

## Article

# Using Agricultural Mixed Waste as a Sustainable Technique for Removing Stable Isotopes and Radioisotopes from the Aquatic Environment

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**Abstract:** In the current study, dried ground mixed waste of olive waste and water hyacinth was subjected to the treatment of wastewater contaminated with stable or radioactive cobalt and cesium. Contact time, temperature, amount of adsorbent, metal ion concentration and pH-value were evaluated as the most potent factors that affect the adsorption process. Concentrations of Co and Cs ions before and after treatment are measured using ICP-OES technique. Moreover, kinetic and equilibrium isotherm parameters were investigated by explaining the equilibrium data by induction of two isotherms, “Langmuir” and “Freundlich”. Experimental results indicated that more than 85% of <sup>60</sup>Co and <sup>134</sup>Cs were efficiently removed from spiked wastewater after one hour of contact time by using 0.2 g of dried mixed waste. The optimal time to remove Cs<sup>+</sup> and Co<sup>2+</sup> was 50 and 90 min, respectively. The kinetic study showed that the adsorption of Cs<sup>+</sup> and Co<sup>2+</sup> were better suited to the second order. The most favorable pH value was at a range of 6–7 for Cs<sup>+</sup> and 5–6 for Co<sup>2+</sup>. R<sup>2</sup> values were higher at the Freundlich isotherm, indicating that the adsorption process was taking place according to Freundlich isotherm at all temperatures for Co<sup>2+</sup> and Cs<sup>+</sup> ions.

**Keywords:** olive waste; water hyacinth; cobalt; cesium; adsorption; kinetics; biomass; wastewater; ICP-OES spectroscopy



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## 1. Introduction

Water covers the vast majority of the Earth’s surface (71%), including only a small portion (3%) of freshwater that plays a vital role in the global economy. This natural essential resource became insufficient in numerous areas. The water scarcity was a major economic and social concern, especially in the agriculture sector, which consumes about 70% of the freshwater [1]. In many attempts in developing countries, 90% of the wastewater remains untreated in freshwater bodies, rendering it unsafe for human consumption, resulting in scarcity or toxic impact [2]. Clean soil and pure water are the most in-demand resources all over the world nowadays [3,4]; also in demand are more efficient selective methods for analysis [5,6].

Pollution is an important issue around the world, especially the contamination with chemical elements, including stable isotopes or radioisotopes. Chemical pollution is the most significant problem because of the easy dispersal of harmful metals, for example, it can be transferred into edibles; however, water contamination is considered a serious ecological problem around the world as well [7].

Physicochemical approaches, such as precipitation, redox-reaction, ion exchanging [8,9] and electrochemically [10], have been reported. Ultrafiltration [11], photocatalysis [12], reverse osmosis [13] and electro flotation [14] are some of the other important techniques. The disadvantages of these techniques are their cost and the production of secondary waste that requires extra treatment [15].

Developing sustainable, simple, efficient, and cost-effective techniques for removing different elements from wastewater is recent worldwide challenge. Because it is a straightforward means of removing metal ions from wastewater, phytoremediation [16,17], using aquatic plants, and adsorption process [18–20], using agricultural waste, become viable choices. For example, the cost of excavation and disposal of contaminated soil based on the Federal Remediation Technologies Roundtable (FRTR) data was in the range of 270 to 460 \$/ton, while the cost of phytoextraction, including plant selection and position, irrigation, soil amendment, field monitoring, harvesting, and residue management, could be in the range 10–35 \$/ton [21].

Although phytoremediation using aquatic plants achieved great effectiveness in the treatment process, the findings of the experiments revealed that dried plants are more effective than fresh plants, and consequently, adsorption techniques have recently been investigated in the management of contaminated wastewater spiked with arsenate, nitrate, phosphate, cadmium, and other heavy metals [7].

Increased cobalt concentrations injure humans, causing lung irritations, asthma, pneumonia, weight loss, paralysis, diarrhea, vomiting, thyroid hormones, liver damage, and nausea, due to the resemblance of cesium-134 to sodium; the ingestion of cesium causes its deposition in soft tissues throughout the body, posing internal hazards [22].

Physical procedures, such as adsorption by natural or manufactured materials, have been used to remove cesium and cobalt. As one of the traditional adsorbents, ion exchangers have limitations because of their competition with other monovalent cations found in liquid radioactive waste, such as sodium and potassium, which can inhibit the adsorption of cesium or other competing divalent cations capable of occupying cobalt adsorption sites [23]. Because the metal adsorption depends on cation exchange sites, it requires the development of new and more active materials with safe and sustainable features for the economical removal of stable or radio pollutants from wastewater.

Water hyacinth is an aquatic plant weed that grows quickly and floats freely in water. It has a reputation of having a very high metal tolerance. The adsorption of  $Pb^{2+}$  from liquid effluents was conducted using the dried biomass of *Eichhornia crassipes*. The performance of the adsorption mechanism was determined to be depended on pH and the adsorbent's uneven surface may aid in the adsorption of  $Pb^{2+}$  on the surface of the adsorbent [24].

The current study focuses on the utilization and effectiveness of biomass originated from dried ground mixed waste of water hyacinth and olive waste to remove cesium and cobalt from wastewater due to their high availability and low operating cost instead of traditional techniques, which are difficult to control and require primarily expensive requirements. Cobalt compounds should be considered genotoxic carcinogens with a practical threshold, and chronic inhalation of cobalt compounds can induce respiratory tumors [25], while the toxicity of cesium can induce gastrointestinal distress, hypotension, syncope, numbness or tingling of the lips [26]. Moreover, radiotoxicity was added on the chemical toxicity when in contact with radio cobalt or radio cesium.

In order to identify the ideal physicochemical conditions for the most effective sorption for treatment applications, the adsorption process was studied in terms of the impact of pH-value, adsorbent dose, metal ion, and interaction time. Additionally, the adsorption process followed the “Langmuir” and “Freundlich” adsorption models, and the kinetic studies were developed to describe the isothermal trend and constants of the adsorption process, with the pH-value being the most important factor. An experiment, using the nominated adsorbent to eliminate radiocobalt and radiocesium, was performed. These treatment processes generated secondary radioactive waste that had to be stabilized and

remediated for safe disposal, as reported in the previous literature, by using cement [27,28], cement polymer composite [29,30], and cement mixed with natural clay [31].

## 2. Experimental Approach

### 2.1. Materials

#### 2.1.1. Adsorptive Materials

Green plants of water hyacinth have been harvested from Egyptian waterways, dehydrated in a 70 °C oven for 2 days and ground to produce a fine powder using an electric mixer. Other types of unimportant materials, such as solid waste derived from the olive oil industry, were prepared by washing thoroughly with methanol, rinsing with pure water, and introducing them to drying at 70 °C. The dry matter was crushed and ground. The two dried ground plant wastes were mixed with a ratio of 50 to 50%. As shown in Figure 1, the chart of spectra confirmed the presence of major functional groups in ground dry single waste and mixed plant waste that were identified and characterized using FTIR spectroscopy with a Shimadzu FTIR-8201 PC. FTIR analysis was performed to identify the functional groups in the dried single waste and mixed waste using the KBr disc method. This investigation showed a strong broad absorption band of O-H stretching vibrations in the region of 3272–3177 cm<sup>-1</sup> and another peak at 3445 cm<sup>-1</sup>. Other peaks appeared at 1739 cm<sup>-1</sup> and 1322 cm<sup>-1</sup>, which corresponded to the C=O stretching vibrations and the phenolic hydroxyl groups, respectively. At 1547 cm<sup>-1</sup>, a stretching vibration regarding the C–N of the peptidic bond of proteins was present; a peak at around 1500 cm<sup>-1</sup> indicated C=C bending. The range 1130–1000 cm<sup>-1</sup> is a vibration of C–O–C and O–H of polysaccharides. The intensity of the adsorption bands confirmed no significant difference between olive waste alone and that mixed with water hyacinth; the functional groups were responsible for the adsorption of many elements.

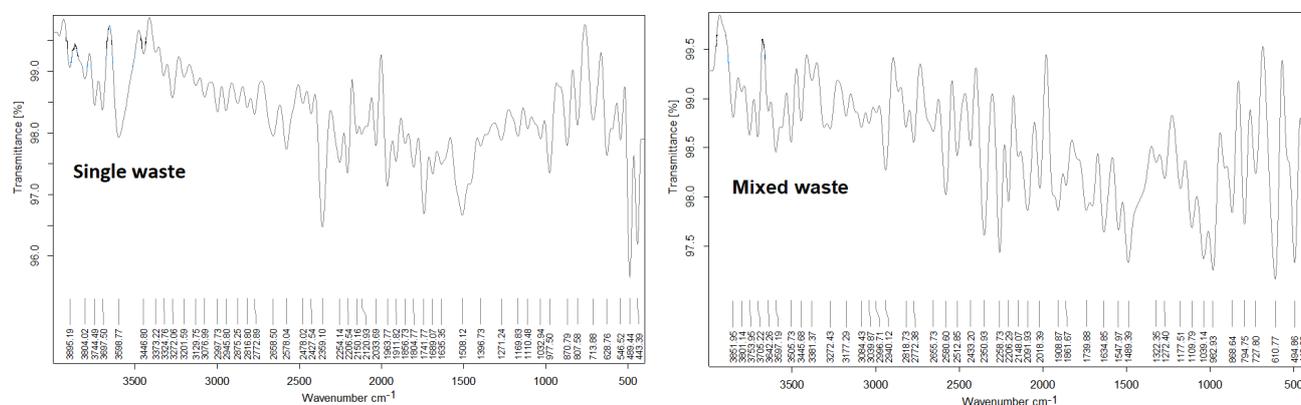


Figure 1. FTIR spectra of single waste and mixed waste.

#### 2.1.2. Adsorbate Elements

Radioisotopes of <sup>134</sup>Cs and <sup>60</sup>Co were produced at the Egyptian Atomic Energy Authority, Egypt. The activity content of two radioisotopes was diluted to various activities in the range of (1400–4850 Bq). High purity and analytical-grade salts of stable cesium and cobalt were purchased from Sigma-Aldrich. To achieve a suitable value of concentration for the subsequent experimentations, dilution was used to adjust the concentration of stable elements in the range of (5–100 mg/L). Additionally, gradual addition of NaOH and 0.1 N of HCl were performed to adjust the acidity or alkalinity of the spiked solutions for various pH values.

## 2.2. Experimentation

### 2.2.1. Adsorption Studies

All experiments have been performed in a bathwater with a constant shaking speed to investigate the influence of contact time, pH-value, adsorbent weight, concentration of Cs<sup>+</sup> or Co<sup>2+</sup> and temperature.

### Adsorption of Single Waste and Mixed Waste

A primary experiment was performed to evaluate adsorption efficiency using different weights of single waste (olive waste) and mixed waste (olive waste) to uptake Cs or Co elements for 2 h at the constant temperature of 25 °C, pH value of 7 and shaking speed of 100 rpm.  $q_e$  was performed according to the recent literature [32] and Equation (1).

$$q_e = \left( \frac{C_0 - C_e}{m} \right) \times V \quad (1)$$

$q_e$  is the adsorption capacity (mg/g),  $C_0$  is the initial concentration of ions (mg/L),  $C_e$  is the equilibrium concentration of ions (mg/L),  $m$  is the mass of biosorbent used in the process (g) and  $V$  is the volume of the solution (L).

### Contact Time

The effect of time on the adsorption of (50 mg/L)  $Cs^+$  or  $Co^{2+}$  on (0.1 g) mixed waste was evaluated at the constant temperature of 25 °C while maintaining the pH value at 7 and the shaking speed of 100 rpm at variable time during two hours (0, 1, 5, 10, 15, 20 and 30, 45, 60, 90 and 120 min). At each contact time, the concentration of non-adsorbed ions was determined by taking 1 mL of clear solution for subsequent analysis using atomic absorption spectroscopy.

### pH Effect

Under varying pH conditions, stable elements were adsorbed at the constant temperature and a shaking rate of 25 °C and 100 rpm, respectively. The pH of non-radioactive solutions was maintained by the addition of 0.1 N of HCl and NaOH to adjust the pH. The removal percentage of both ions (concentration, 50 mg/L) at various pH (3 to 8) by using 0.1 g of dry ground mixed waste was determined at 60 min.

### Adsorbent Dose

Six extended weights of dry mixed waste (0.025, 0.05, 0.1, 0.25, 0.5, and 1.0 g) were poured to six bottles including 100 mL of  $Cs^+$  or  $Co^{2+}$  at 25 °C, shaken at 100 rpm, with pH-value (7 for  $Cs^+$  and 6 for  $Co^{2+}$ ) for 60 min. Then, 1 mL of each bottle was collected and analyzed to determine the remaining concentration of each element after completing the process.

### 2.2.2. Inductively Coupled Plasma Optical Emissions Spectroscopy (ICP-OES)

ICP-OES, inductively coupled plasma optical emission spectroscopy (Prodigy High Dispersion ICP, Leeman in the United States) was used to evaluate the initial concentrations of Co and Cs, as well as the concentration of the remaining metal in the solution. The liquid was converted into an aerosol inside the instrument via a process known as nebulization. The sample aerosol was then transported to the plasma and desolvated, vaporized, atomized, excited, and/or ionized by the plasma. The excited atoms and ions emitted their characteristic radiation, which was collected and sorted by wavelength by a device [33].

The limit of detection (LOD) was primarily determined by instrumental sensitivity, spectral interferences, memory effects, digestion vessel cleanliness, and analytical reagent blank level, and it is possible to define the lowest concentrations that can be reliably detected and quantified.

The LODs for each element were calculated as the concentration equivalent of three times the standard deviation of the ion counts obtained from duplicate runs of reagent blank solutions (3% *v/v*  $HNO_3$  including the internal standard spikes) [34].

ICP-OES has the following features:

Available in radial, axial and dual view configurations;

Wavelength range from 165 to 1100 nm;

Resolution < 0.008 nm;

High precision elemental ratio measurements (0.01–0.02% RSD).

### 2.2.3. Adsorption Isotherms and Data Analysis

At various temperatures and constant pH and shaking speed (pH = 7, 100 rpm), adsorption isotherms were extrapolated for Cs and Co ions at the two contact times of 45 and 90 min, respectively. At 15, 25, 35, and 45 °C, 0.1 g of dried mixed waste was added to 100 mL of Cs or Co ions of various initial concentrations ( $C_0$ ), (5, 10, 20, 50, 75, and 100 mg/L), respectively, until equilibrium time was reached. A total of 1 mL of the remaining clear solution was analyzed by an atomic absorption apparatus to determine the non-adsorbed ions under various conditions.

Batch data from various contact times with variable initial concentrations of metal ions were analyzed using kinetic studies (pseudo-first order, pseudo-second order, and intra-particle diffusion).

Furthermore, different models of isothermal adsorption, such as “Langmuir” and “Freundlich”, were applied to investigate the adsorption potential of different ions.

### 2.2.4. Adsorption of Radioisotopes ( $^{60}\text{Co}$ or $^{134}\text{Cs}$ )

Absorption processes at constant room temperature were followed by using 50 mL of  $^{60}\text{Co}$  or  $^{134}\text{Cs}$  solutions of various initial activity in the range of (1400 to 4350 Bq for  $^{134}\text{Cs}$ , and 3800 to 11000 Bq for  $^{60}\text{Co}$ ). In this experiment, extended doses of adsorbent (0.5, 1.0, 1.5, 2.0, 2.5 g) were mixed with  $^{60}\text{Co}$  and  $^{134}\text{Cs}$  for contact time (120 min.), constant shaking speed (100 rpm), and constant pH-value (7). The removal of radionuclides was assessed by periodically recording the radioactivity content of the remaining solution using a multichannel analyzer of a NaI detector, PCAP, USA.

### 2.2.5. Statistical Analysis

All results are expressed as means  $\pm$  SEM and regression models with the SPSS software (Statistical Package for the Social Sciences), version 24 (IBM SPSS Statistics for Windows, Armonk, NY, USA: IBM Corp.). Comparisons made between more than two groups were performed using multifactorial analysis of variance (ANOVA) followed by the Least Significant Difference (LSD) test. Differences were considered statistically significant when the alpha probability was  $\leq 0.05$ .

## 3. Results and Discussions

### 3.1. Single Waste and Mixed Waste

The potential of agricultural waste has been conducted in recent decades according to its ability to accumulate some elements from contaminated water or soil [35]. At least the same potential, such as terrestrial plants, can eliminate heavy metal contaminants that are commonly found in the environment, such as Cd, Cr, Cu, Hg, Pb, Ni, Zn and Fe [36].

Some of these elements are essential for plants but the others are toxic or still have unknown biological effects. A primary experiment was performed to evaluate the efficiency of using single waste, such as dry olive waste, and mixed waste of the same waste with dry water hyacinth to uptake Cs or Co elements after 2 h of contact. Figure 2 shows the superiority of mixed waste in the accumulation of constant concentration of Cs and Co by different amounts of adsorbents. According to the observed results, it is confirmed that mixed waste has a tendency to accumulate more than single waste, especially in the case of Co. From the statistical analysis, it is clear that F for treatment = 19.336 at  $p = 0.00$  ( $<0.05$ ) and this confirms the statistically significant differences between the averages in the case of treatment. Consequently, in this study, the mixed waste of olive waste and water hyacinth was selected to evaluate its efficiency to eliminate cobalt and cesium elements from artificial contaminated water. This result is consistent with the recently published literature conducted to evaluate the use of mixed biomass (*Aspergillus campestris* and agro waste) [37].

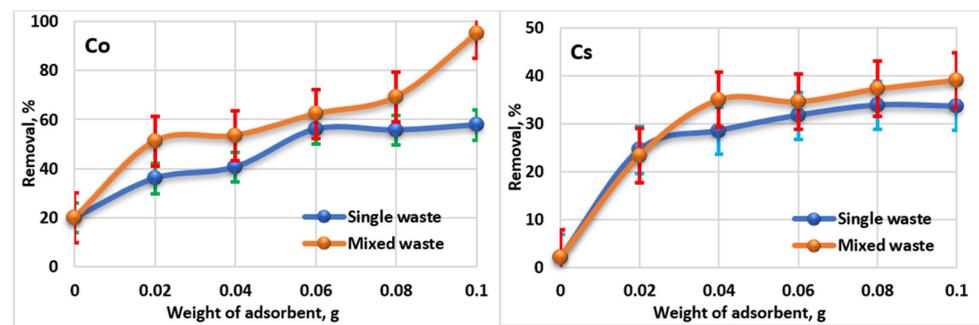


Figure 2. Adsorption of both ions separately on dry single waste and mixed waste.

### 3.2. Contact Time

Contact time acts as a vital key for determining the equilibrium point at the highest adsorption value. The optimal contact time is the time spent to achieve the highest removal percent with respect to different adsorption concentrations by evaluating the biosorption kinetics [38]. In this experiment, the statistical analysis confirmed that  $F$  for time = 70.681 when  $p = 0.000$  ( $<0.05$ ) and this confirms the statistically significant differences between the averages in the case of the time factor.

At a constant pH (7) and constant temperature (25 °C), the adsorption capacities of both ions ( $\text{Cs}^+$  or  $\text{Co}^{2+}$ ) onto dry biowaste was evaluated. As shown in Figure 3, as the contact time increases, the rate of adsorption first increases and then becomes constant due to the aggregation of adsorbed ions that diffuse deeper into the adsorbent structure at higher energy sites with time. This aggregation negates the influence of contact time as the pores get filled up and start offering resistance to the diffusion of aggregated ions in the adsorbents [39]. However, the elimination efficiency was increased and maximum biosorption capacity was achieved in 50 and 90 min, respectively.

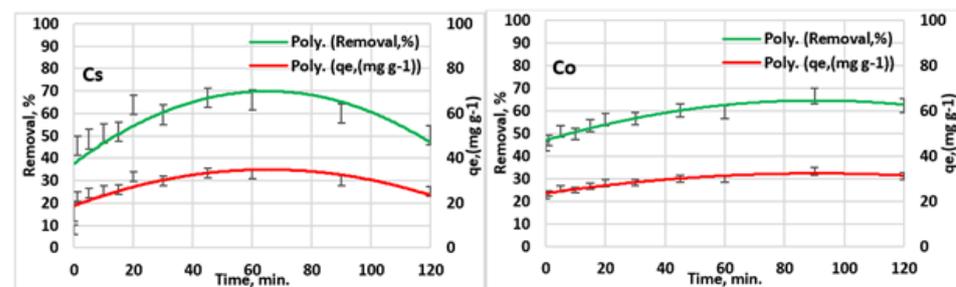


Figure 3. Removal (%) and adsorption capacity( $q_e$ ) of both ions separately using dry mixed waste during contact time.

The data are means  $\pm$  SE ( $n = 5$ ). ANOVA followed by LSD's multiple comparison post hoc test; the differences were considered significant at  $p \leq 0.05$ . A statistically significant difference  $F$  (contact time) was 70.681 at  $p = 0.000$  ( $<0.05$ ).

The calculated data of adsorption/time were employed to both kinetic models; pseudo-first order as shown in Equation (2) and pseudo-second order as shown in Equation (3).

$$\text{Log} (q_e - q_t) = \text{log} q_e - (k_1/2.303) t \quad (2)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} \quad (3)$$

where  $k_1$  ( $\text{min}^{-1}$ ) is the pseudo-first order rate constant of adsorption,  $q_e$  and  $q_t$  ( $\text{mg/g}$ ) refer to amount of ions adsorbed at equilibrium and at time  $t$ , respectively.  $K_2$  ( $\text{g mg}^{-1} \text{min}^{-1}$ ) is the pseudo-second order rate constant of adsorption.

The suitable model was investigated by computing the correction coefficient value of straight line ( $R^2$ ). As indicated in Figure 4, the adsorption of  $\text{Cs}^+$  ion processes were in pseudo second order greater than in pseudo-first order because the high regression coefficient ( $R^2$ ) of the pseudo-second order model was higher than that in the pseudo-first order model. Similarly, the adsorption of  $\text{Co}^{2+}$  ion processes fit the second-order model more than the first-order model according to the higher value of regression coefficient ( $R^2$ ) in the case of the pseudo-second order model, as shown in Figure 5 [40]. The adsorption kinetics of  $\text{Cs}^+$  and  $\text{Co}^{2+}$  suited the pseudo-second order model, indicating that the key step in the adsorption mechanism was the ion exchange by electrostatic interactions.

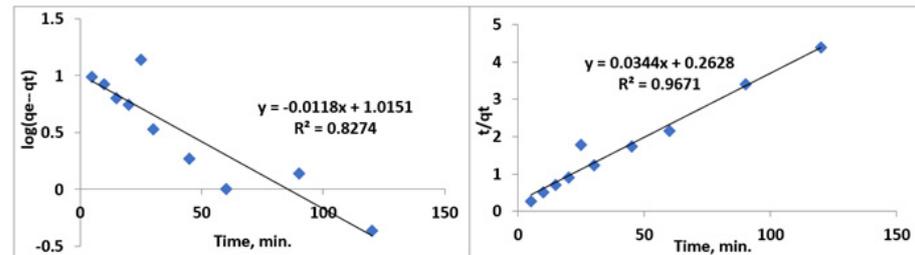


Figure 4. Adsorption kinetics modeled using pseudo-first and pseudo-second orders for  $\text{Cs}^+$ .

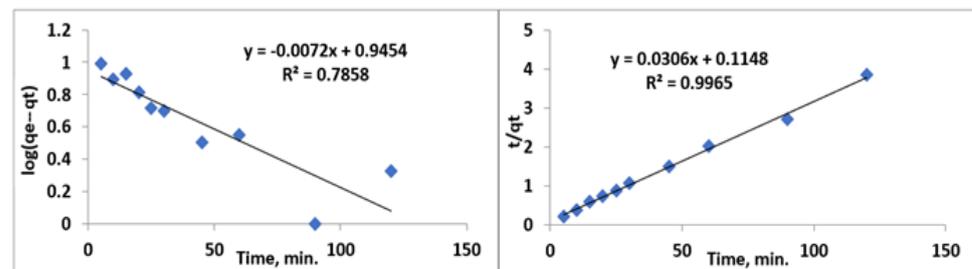
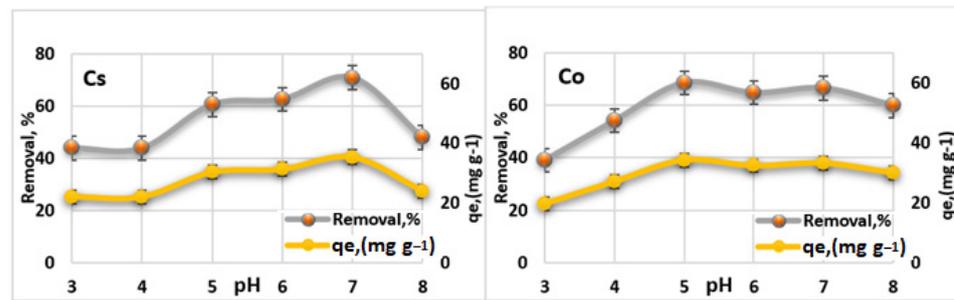


Figure 5. Adsorption kinetics modeled using pseudo-first and pseudo-second orders for  $\text{Co}^{2+}$ .

### 3.3. pH Effect

pH is one of the most important factors that largely affects the adsorption processes due to the effect of  $\text{H}^+$  ions on the biosorption of  $\text{Cs}^+$  and  $\text{Co}^{2+}$  ions by using dry plant mixed waste [41]. The potential of changing the pH from 3 to 8 was evaluated and presented in Figure 6, significantly pH could change the relative distribution of ions in the solution, as well as the surface properties of the adsorbent. The adsorption of the mixed waste was clearly affected by the varying solution pH values, which considerably increased by enhancing the solution's pH from 2 to 5, and basically remained unchanged in the case of the Co solution. In the case of the Cs solution, at pH 7, the removal capacity reached maximum value; however, afterwards rapidly decreased at a solution pH of more than 7.0. This behavior can be explained by the fact that at lower pH values, excess  $\text{H}^+$  ions compete with  $\text{Cs}^+$  and  $\text{Co}^{2+}$  for free adsorption sites, reducing their adsorption. With low pH, the solubility of some heavy elements in water increases, which causes an increase in their spread; pH plays a critical role in the transfer of heavy metals between the liquid and solid phases [42]. Another factor that could contribute to enhancing metal ion adsorption is the increasing pH, which encourages metal ion precipitation from the solution in the form of hydroxides [43]. From the statistical analysis it is clear that the F value (for pH) was 7.274 when  $p = 0.016$  ( $<0.05$ ) and this confirms the statistically significant differences between the averages in the case of this factor.

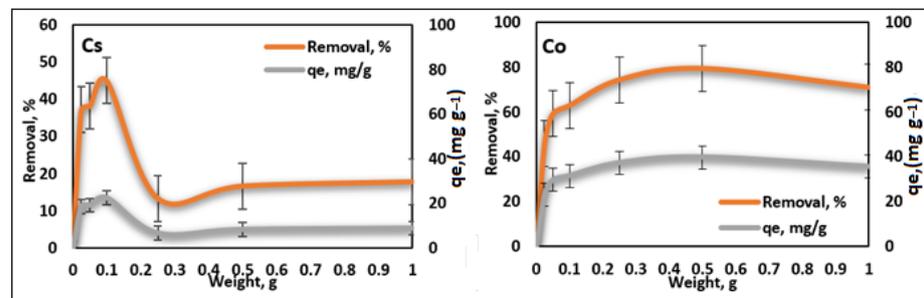


**Figure 6.** Removal (%) and adsorption capacity ( $q_e$ ) of both ions separately in dry mixed waste at different pH-values.

The data are means  $\pm$  SE ( $n = 5$ ). ANOVA followed by LSD's multiple comparison post hoc test; the differences were considered significant at  $p \leq 0.05$ . A statistically significant difference F (pH) was 7.274 at  $p = 0.016$  ( $< 0.05$ ).

### 3.4. Dosage Weight of Dried Mixed Waste

Knowledge of the effect of the adsorbent dose on adsorption is critical for optimal adsorbent use. Figure 7 shows that batch studies with varying quantities of mixed waste were carried out to determine the best adsorbent dose. The adsorption capacity ( $q_e$ ) was increased by increasing the adsorbent dose and then proceeded steadily at a high level in the case of Co, while in case of Cs it increased with the increasing the dose, then dropped suddenly, and then settled at a low level. The rise in removal with the dose is associated with the increased number of active sites, it is readily understood that the number of available adsorption sites and the surface area increase by increasing the adsorbent dose [44].



**Figure 7.** Removal (%) and adsorption capacity ( $q_e$ ) of both ions separately at different loading weights of mixed waste.

The steady removal thereafter is an indication of the adsorption equilibrium, and the decline in removal at a high dose could have resulted from the aggregation of adsorbent particles [45]. At low adsorbent dosages, however, the adsorbed amount ( $q_e$ ) reduced dramatically in the case of Cs. The optimum adsorbent doses of  $\text{Cs}^+$  and  $\text{Co}^{2+}$  were 0.1 g for  $\text{Cs}^+$  and 0.5 g for  $\text{Co}^{2+}$  ions. Statistically, F (for weight) was 0.041 at  $p = 0.998$  ( $> 0.05$ ) non-significant.

The data are means  $\pm$  SE ( $n = 5$ ). A significant difference F (loading weight) was 0.041 at  $p = 0.998$  ( $> 0.05$ ) non-significant.

### 3.5. Comparison of the Study Findings with Other Similar Published Work

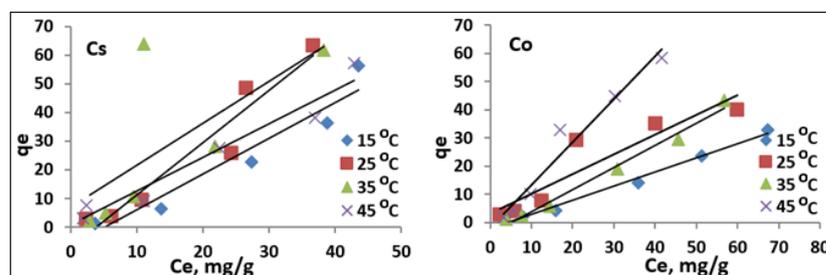
The economic and ecological features of the adsorption technique, various adsorbents have been studied in the literature. Some of these adsorbents have been collected in Table 1, where they are compared with the mixed waste used in the current study to evaluate the adsorption capacity.

**Table 1.** Adsorption capacity of mixed waste relative to other adsorbents for both ions separately.

Material	Adsorption Capacity ( $q_e$ ) of Co	Literature
Kaolinite	0.92	[46]
Soil	1.50	[47]
Marine bacterium	4.38	[48]
Nedalco sludge	11.71	[49]
Eerbeek sludge	12.34	[49]
Coir pith	12.82	[50]
Brown seaweed	20.63	[51]
<i>Myriophyllum spicatum</i>	43.40	[18]
Mixed waste	37.45	Current study
Adsorption Capacity ( $q_e$ ) of Cs		
Ceiling tiles	0.21	[52]
Coal and chitosan	3.00	[53]
Bure mudrock	13.30	[54]
Modified akadama clay	16.10	[55]
Kaolinite clay	17.10	[56]
Coir pith	32.00	[57]
Bentonite	40.00	[55]
<i>Myriophyllum spicatum</i>	58.00	[18]
Mixed waste	48.30	Current study

### 3.6. Adsorption Isotherm at Different Teperatures

At various temperatures, the adsorption behavior attained a maximum uptake value of 35 mg/g at 25 °C and 45 mg/g at 45 °C for both elements Cs<sup>+</sup> or Co<sup>2+</sup>, respectively, as shown in Figure 8. Due to the ratio of the interaction energies of the cations with water molecules between the layers and with the charges on the surface of the biomass layer, ions with a small radius, (Co), have lower competitive advantages over those with a large radius, (Cs) [58]. However, the change in the cation diffusion coefficient with increasing temperature is slightly indicated in the case of Cs.

**Figure 8.** Adsorption capacity ( $q_e$ ) of both ions separately on dry mixed waste at various temperatures.

The Langmuir adsorption isotherm implies the presence of a homogenous monolayer at all sorbent surface sites with no adsorbed molecules interacting with nearby adsorption sites. The Langmuir model was applied by Equation (4).

$$\frac{1}{q_e} = \frac{1}{k_L q_{\max}} \frac{1}{C_e} + \frac{1}{q_{\max}} \quad (4)$$

where  $q_e$  (mg/g) is the uptake at equilibrium concentration,  $C_e$  (mg/L), and  $q_{max}$  (mg/g) is the maximum number of ions required to create a monolayer. The linearized Langmuir adsorption isotherm was used to examine the equilibrium data, as shown in Figures 9 and 10. Table 2 shows the Langmuir constants,  $k_L$ , and monolayer sorption capacity,  $q_{max}$ , which were computed using the slope and intercept of the curve between  $1/q_e$  and  $1/C_e$ . The  $q_{max}$  values obtained from “Langmuir” plots do not agree with the experimental result [59].

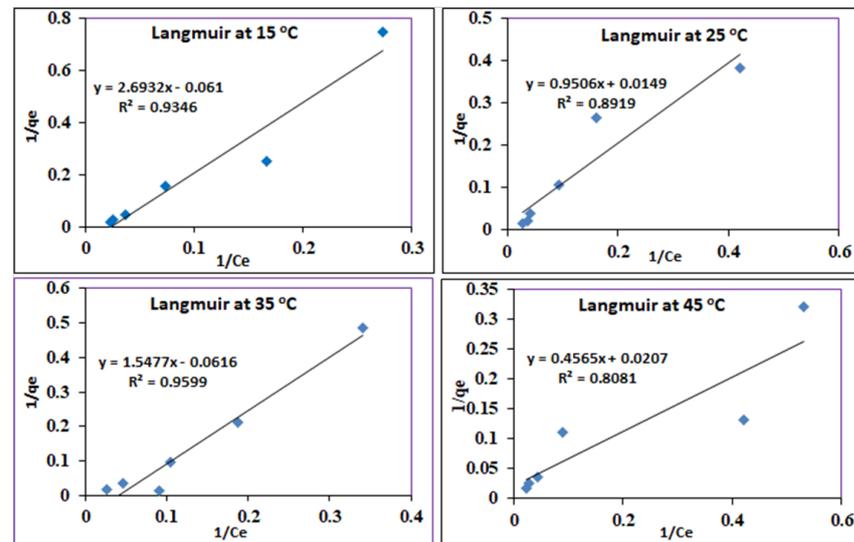


Figure 9. Adsorption of Cs<sup>+</sup> ions on mixed waste at different temperatures (Langmuir isotherm).

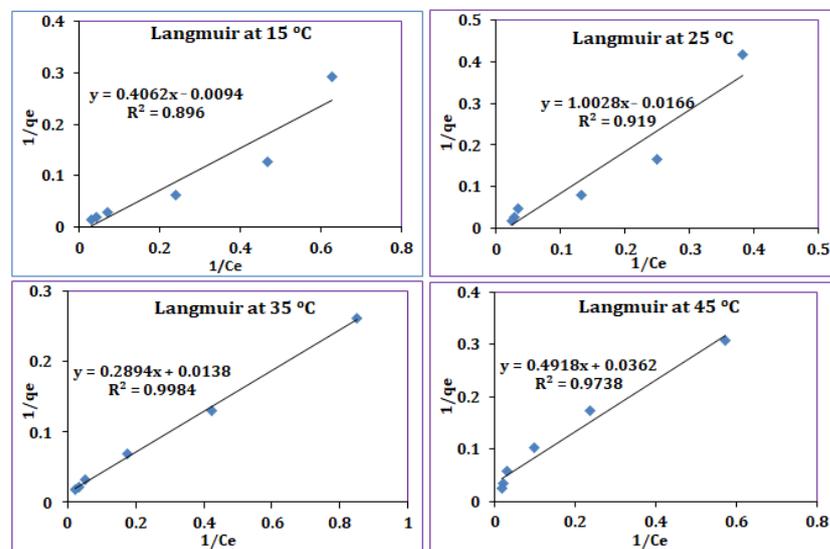


Figure 10. Adsorption of Co<sup>2+</sup> ions on mixed waste at different temperatures (Langmuir isotherm).

Table 2. Value of Langmuir parameters for adsorption of both ions separately on mixed waste.

Temp.	Cs <sup>+</sup>				Co <sup>2+</sup>			
	q <sub>e</sub>	q <sub>max</sub>	k <sub>L</sub>	R <sup>2</sup>	q <sub>e</sub>	q <sub>max</sub>	k <sub>L</sub>	R <sup>2</sup>
15 °C	43	16.3	0.022	0.934	67	21.55	0.021	0.896
25 °C	36	67.0	0.015	0.891	59	37.45	0.029	0.919
35 °C	38	16.2	0.039	0.959	56	21.23	0.013	0.998
45 °C	42	48.3	0.045	0.808	41	14.80	0.034	0.973

An empirical equation used to define heterogeneous schemes is the Freundlich isotherm. The “Freundlich” equation is represented as

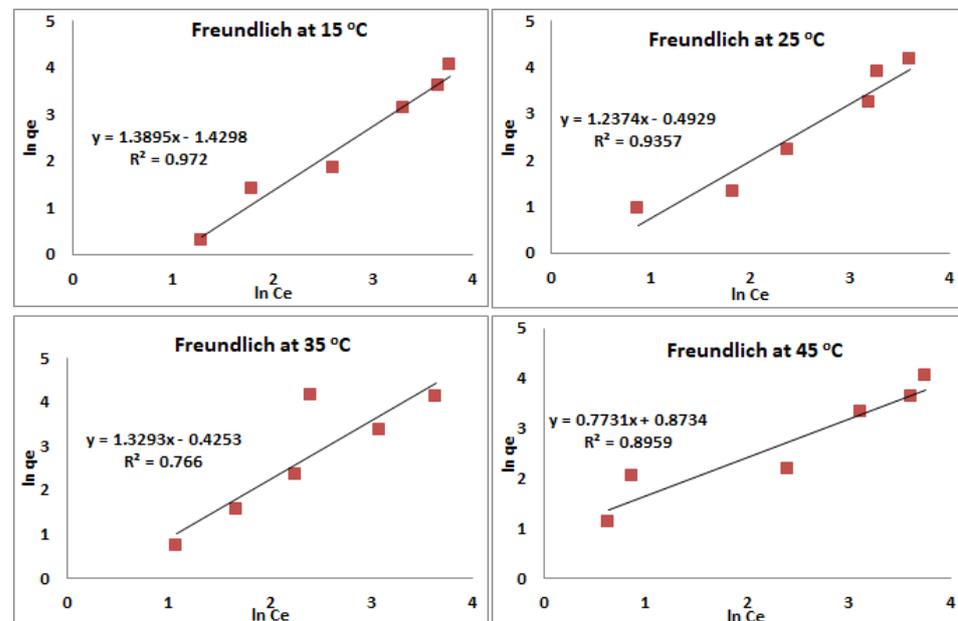
$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e \quad (5)$$

The Freundlich equation’s linear formula is given in Equation (5), where  $K_f$  and  $n$  are Freundlich constants, with  $K_f$  (mg/g (L/mg)<sup>1/n</sup>) denoting the adsorption capability and  $n$  denoting how beneficial the adsorption progression is. The adsorption favorability is determined by the extent of the exponent,  $1/n$ .  $n > 1$ , which indicates that the adsorption conditions are good. The intercept and slope of the curve illustrated in Figures 11 and 12 are used to estimate  $K_f$  and  $n$ , which are provided in Table 3.

The explanation behind the isotherm’s compatibility with the Freundlich isotherm model rather than the Langmuir isotherm model is that  $R^2$  was higher at Freundlich, which indicates that the adsorption process were taking place according to the Freundlich isotherm at all temperatures for  $\text{Co}^{2+}$  and  $\text{Cs}^+$  ions. In this study, the Freundlich model is more significantly correlated with high  $R^2$  than the Langmuir model, assuming that the stronger binding sites are occupied first and that binding strength decreases with the increasing degree of site occupation [60].

**Table 3.** Value of Freundlich parameters for adsorption of both ions separately on mixed waste.

Temp.	$\text{Cs}^+$			$\text{Co}^{2+}$		
	$n$	$K_f$	$R^2$	$n$	$K_f$	$R^2$
15 °C	0.719	0.239	0.972	0.926	0.298	0.957
25 °C	0.935	0.951	0.935	1.048	0.953	0.926
35 °C	0.750	0.653	0.766	0.734	0.166	0.995
45 °C	1.29	2.3	0.895	0.703	0.372	0.970



**Figure 11.** Adsorption of  $\text{Cs}^+$  ions on mixed waste at different temperatures (Freundlich isotherm).

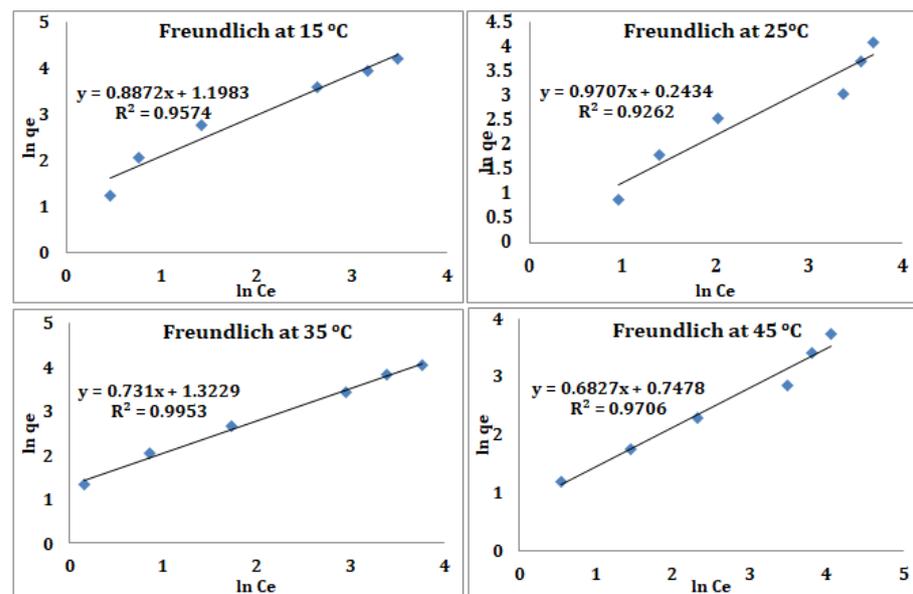


Figure 12. Adsorption of  $\text{Co}^{2+}$  ions on mixed waste at different temperatures (Freundlich isotherm).

To acquire the thermodynamic parameters of the adsorption reaction,  $k_f$  values were processed at different temperatures according to van't Hoff Equation (6):

$$\ln k_f = \frac{-\Delta H^0}{RT} + \frac{\Delta S^0}{R} \quad (6)$$

where  $\Delta H^0$  ( $\text{KJ}\cdot\text{mol}^{-1}$ ) and  $\Delta S^0$  ( $\text{KJ}\cdot\text{mol}\cdot\text{k}^{-1}$ ) are enthalpy and entropy changes, respectively,  $R$  is gas constant. Plotting  $k_f$  against  $1/T$ , as shown in Figure 13, gives a straight line with a slope and intercept  $-\Delta H^0/R$  and  $\Delta S^0/R$ , respectively. The value of  $\Delta H^0$  and  $\Delta S^0$  were calculated and listed in Table 4.

The positive value of  $\Delta H^0$  corresponds to the endothermic adsorption process of  $\text{Cs}^+$  and the negative value of  $\Delta H^0$  corresponds to the exothermic adsorption process  $\text{Co}^{2+}$  ions. The Gibbs free energy of adsorption was estimated from the following relation Equation (7) Table 4.

$$\Delta G^0 = \Delta H^0 - T\Delta S^0 \quad (7)$$

Table 4 indicated that little change in  $T\Delta S^0$  has been conducted at all temperatures for  $\text{Cs}^+$  ion and  $T\Delta S^0 < \Delta H^0$ . This finding implied that enthalpy, rather than entropic change, is the driving force of adsorption. These findings are consistent with prior research indicating that the adsorption process is non-spontaneous due to positive  $\Delta G^0$  values obtained [40]. Result for  $\text{Co}^{2+}$  show that  $T\Delta S^0 > \Delta H^0$  suggest an entropic rather than an enthalpic change in the adsorption process. The positive value of  $\Delta G^0$  obtained indicated that the adsorption process of  $\text{Cs}^+$  ion is nonspontaneous.

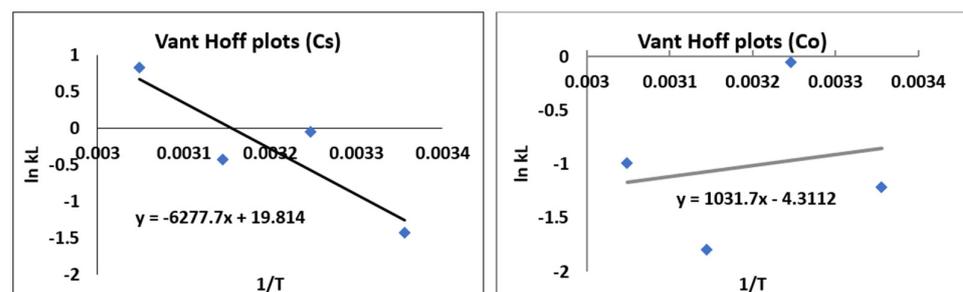


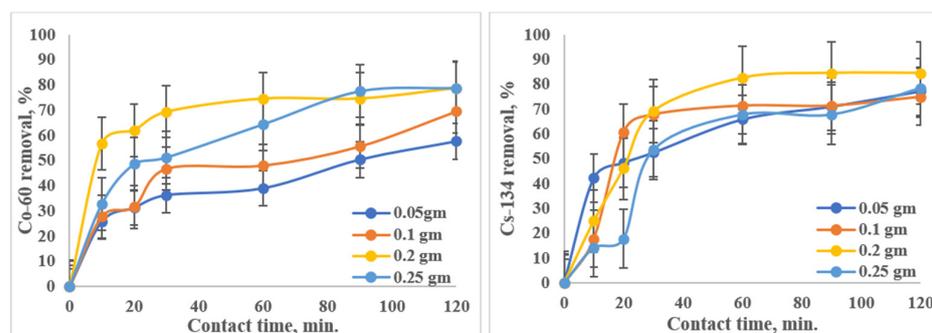
Figure 13. Van't Hoff plots related to adsorption of both ions separately on mixed waste.

**Table 4.** Thermodynamic parameters of adsorption of both ions on mixed waste.

Element	$\Delta S$	$\Delta H$	T(K)	T $\Delta S$	$\Delta G$
$\text{Co}^{2+}$	−0.035	−8.5	288	−10.08	1.58
			298	−10.43	1.39
			308	−10.78	2.28
			318	−11.13	2.63
$\text{Cs}^+$	0.16	52.1	288	46.08	6.02
			298	47.68	4.42
			308	49.28	2.82
			318	50.88	1.22

### 3.7. Adsorption of Radioisotopes ( $^{60}\text{Co}$ and $^{134}\text{Cs}$ ) on Dry Mixed Waste

Figure 14 depicts the adsorption of radioactive cesium and cobalt; the removal efficiency increases with time until it reaches a constant value at equilibrium. The removal percentage of  $^{60}\text{Co}$  after 120 min with different weights of dried mixed waste (0.05, 0.1, 0.2, and 0.25 g) was 55, 70, 80, and 80%, respectively. While the removal percentage of  $^{134}\text{Cs}$  after 120 min with different weights of dried mixed waste (0.05, 0.1, 0.2, and 0.25 g) was between 78.5 and 84.6%. Because there are no chemical differences between stable and radioactive elements, the same adsorption pattern was produced as in a previously reported study of the stable and radioisotopes [18]. With the difference in the radii of cesium and cobalt elements, the ability to impact diffusion mobility and to improve the adsorption process is different, and consequently, the diffusive mobility of the large radius of  $^{134}\text{Cs}$  from the solution to the surface of the biosorbent in aqueous media is lower than the diffusive mobility of the small radius of  $^{60}\text{Co}$  [61].

**Figure 14.** Adsorption of  $^{134}\text{Cs}$  and  $^{60}\text{Co}$  separately on mixed waste.

## 4. Conclusions

The novelty in this study lies in a sustainable treatment technology because it employs dry biomass from mixed waste to demonstrate its synergistic benefits, such as high efficiency and cost effectiveness.

In this study, the removal of stable and radioisotopes of cobalt and cesium from aqueous solutions by biosorption in dried mixed waste of olive waste and water hyacinth as a low-cost and natural available sorbent was investigated. The results show that the natural biomass of mixed waste is an excellent biosorbent for the examined isotopes. The pH experiments showed that the significant biosorption takes place in the acidic range in the case of Co and in the neutral medium in case of Cs. Contact times of 50 and 90 min were the optimal times for the maximum biosorption efficiency of  $\text{Cs}^+$  or  $\text{Co}^{2+}$ . Generally, the increase in mass biosorbent leads to the increase in biosorption due to an increase in the number of biosorption sites. Experimental data were better described by the pseudo-second

order model. The adsorption isotherm could be well-fitted to the Freundlich isotherm model. The biosorption capacity decreases with an increase in solution temperature.

Based on the experimental and mathematical data obtained, mixed dry waste of water hyacinth and olive waste could be nominated as a sustainable low cost and efficient natural adsorbent material suggested for the efficient remediation of radio or stable cobalt and cesium ions from wastewater. However, this technology is predicted to achieve the successful decontamination of hazardous metals and radionuclides in an environmentally friendly and sustainable manner with the extensively decreasing the cost of wastewater treatment.

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