



## COMPARISON STUDY FOR SOME HEAVY METALS REMOVAL BY PLANT RESIDUES FROM ITS AQUEOUS SOLUTIONS

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<b>Received:</b> 11 <sup>th</sup> June 2023 <b>Accepted:</b> 10 <sup>th</sup> July 2023 <b>Published:</b> 14 <sup>th</sup> August 2023	Heavy metals are dangerous pollutants for the environment because they come from different sources to be put into the ecosystem. The adsorption process is one of the modern methods used to treat water contaminated with these metals. In this study parts of dry plant materials were used for Sider ( <i>Zizyophus SP.</i> ) branches, Bamper ( <i>Cordia SP.</i> ) branches, Palm fronds, Eucalyptus branches, and Rice husk ash to test them for the absorption of polluting elements, and they were conducted in the laboratories of the Department of Soil Sciences and Water Resources at the College of Agriculture / University of Basra, Iraq, for the year 2020-2021. Several parameters were used in the study of the adsorption process to show the role of plant material type and ion element type (copper, nickel, cadmium, zinc, and lead), including pH value, different particle diameters and temperature. The results showed an increase in the adsorption of ions by plant residues with high pH values (3,4,5,6,7,8 and 9) for aqueous solutions contaminated with the studied elements. There was a decrease in the removal percentage with increasing particle diameters (less than 200,200-400,400-600,600-800) $\mu$ m. The study also showed that the adsorption of these elements by the used plant residues increases with the increase in temperature.

**Keywords:** heavy metals - plant residues - solution pH - particle diameters - temperature.

### 1. INTRODUCTION:

The toxic and harmful effects of heavy metals on the environment have become known and diagnosed by a number of researchers, and it is considered one of the greater, most complex, and most difficult problems to solve. Pollution results from various sources, the most important of which is the parent material rich in heavy metals or groundwater, as well as due to human activities resulting from the disposal of domestic sewage, industrial liquid waste and various mining operations. Where heavy metals are pollutants capable of causing environmental and health problems in water, soil, various microorganisms and the atmosphere. (Soran *et al.*,2019). Unlike on the contrary of decompose in the environment, these elements are not biodegradable and accumulate in the environment, although some of them, which present in low concentrations, are used in the human diet, but within the permissible limits (Uddin 2017), given that they bioaccumulate and tend to be dangerous (Ajala and Ojoawa,2022), because of its long half-life. If there are no appropriate treatments and appropriate solutions for the disposal of mine waste, factories, mining and sewage, this will affect the ecosystem. It has become necessary to use techniques to remove these pollutants such as chemical precipitation, membrane filtration, activated carbon, extraction solvents, and others (Dixit and Sinha 2021). However, these methods are expensive and require more complex chemicals and techniques that require time, as well as being a source of pollution with secondary environmental materials resulting from purification processes, and need more than one stage to get rid of these pollutants (Obloh *et al.*, 2012).

Recently, the adsorption process using plant residues, or what is known as biosorption, has gained a lot of attention because it is cheap, economically preferred, simple technology, and has advantages in the removal process because it has high

properties, as well as the ease of devising multiple methods for its use when dealing with waste polluted water. In addition to that, it is environmentally friendly and can be reused, its abundance in nature, non-toxicity, and chemical stability, as it makes plant residues absorbent materials for various pollutants (Sang and Si, 2021).

plants chosen in this study are widely spread in arid and semi-arid regions. The *Eucalyptus* is one of the industrially important forest trees because of its rapid growth and tolerance to different environmental conditions, it has spread successfully in most countries of the world, as this plant has different and effective biological effects. The Bamber tree (*Cordia SP.*), belongs to the *Boraginaceae* family (Tahervand, S. and M. Jalali 2017). It is a semi-tropical, evergreen, medium-sized tree with a height of 5-7 m (Malik *et al.*, 2017). In terms of chemical composition and active ingredients,

the Bamber contains alkaloids, which have great pharmacological effects, including analgesics, stimulants, vasodilators and antiparasitics (Yahya, 2003). Sidr tree *Zizyphus SP.* is a small fruit tree ideal for cultivation in arid and semi-arid regions and prefers to grow in alkaline soils (Erhim,2002) Sidr belongs to the *Rhamnaceae* family, belonging to the order *Rhamnales*, which includes about 58 genera and 600 species among trees, shrubs, and climbers. In Iraq, there are four genera comprising 32 species, including the Iraqi species (AI-Rawi ,1982). The palm tree, *Phoenix dactylifera*, belongs to the family *Areca*. Previously, it is a perennial tree, with a thick stem (trunk), and the highest recorded it reached 28.20 m (Sarmiento *et al.*,2023). The tree grows annually about 45 cm, forming about 20-25 fronds on it Iraq ranks first with about 26% of the production of the Arab world among the countries exporting dates. Rice husk is an agricultural waste, which is available in large quantities in rice milling factories. The husk may be used to produce the energy needed to operate some factories. As a result of using it for this purpose, it results in large quantities of ash, so the use of ash has become widespread in industry and of economic importance (Patel *et al.*,1987). This ash comes as a result of the breakdown of the organic materials in the husks upon burning, which causes cellulose and lignin to volatilize, leaving pores in the silica structure (Esmaeili and Eslami ,2019). The study aims to test the efficiency of plant residues in the adsorption of some heavy metals and their ability to reduce the level of environmental pollution from these metals, especially ( $Cd^{+2}$ ,  $Pb^{+2}$ ,  $Zn^{+2}$ ,  $Cu^{+2}$ ,  $Ni^{+2}$ ) and to understand some physicochemical parameters in controlling the process of sequestration of these ions by the studied plant residues, such as the pH of the solution, particle size of the plant residues, and temperature.

**2. MATERIALS AND METHODS:**

The laboratory experiments were conducted in the laboratories of the Department of Soil Sciences and Water Resources / College of Agriculture, University of Basra for the academic year 2020/2021.

Plant residues of Eucalyptus, Sidr(*Zizyphus SP.*),Bampere( *Cordia SP.*) and Palm fronds were collected in different regions in Basra Governorate. These plant residues were washed with distilled water after excluding leaves from them and keeping twigs and small branches. Then these samples were dried at a temperature of 60-70 degrees Celsius. After that, these dry plant parts were ground and kept in plastic containers until used. As for the ashes of Rice husks, they were collected from rice farms in Najaf Governorate, the Amber variety. These husks were washed several times with distilled water, then dried under a temperature of 100 C°, and then placed in Mfful furnace at a temperature of 800C°for 3 hours.

**3-1 Effect of pH .**

40 ml of a solution containing cadmium, zinc, lead, copper and lead ions at a concentration of 400 µgml<sup>-1</sup> was added to 1 gm of plant residue (crushed plant parts) with a diameter of 400 µm, then placed in plastic bottles of 100 ml and the pH of the solution was adjusted (3,4,5,6,7, 8 and 9) by a pH\_meter using a dilute solution of sodium hydroxide with a concentration of 0.1N and dilute hydrochloric acid with a concentration of 0.1N by placing drops until the required degree is reached. The bottles were tightly closed and shaken by mechanical shaking at a speed of 2000 rpm for 3 hours. After the end of the shaking period, the samples were filtered through filter paper No. (1). The remaining concentration of lead, cadmium, zinc, copper and nickel ions in the solution was estimated by an atomic absorptionspectrophotometry (A.A.S). The number of experimental units for each metals was 350 experimental units, which came from (5 x 7 x 5 x 2) to represent (plant residues x pH x heavy metals x replicates) respectively. Then the equation below was applied to calculate the adsorbed amount of the metals (Ajala and Ojoawa,2022)

$$X = \frac{(Co - Ce)V}{m}$$

Where:-

X: the amount adsorbed in µg gm<sup>-1</sup>

Co: initial metal concentration µg ml<sup>-1</sup>

Ce: metal equilibrium concentration µg ml<sup>-1</sup>

V: volume of heavy metal solution ml

m: weight of the plant residue g

**3-2 Particle diameter :**

The ground plant residues (for each of the five plant species) were passed through a series of sieves with different sizes of apertures less than 200,200-400,400-600 and 600-800 µm . The weight of 1 gm of these particles of different sizes was taken for each of the plant species separately, and they were placed in plastic containers with a capacity of 100 ml, then solutions were prepared with a concentration of 400 µgml<sup>-1</sup> and pH = 6, and 40 ml of solutions were placed in containers containing plant residues, same steps were repeated in the previous experiment for estimating and shaking. The removal percentage was calculated from the following equation :

$$\% \text{ Removal} = \left( \frac{Co - Ce}{Co} \right) * 100$$

**3-3 Effect of temperature .**

The weight of 1 gm of plant residue was taken and placed in plastic containers with a capacity of 100 ml and 40 ml of the solution was added to each element at a concentration of 400 µgml<sup>-1</sup> at pH = 6, the samples were placed in

the shaker incubator and the temperature was set at 283, 293, 303,313 and 323 Kelvin For a period of 3 hours, the concentration of heavy elements was estimated using the atomic absorption spectrophotometry and the adsorbed amount of the element was calculated as previously mentioned

**4. RESULTS AND DISCUSSION :**

**4-1 Effect of the pH**

The pH of the solution is an important unit in the adsorption process, as the state of solubility of these elements in the solution, and thus the interactions of complex formation, electrostatic interference, and sedimentation in the physical adsorption processes on the adsorption surfaces, are controlled by the pH values. The results showed in general (Fig. 1, 2, 3,4,5) there is a decrease in the adsorption of element ions at low pH values this may be due to an increase in the concentration of  $H_3O^+$  ions at low pH values in the solution, which puts it in great competition with the studied element ions on the exchange sites for used plant materials and thus less chance of engagement on exchange sites (Khalid *et al.*,2019). The results also showed a variation in the amount of adsorption of the ions of the studied heavy metals in different plant residues, which may be attributed to a difference in the affinity of these elements with the sites of adsorption and may be to the difference in the diameters of these ions and their suitability to the binding sites or what is known as the electronic negativity of the metal electronegativity (Jasperi *et al.*,2020 ). Which may mean the tendency or susceptibility of the atom or the active group to attract electrons (or the electronic density) towards it and also may be due to the decrease in the concentrations of ions of the studied elements in the solution with a rise of pH more than 7,not due to the association of the ions of the studied elements with the locations on the surfaces of plant residues as much as they are Complexation and precipitation of these ions in solution (Bhattacharyya and Gupta, 2008), which lowered their concentration and did not bind to the active sites, because these elements tend to precipitate and form complexes at high pH. (Wang *et al.*, 2007) indicated that nickel is likely to precipitate at  $pH > 7$  and that the maximum adsorption of nickel is at  $pH = 6$  ( Zagorscak and Thomas.,2022), while (Nasrabadi *et al.*,2020) mentioned that zinc decreases its solubility at high pH ( $pH > 7$ ), and also for the other studied elements, It is believed, and according to what appeared from the results, and to avoid the state of sedimentation that may result from the high pH of the solution, the value of  $pH = 6$  may be more appropriate to explain the efficiency of plant residues in the adsorption capacity because the element is dissolved at this value of pH, and therefore the increase in adsorption is due only to the efficiency adsorbent.

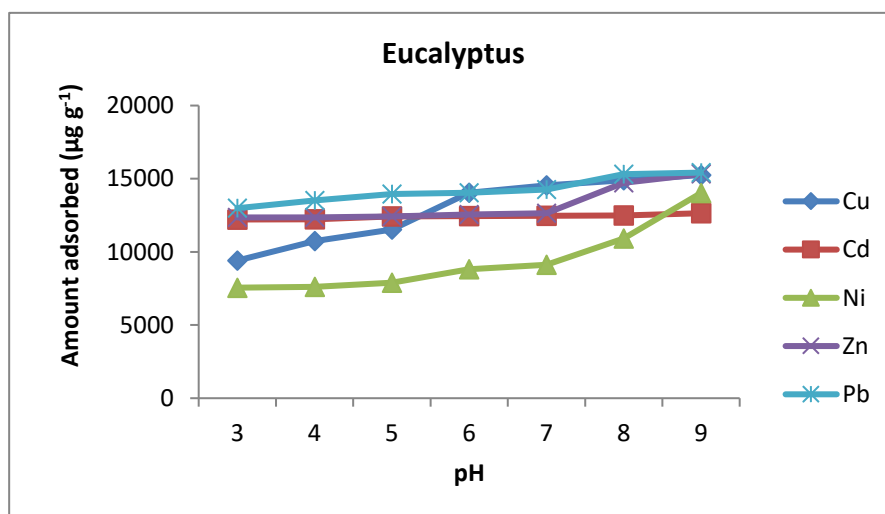


Figure1: The adsorption amount of the heavy metals on the Eucalyptus at different pH

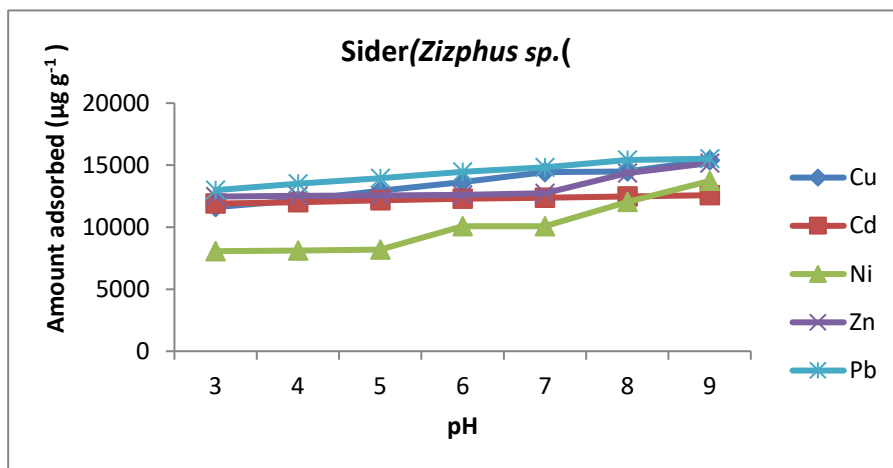


Figure2: The adsorption amount of the heavy metals on **Sider(*Zizphus sp.*)** at different pH

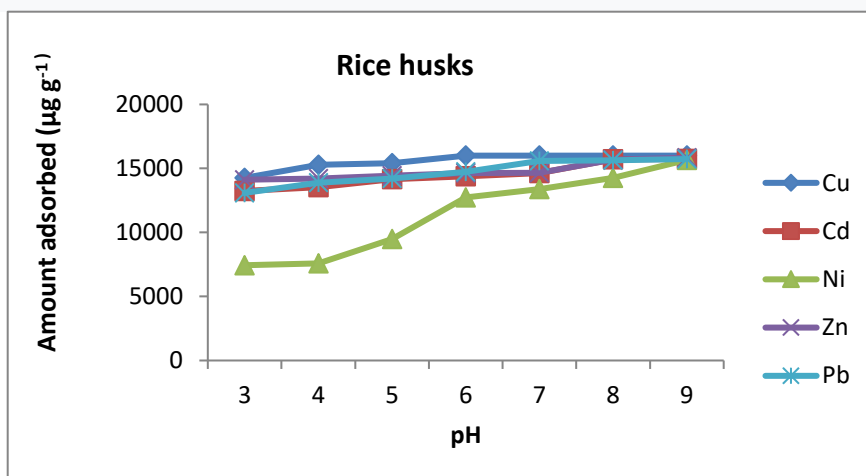


Figure3: The adsorption amount of the heavy metal on **Rice husks** at different pH

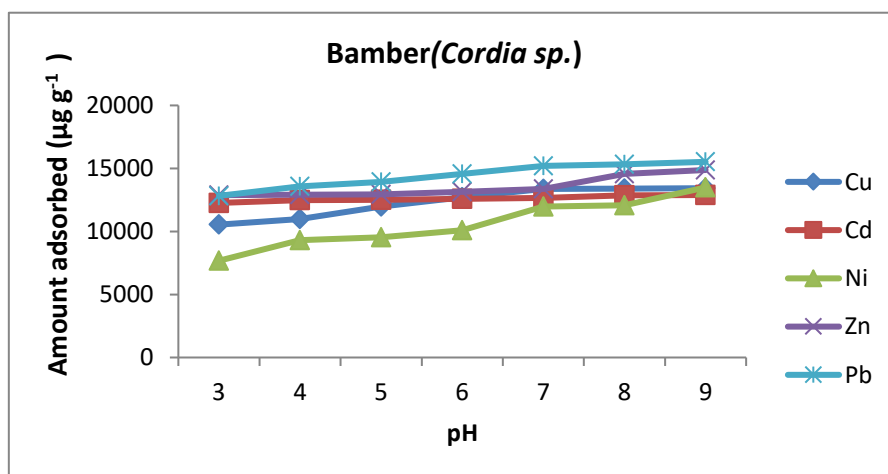


Figure4: The adsorption amount of the heavy metal on **Bamber(*Cordia sp.*)** at different pH

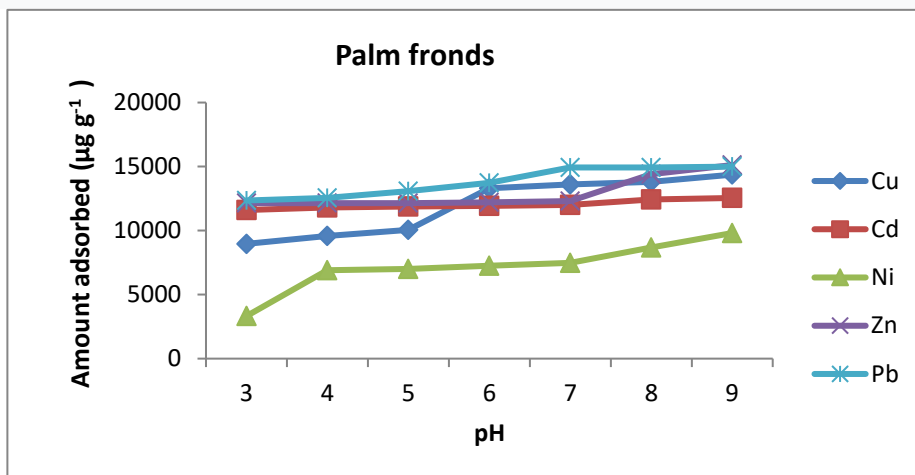


Figure5: The adsorption amount of the heavy metal on **Palm fronds** at different pH

**4-2 Effect of diameter :**

The results indicated a difference in the effect of particle sizes of plant residues on the adsorption of heavy metals (Cd<sup>+2</sup>, Pb<sup>+2</sup>, Ni<sup>+2</sup>, Cu<sup>+2</sup>, Zn<sup>+2</sup>), and the percentage of heavy metals removal decreased with increasing the size of residue particles (table 1), as the percentage of removal of Bamber (*Cordia sp.*) branches ranged between (77.505-80.599, 92.389-95.228, 53.388-73.665, 71.782-83.515, 95.527-95.988)%, and the percentage of Sidr (*Zizphus SP.*) branches ranged between (77.938-78.612, 91.618-94.291, 56.389-68.529, 55.808-82.643, 97.446-98.040) % and for Rice husks ash (69.463-78.025, 81.637-85.871, 90.274-92.198, 93.094-97.480, 100.000-100.000)% and for *Eucalyptus* branches (78.024-80.047, 62.326-73.611, 53.426-56.189, 91.774-93.023, 95.948-99.654)% and finally the Palm fronds ranged between (47.785-54.055, 59.525-68.993, 77.523-80.599, 91.310-92.204, 95.833-97.614)% for the elements Zinc, Cadmium, Nickel, Copper and Lead, respectively. The highest percentage of removal was achieved in all plant residues at the size of the particles with diameters less than 200 microns, and vice versa at the diameters of 800 microns, and the possibility that this is due to the increase in the surface area of the small particles compared to the large particles, and thus the increase in the presence of effective adsorption sites. It provides greater opportunities for contact and association with ions of heavy elements, and these results came in the same direction with the (Jain *et al.*, 2004). In order to compare the efficiency of the plant residues in percentages of removal at the maximum removal, which occurred when the diameters of the particles are less than 200µm, regardless of the type of element ion removed, the plant residues took the following order :

Ashes of Rice husks > Bamber branches > Sidr branches > Eucalyptus branches > Palm fronds, with average : 93.165%, 85.799%, 84.423%, 81.732%, 78.693%, respectively (not shown in the table).

**Table (1) Percentage of heavy metals removal by plant residues in different particle sizes**

plant residues	diameter (micron)	Heavy metales				
		Zn	Cd	Ni	Cu	pb
Bamber	less than200	80.599	95.228	73.665	83.515	95.988
	200-400	80.409	94.641	63.526	81.017	95.695
	400-600	79.165	94.262	63.193	80.813	95.452
	600-800	77.505	92.389	53.388	71.782	95.527
Sider	less than200	78.612	94.291	68.529	82.643	98.040
	200-400	78.560	92.682	64.393	69.661	97.840
	400-600	77.868	92.445	62.860	65.595	97.690
	600-800	77.938	91.618	56.389	55.808	97.446
Eucalyptus	less than200	80.047	93.023	62.326	73.611	99.654
	200-400	79.424	92.654	56.456	71.491	99.605
	400-600	78.318	91.925	52.587	64.434	98.160
	600-800	78.024	91.774	56.189	53.426	95.948
Rice huck ash	less than200	85.871	97.480	90.274	92.198	100.000
	200-400	85.508	95.876	86.872	90.398	100.000
	400-600	83.071	95.290	82.870	87.580	100.000

	600-800	81.637	93.094	69.463	78.025	100.000
Plam frondes	less than200	80.599	92.204	54.055	68.993	97.614
	200-400	80.340	91.618	51.787	63.649	96.613
	400-600	77.799	91.377	49.252	61.646	95.527
	600-800	77.523	91.310	47.785	59.525	95.833

**4-3 Effect of temperature .**

The temperature is directly related to the kinetic energy of the element ion in the solution and with an increase in the temperature followed by an increase in the rate of diffusion of the adsorbent. In addition to that, the effect of temperature succession on the adsorption capacity of the equilibrium of the adsorption material towards the adsorbent (Renu *et al.*,2020). The adsorption results showed that there was an increase in the amount of adsorption (removed) for all elements under study with increasing temperature and for all plant residues, (table 2). There was a clear variation in The association of the elements with the adsorption sites on the plant residues with the change in the temperature of the medium, and this is due to a change in the volume of the void (expansion of the pores) of the adsorbent leading to diffusion of ions within the particles or the enhancement of the chemical affinity of the element ion with the surface of the adsorbent material and or an increase in the active sites of the solid part. Accompanied by high temperatures bonding of the element on the surfaces of the adsorbent material. This leads to an increase in the irregularity or arrangement on the surface of the adsorption material (plant residues) and an increase in the chemical reactions that occur during the adsorption process, which results in an increase in the adsorption capacity (Jafari *et al.*,2023).The temperature of 323 K achieved the maximum amount of removal at the different plant residue treatments regardless of the studied metales, the following order: Rice husk ash > Bamber branches > Sidr branches > Palm fronds > Eucalyptus branches at average 14814, 13929, 13426, 12618, 12480 µgm gm<sup>-1</sup>. (not mentioned in the table.

**Table (2) The amount removed µg gm<sup>-1</sup> of the heavy metals zinc, cadmium, nickel, lead and copper on the plant residues at different temperatures (Kelvin)**

plant residues	metals	temperatures (Kelvin)				
		283	293	303	313	323
Bamber	Zn	12165	12266	12440	12534	12808
	Cd	15461	15551	15636	15726	15775
	Ni	9794	11120	11635	11999	12215
	pb	15135	15199	15224	15300	15353
	Cu	9588	11000	11768	13006	13495
Sider	Zn	11902	12185	12772	12889	12996
	Cd	14897	15025	15304	15457	15572
	Ni	8558	9354	10374	10681	11834
	pb	15078	15146	15278	15365	15445
	Cu	7422	8261	9868	10357	11287
Rice huck ash	Zn	12850	12940	13057	13382	13444
	Cd	15501	15598	15695	15813	15947
	Ni	10606	12290	12414	13707	13,890
	pb	15958	15986	15990	15992	15995
	Cu	12454	13460	14445	14655	14795
Eucalyptus	Zn	12406	12554	12604	12898	13038
	Cd	14670	14743	14810	15071	15162
	Ni	8691	10092	9653	10051	10374
	pb	15441	15518	15718	15746	15952
	Cu	3544	4976	66191	7247	7876
Palm fronds	Zn	12423	12582	12766	12912	13046
	Cd	14599	14624	14757	14788	15108
	Ni	7870	8376	8,757	9421	9628



pb	15920	15943	15950	15961	15966
Cu	683	3935	4955	6199	9344

### CONCLUSIONS:

The high pH values of the solutions achieved the removal of the polluting metals (Cu<sup>+2</sup>, Cd<sup>+2</sup>, Ni<sup>+2</sup>, Zn<sup>+2</sup>, Pb<sup>+2</sup>) and the efficiency of Rice husk ash ranked first in removal, followed by Bamber branches and then Sidr branches. The diameters of the particles of plant residues less than 200 µm have the highest capacity for adsorption of polluting metals compared to the diameters of the other particles studied. The plant residues contributed to the removal of polluting elements from their solutions at high rates that reached 100% in some of them, as in the ashes of Rice husks. All plant residues agreed to achieve the highest removal of lead compared to other studied elements, reaching between 95.5-100%. The temperature also contributed to increasing the amount of elements removed, as the temperature of 323 K gave the maximum removal of the studied elements compared to other temperatures under study.

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