dose of 0.2g and a contact time of 15 minutes where in Table XXVII the trend for the removal of chromium is cow dung > jamun leaves > coconut husk > amaltas leaves > maize cob > bar leaves > olive stone > neem leaves. Cow dung with the highest removal rate of 76.19% in time duration of 30 minutes and neem leaves being the least effective at 30% removal after 60 mins of contact time.

Table XIV. Optimal conditions for removal of heavy metals using neem leaves

Adsorbent: Neem leaves				
Conc. before treatment (Ci)	Conc. after treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)	
		Chromium (Cr)		
0.4	0.1	Copper (Cu)	0.03	75
0.2	0.1	Manganese (Mn)	0.01	50
0.3	0.1	Nickel(Ni)	0.02	66.7
0.6	0.2	Zinc (Zn)	0.04	66.6
0.4	0.1		0.03	75

Table XV. Optimal conditions for removal of heavy metals using oat husk

Adsorbent: Oat Husk				
Conc. before treatment (Ci)	Conc. after treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)	
		Chromium (Cr)		
0.2	0.05	Copper (Cu)	0.015	75
0.3	0.1	Manganese (Mn)	0.02	66.7
0.2	0.1	Nickel(Ni)	0.01	50
0.1	0	Zinc (Zn)	0.01	100
0.3	0.2		0.01	33.3

Table XVI. Effects of varying contact time in the removal of heavy metals using chickoo leaves

Adsorbent: Chickoo Leaves			
Contact Time (CT)	Conc. After Treatment (Cf)	Adsorption Capacity (A.C) (g)	Percentage Removal (%)
Chromium (Cr)			
15	0.1	0.036	90
30	0.1	0.036	90
45	0.2	0.032	80
60	0.2	0.032	80
Copper (Cu)			
15	0.2	0.032	80
30	0.2	0.032	80
45	0.1	0.036	90
60	0.1	0.036	90
Manganese (Mn)			
15	0.1	0.036	90
30	0.2	0.032	80
45	0.1	0.036	90
60	0.2	0.032	80
Nickel(Ni)			
15	0.2	0.32	80
30	0.1	0.36	90
45	0.1	0.36	90
60	0.1	0.35	90
Zinc (Zn)			
15	0.2	0.032	80
30	0.2	0.032	80
45	0.1	0.036	90
60	0.1	0.036	90

Heavy metal contamination and need for low-cost adsorbents

Heavy metals, particularly copper (Cu^{2+}), chromium (Cr^{6+}), nickel (Ni^{2+}), manganese (Mn^{2+}), and zinc (Zn^{2+}), are prominent contaminants in industrial effluents, including those from metal cleaning, textile processing, paper manufacturing, and chemical industries. While copper is an essential trace element, its concentration in drinking water must not exceed 1.5 mg/L, as per WHO guidelines (Tumin *et al.* 2008). Elevated levels pose toxicological risks including

gastrointestinal distress, liver and kidney damage, neurological effects, and mortality (Liu *et al.* 2008). Consequently, cost-effective and sustainable remediation techniques are urgently needed.

Adsorption has emerged as a viable alternative due to its operational simplicity, minimal sludge generation, and environmental compatibility (Al-Ashesh *et al.* 2000). However, the economic feasibility depends on the development and optimization of low-cost, efficient adsorbents derived from agricultural and industrial biomass.

Table XVII. Effects of varying Contact Time in the Removal of Heavy Metals using Peanut Husk

Adsorbent: Peanut Husk			
Contact Time (CT)	Conc. After Treatment (Cf)	Adsorption Capacity (A.C) (g)	Percentage Removal (%)
Chromium (Cr)			
15	0.9	0.004	10
30	0.9	0.004	10
45	0.7	0.012	30
60	0.5	0.02	50
Copper (Cu)			
15	0.8	0.008	20
30	0.7	0.012	30
45	0.7	0.012	30
60	0.1	0.036	90
Manganese (Mn)			
15	0.3	0.028	70
30	0.3	0.028	70
45	0.2	0.032	20
60	0.2	0.032	20
Nickel(Ni)			
15	1	0	0
30	0.6	0.016	40
45	0.8	0.008	20
60	0.1	0.036	90
Zinc (Zn)			
15	0.9	0.004	10
30	0.6	0.016	40
45	0.8	0.004	10
60	0.1	0.028	70

*Evaluation of adsorbents for individual heavy metals**Chickoo leaves*

Optimization studies identified Chickoo leaves as particularly effective for Cu^{2+} removal, achieving 90%

efficiency at a contact time of 45 minutes. Cr^{6+} removal was also notable, with 83% efficiency reached in only 15 minutes. However, performance was comparatively lower for Ni^{2+} and Mn^{2+} , with maximum removal efficiencies of 75% (Table I; Table XVI).

Table XVIII. Effects of varying contact time in the removal of heavy metals using almond husk

Adsorbent: Almond husk				
Contact time (CT)	Conc. After treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)	
15	0.8	Chromium (Cr)	0.008	20
30	0.6		0.016	40
45	0.5		0.02	50
60	0.3		0.028	70
15	0.9	Copper (Cu)	0.004	20
30	0.9		0.004	20
45	0.7		0.012	30
60	0.3		0.028	70
15	0.7	Manganese (Mn)	0.012	30
30	0.7		0.012	30
45	0.7		0.012	30
60	0.7		0.012	30
15	0.4	Nickel(Ni)	0.024	60
30	0.2		0.032	80
45	0.3		0.028	70
60	0.2		0.032	80
15	0.9	Zinc (Zn)	0.004	20
30	0.7		0.012	30
45	0.4		0.024	60
60	0.4		0.024	60

Table XIX. Effects of varying contact time in the removal of heavy metals using date stone

Adsorbent: Date Stone				
Contact time (CT)	Conc. After treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)	
15	0.5	Chromium (Cr)	0.02	50
30	0.4		0.024	60
45	0.4		0.024	60
60	0.3		0.028	70
15	0.8	Copper (Cu)	0.008	20
30	0.2		0.032	80
45	0.1		0.036	90
60	0.1		0.036	90
15	0.9	Manganese (Mn)	0.004	10
30	0.9		0.004	10
45	0.8		0.003	20
60	0.3		0.028	70
15	0.3	Nickel(Ni)	0.028	70
30	0.3		0.028	70
45	0.2		0.032	80
60	0.2		0.032	80
15	0.7	Zinc (Zn)	0.012	30
30	0.3		0.028	70
45	0.3		0.028	70
60	0.4		0.024	60

Peanut husk

Peanut husk displayed highest efficiency for Cr^{6+} (90%) at 60 minutes but performed poorly for Ni^{2+} (55%) and Mn^{2+} (67%) (Table II; Table XVII). These results suggest a selective affinity toward certain metals, especially chromium.

Almond husk and olive stone

Both almond husk and olive stone demonstrated broad-spectrum metal removal efficiencies ranging from 80–90% for all tested heavy metals within 60 minutes of contact time (Tables III, V; Tables XVIII, XX). These findings position them as versatile and highly effective adsorbents for multi-metal wastewater streams.

Table XX. Effects of varying contact time in the removal of heavy metals using olive stone

Adsorbent: Olive stone			
Contact time (CT)	Conc. After treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)
Chromium (Cr)			
15	0.6	0.016	40
30	0.4	0.024	60
45	0.2	0.032	80
60	0.4	0.024	60
Copper (Cu)			
15	0.9	0.004	10
30	0.8	0.008	20
45	0.9	0.004	10
60	0.6	0.016	40
Manganese (Mn)			
15	0.2	0.032	80
30	0.3	0.028	70
45	0.4	0.024	60
60	0.3	0.028	70
Nickel(Ni)			
15	0.3	0.028	70
30	0.1	0.036	90
45	0.1	0.036	90
60	0.1	0.036	90
Zinc (Zn)			
15	0.7	0.012	30
30	0.6	0.016	40
45	0.7	0.012	30
60	0.6	0.016	40

Table XXI. Effects of varying contact time in the removal of heavy metals using amaltas leaves

Adsorbent: Amaltas leaves			
Contact time (CT)	Conc. After treatment (Cf)	Adsorption capacity (A.C) (g)	Percentage removal (%)
Chromium (Cr)			
15	0.9	0.004	10
30	0.8	0.008	20
45	0.7	0.012	30
60	0.6	0.016	40
Copper (Cu)			
15	0.9	0.004	10
30	0.9	0.004	10
45	0.8	0.008	20
60	0.6	0.0016	40
Manganese (Mn)			
15	0.8	0.008	20
30	0.6	0.004	10
45	0.4	0.012	30
60	0.2	0.032	80
Nickel(Ni)			
15	0.4	0.012	30
30	0.4	0.012	30
45	0.6	0.004	10
60	0.8	0.008	20
Zinc (Zn)			
15	0.4	0.012	30
30	0.2	0.032	80
45	0.3	0.028	70
60	0.1	0.036	90

Date stone

Date stone showed maximum affinity for Zn^{2+} , achieving 90% removal in 45 minutes. Cr^{6+} removal was 83% at 60 minutes, while Cu^{2+} was least efficiently adsorbed (67%) under similar conditions (Table IV; Table XIX).

Amaltas leaves

Amaltas leaves demonstrated 90% removal efficiency for Cu^{2+} at 60 minutes but were less effective for Mn^{2+} even after prolonged exposure (Table VI; Table XXI), indicating selectivity dependent on metal ion characteristics.

*Performance in industry-specific effluents**Textile industry effluent*

The textile sector, a major contributor to economic output in South Asia, is also a significant polluter due to the extensive use of dyes, bleaches, and metallic mordants. Zinc and chromium were targeted for removal from textile waste water.

Zinc removal

Maximum Zn^{2+} adsorption (60%) was achieved using jamun leaves within 15 minutes. other effective adsorbents, in decreasing order of performance, included date stone, amaltas leaves, and neem leaves (Table XXII).

Chromium removal

Maize cob exhibited the highest Cr^{6+} removal (90%) within 45 minutes, followed by cow dung and neem leaves (Table XXIII).

Paper industry effluent

Effluent from pulp and paper mills is characterized by high COD, BOD, and metal concentrations.

Zinc removal

Cow dung showed superior removal efficiency (85%) in just 15 minutes. The least effective was date stone (39%) despite longer contact duration (Table XXIV).

Table XXII. Removal of zinc using optimally conditioned bio-adsorbents from textile waste water

ZINC					
Adsorbent: Date stone pH: 7					
Adsorbent dose (g)	CT (Mins)	Ci	Cf	AC (g)	% Removal
	45	0.3	0.125	0.0175	58.33%
Adsorbent: Coconut husk pH: 7					
0.2	15	0.3	0.135	0.0165	55%
	30	0.3	0.275	0.0025	8.33%
	45	0.3	0.295	0.00075	1.66%
Adsorbent: Bar leaves pH: 7					
0.2	15	0.3	0.135	0.0165	55%
	45	0.3	0.260	0.0002	3.4%
Adsorbent: Amaltas leaves pH: 7					
0.2	15	0.3	0.200	0.01	33.33%
	60	0.3	0.130	0.017	56.67%
Adsorbent: Neem leaves pH: 7					
0.2	15	0.3	0.135	0.0165	55%
	30	0.3	0.200	0.01	33.33%
	45	0.3	0.130	0.017	56.67%
	60	0.3	0.220	0.008	26.67%
Adsorbent: Jamun leaves pH: 7					
0.2	15	0.3	0.120	0.018	60%

Chromium removal

Coconut husk was the most effective (82%), while cow dung, although effective for Zn^{2+} , was least efficient for Cr^{6+} removal (16.7%) (Table XXV). This highlights the need to match adsorbent type with specific target metals.

Car wash effluent

Car wash wastewater is an under-studied source of pollution, rich in detergents, oils, metals, and suspended solids.

Zinc removal

Bar leaves showed moderate Zn^{2+} removal efficiency (52%) within 15 minutes (Table XXVI).

Chromium removal

Cow dung again emerged as the most efficient adsorbent for Cr^{6+} , achieving 76.2% removal at 30 minutes. Neem leaves

showed the lowest removal capacity (30%) even after 60 minutes (Table XXVII). The comparative analysis of all adsorbents across different wastewater types suggests:

Best universal adsorbents

Almond husk and olive stone due to consistently high removal rates across all tested metals.

*Industry specific recommendations**Textile effluent*

Jamun leaves for Zn^{2+} , maize cob for Cr^{6+} .

Paper industry

Cow dung for Zn^{2+} , coconut husk for Cr^{6+} .

Car wash waste water

Bar leaves for Zn^{2+} , cow dung for Cr^{6+} .

Table XXIII. Removal of chromium using optimally conditioned bio-adsorbents from textile waste water

CHROMIUM					
Adsorbent: Date stone pH: 7					
Adsorbent dose (g)	CT (Mins)	Ci	Cf	AC (g)	% Rem oval
0.2	45	0.05	0.025	0.0025	50%
Adsorbent: Olive stone pH: 7					
0.2	60	0.05	0.02	0.003	60%
Adsorbent: Coconut husk pH: 7					
	30	0.05	0.03	0.002	40%
	45	0.05	0.01	0.004	80%
Adsorbent: Bar leaves pH: 7					
	15	0.05	0.042	0.008	16%
0.2	60	0.05	0.0295	0.00205	45%
Adsorbent: Amaltas leaves pH: 7					
0.2	15	0.05	0.011	0.001	78%
Adsorbent: Neem leaves pH: 7					
	15	0.05	0.04	0.001	20%
0.2	30	0.05	0.0325	0.00175	30%
	45	0.05	0.02	0.003	60%
	60	0.05	0.01	0.004	80%
Adsorbent: Jamun leaves pH: 7					
	30	0.05	0.013	0.037	74%
0.2	45	0.05	0.025	0.0025	50%
	60	0.05	0.015	0.0035	70%
Adsorbent: Cow dung pH: 7					
0.2	45	0.05	0.1	0.004	80%
Adsorbent: Maize cob pH: 7					
0.2	45	0.05	0.005	0.0045	90%

Table XXIV. Removal of zinc using optimally conditioned bioadsorbents from paper industry waste water

ZINC					
Adsorbent: Date stone pH: 7					
Adsorbent Dose (g)	CT (Mins)	Ci	Cf	AC (g)	% Removal
0.2	45	0.75	0.270	0.048	39%
Adsorbent: Olive stone pH: 7					
0.2	15	0.75	0.124	0.0626	83.46%
Adsorbent: Coconut husk pH: 7					
	15	0.75	0.270	0.048	39%
0.2	30	0.75	0.115	0.0635	84.66%
	45	0.75	0.125	0.0625	83.33%
	60	0.75	0.220	0.053	70.66%

Continued

Adsorbent: Bar Leaves						
0.2	15	0.75	pH: 7	0.210	0.054	72%
	30	0.75		0.140	0.061	81.33%
	45	0.75		0.140	0.061	81.33%
	60	0.75		0.135	0.0615	82%
Adsorbent: Am altas Leaves						
0.2	15	0.75	pH: 7	0.130	0.062	82.66%
Adsorbent: Neem Leaves						
0.2	15	0.75	pH: 7	0.285	0.465	62%
	30	0.75		0.272	0.0478	63.73%
	45	0.75		0.290	0.046	61.33%
	60	0.75		0.278	0.0472	62.93%
Adsorbent: Jamun Leaves						
0.2	30	0.75	pH: 7	0.200	0.055	73.33%
	45	0.75		0.250	0.05	66.67%
	60	0.75		0.280	0.047	62.67%
Adsorbent: Cow Dung						
0.2	15	0.75	pH: 7	0.115	0.0635	84.66%
	30	0.75		0.115	0.0635	84.66%
Adsorbent: Maize Cob						
0.2	45	0.75	pH:7	0.125	0.0625	83.33%

table xxv. Removal of chromium using optimally conditioned bio-adsorbents from paper industry waste water

CHROMIUM					
Adsorbent: Coconut husk pH: 7					
Adsorbent dose (g)	CT (Mins)	Ci	Cf	AC (g)	% Removal
0.2	15	0.9	0.465	0.0435	48.33%
	30	0.9	0.366	0.0534	59.33%
	45	0.9	0.225	0.0675	75%
	60	0.9	0.215	0.0685	76.11%
Adsorbent: Bar Leaves					
0.2	15	0.9	pH: 7a 0.815	0.0085	9.44%
	30	0.9	0.765	0.0135	15%
	45	0.9	0.629	0.0271	30.11%
	60	0.9	0.425	0.0475	52.77%
Adsorbent: Amaltas Leaves					
0.2	15	0.9	pH: 7a 0.350	0.055	61.11%
Adsorbent: Neem Leaves					
0.2	15	0.9	pH: 7a 0.785	0.0115	12.77%
	30	0.9	0.653	0.0247	27.44%
	45	0.9	0.592	0.0308	34.22%
	60	0.9	0.320	0.058	64.44%
Adsorbent: Jamun Leaves					
0.2	30	0.9	pH: 7a 0.435	0.0465	51.67%
	45	0.9	0.360	0.054	60%
	60	0.9	0.385	0.0515	57.2%
Adsorbent: Cow Dung					
0.2	15	0.9	pH: 7a 0.825	0.0075	8.3%
	30	0.9	0.750	0.0015	16.7%
Adsorbent: Maize Cob pH:7					
0.2	45	0.9	pH: 7a 0.650	0.0025	27.78%

table xxvi. Removal of zinc using optimally conditioned bio-adsorbents from car wash waste water

ZINC					
Adsorbent: Bar Leaves pH: 7					
Adsorbent Dose (g)	CT (Mins)	Ci	Cf	AC (g)	% Removal
0.2	15	0.250	0.130	0.012	52%
	30	0.250	0.120	0.013	52%
	45	0.250	0.120	0.013	52%

Table XXVII. Removal of chromium using optimally conditioned bio-adsorbents from car wash waste water

CHROM IUM					
Adsorbent: Amaltas leaves pH: 7					
Adsorbent dose (g)	CT (Mins)	Ci	Cf	AC (g)	% Removal
0.2	30	0.63	0.22	0.041	65.07%
Adsorbent: Olive stone pH: 7					
0.2	45	0.63	0.40	0.023	36.5%
Adsorbent: Coconut husk pH: 7					
0.2	15	0.63	0.42	0.023	33.33%
	30	0.63	0.38	0.025	39.6%
	45	0.63	0.28	0.035	55.5%
	60	0.63	0.20	0.043	68.25%
Adsorbent: Bar leaves pH: 7					
0.2	15	0.63	0.58	0.005	7.93%
	30	0.63	0.60	0.003	4.76%
	45	0.63	0.36	0.027	42.85%
	60	0.63	0.45	0.018	28.57%
Adsorbent: Neem leaves pH: 7					
0.2	15	0.63	0.56	0.007	11.11%
	30	0.63	0.48	0.015	23.80%
	45	0.63	0.59	0.004	6.34%
	60	0.63	0.44	0.019	30.15%
Adsorbent: Jamun leaves pH: 7					
0.2	30	0.63	0.23	0.04	63.49%
	45	0.63	0.21	0.042	66.66%
	60	0.63	0.19	0.044	69.841%
Adsorbent: Cow dung pH: 7					
0.2	15	0.63	0.21	0.042	66.67%
	30	0.63	0.15	0.048	76.19%
Adsorbent: Maize cob pH:7					
0.2	45	0.63	0.23	0.04	63.49%

These findings demonstrate the efficacy of low-cost agricultural wastes as bio-adsorbents for heavy metal remediation in industrial effluents. The variability in adsorption capacity across different metals and effluent types underlines the importance of targeted adsorbent selection based on pollutant profile. Recent studies have confirmed the effectiveness of agricultural waste-derived adsorbents for heavy-metal remediation, with substantial removal of Cu^{2+} , Ni^{2+} , Pb^{2+} , Zn^{2+} and Cr^{6+} reported for different biomass materials. Alsulaili *et al.* (2024) further demonstrated that agricultural residues such as husks, stones and seed coats can provide promising and economical alternatives to conventional adsorbents. In the present study, almond husk and olive stone achieved 80–90% removal across all tested metals within 60 min, indicating broad-spectrum adsorption performance and supporting their potential for treatment of multi-metal industrial effluents.

Conclusion

The current study demonstrates that low-cost agricultural and plant based waste materials can serve as effective bio-adsorbents for the removal of heavy water from industrial waste water. Almond husk and olive stone showed the most consistent performance, achieving 80–90% removal of the tested metals, while several adsorbents exhibited high metal-specific removal efficiencies. Industry specific results demonstrated up to 90% Cr^{6+} removal from textile wastewater, 85% Zn^{2+} removal from paper-industry wastewater, and 76.2% Cr^{6+} removal from car-wash wastewater. Overall, these findings highlight the potential of readily available waste materials as economical, environmentally sustainable, and practical alternatives for heavy-metal remediation.

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