Chapter

Removal of Heavy Metals from Wastewater with Special Reference to Groundnut Shells: Recent Advances

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Abstract

Wastewater contains organic pollutants and heavy metals which presents a significant threat to aquatic life and impacts human health and animals. In the past few years, the incomplete remediation of wastewater has made living beings suffer from various problems, and many health diseases are being noticed at a peak rate. Different methods have been employed to remove heavy metals from wastewater to date. However, the adsorption technique is the most efficient and eco-friendly for removing heavy metals and pollutants in wastewater remediation. Many agricultural wastes have been used as adsorbents for removing toxic pollutants and heavy metals from wastewater. Groundnut shell is widely considered agro-industrial waste. Groundnut shells account for nearly 20% of the dried peanut pod by weight, and millions of tons of its quantity are wasted every year. An increase in groundnut production leads to accumulating these groundnut shells in colossal quantities, which is not utilized; thus, they are either burnt or buried. Groundnut shells undergo slow degradation in the natural environment because they are rich in lignin content. Therefore, these shells can be converted into a valuable bio-product to produce less waste. Groundnut shells and groundnut shell-derived biochar act as good biosorbents in the wastewater treatment.

Keywords: heavy metal, groundnut shell, wastewater treatment, adsorption, biochar

1. Introduction

To begin with, freshwater is a prerequisite for human beings as well as animals. In addition, availability of clean water for maintaining a healthy life is a critical factor. However, different water sources have become polluted with increased global water demand [1]. Furthermore, researchers have found that the impacts of climate change, such as rise in temperature and water cycle changes, also aggravate the water issues, which potentially result in floods, severe droughts, and contamination of water bodies with various pollutants and toxic heavy metals [2–4]. The contaminated water sources can be harmful and perilous to humans as well as wildlife, also the aquatic life is negatively affected [4]. However, in developing countries, human health is mainly affected by the direct consumption of contaminated water because they are destitute in the efficient technologies for remediation of water sources, resulting in increased water pollution.

The World Health Organization (WHO) estimates that nearly 844 million population lack primary drinking water sources out of which approximately 230 million people spend more than 30 min/d for collecting water from an improved water source, which includes piped water, protected wells and springs, boreholes, rainwater and stored water [5, 6]. The lacking of efficient water sources and its accessibility in developing countries increases waterborne ailments. According to WHO, approximately 1.6 million people succumb to curable waterborne diseases, of which 90% of deaths are of children under 5 years of age [7].

Landfilling and mining sites have become the main reason for the introduction of heavy metals into freshwater bodies, eventually leading to water pollution. Due to their toxic nature, these heavy metals can bioaccumulate in living cells [8]. There are many conventional methods and technologies for removing heavy metals from an aqueous environment to protect human beings, wildlife, and aquatic life. However, their utilization for remediation is confronted with high costs, waste disposal issues, and effectiveness in removal. However, adsorption is a unique and promising technique and has been found as one of the most suitable methods for the removal and recovery of heavy metals from water economically and efficiently [9].

Groundnut shell is a cheaper agricultural waste and occupies an important position in agricultural waste and biomass because it is produced in larger quantities worldwide. Groundnut shell and groundnut shell-derived biochar play a crucial role in eliminating heavy metals from water sources [10, 11]. This chapter highlights the results of several scientific studies illustrating the efficacy of groundnut shells for the removal of heavy metals from wastewater. Furthermore, the chapter also discusses the characteristics of groundnut shell-derived biochar.

2. Chemical characteristics of heavy metals

Heavy metals contaminate water sources through various industrial activities, agricultural practices, and improper waste disposal [12]. Heavy metals are non-biodegradable in nature and accumulate in humans and animals when they consume contaminated food and water. Many researchers have investigated the contamination of water sources with heavy metals [13]. **Table 1** provides the characteristics of some common heavy metals found in a water sources.

Among the heavy metals, arsenic, cadmium, and lead were extensively studied for their toxicity in water sources. Studies have estimated various effects of heavy metals in drinking water [14, 15]. Arsenic and cadmium have been considered cancer-causing agents in humans [16]. Arsenic also leads to skin damage. It has also been investigated that 1 L of drinking water contaminated with 50 μg of arsenic can cause liver, kidney, bladder, and lung cancer. Lead has been examined to affect the central nervous system, cardiovascular system, gastrointestinal system, reproductive system, neurological system, renal system, endocrine system, and immune system [14]. Exposure to the low concentration levels of lead can also reduce neurodevelopment. The presence of lead in bloodstream alters the functioning of neurons and interferes

Heavy metal	Human health effects	Common sources
Arsenic (As)	Skin damage and circulatory system problems	Electronics production
Cadmium (Cd)	Kidney damage, causing cancer	Chemical industries
Chromium (Cr)	Skin allergies, nausea, vomiting, diarrhea	Steel manufacturing industries
Copper (Cu)	Gastrointestinal damage, liver damage	House plumbing systems
Lead (Pb)	Kidney damage, neurodevelopment problems	Lead-based products
Mercury (Hg)	Nervous system damage	Fossil fuel combustion

Table 1.
Characteristics of common heavy metals [13].

Heavy metal	Molecular weight (g mol-1)	Oxidation state	Van der Waals radius (10–12 m)	Electronegativity (Pauling Scale)
Arsenic (As)	74.9	−3, +3, +5	119	2.18
Cadmium (Cd)	112.4	+2	158	1.69
Chromium (Cr)	52	0, +2, +3, +6	200	1.66
Copper (Cu)	63.5	+1, +2	140	1.9
Lead (Pb)	207.2	+2, +4	202	2.33
Mercury (Hg)	200.6	+1, +2	155	2

Table 2.
Chemical properties of heavy metals [13].

with the trimming and pruning of synapses during brain development, which may result in permanently altered functions [14].

Cadmium is the most often reported heavy metal in the water sources [14, 15, 17]. Cadmium-contaminated drinking water leads to chronic renal failure, anemia, cardiovascular diseases, osteoporosis, and also hypertension [14]. There are also other heavy metals that can also put a living being in jeopardy. Researchers have estimated that 1 L of drinking water contaminated with nearly 8.29–51 μg chromium can cause liver, lung, and kidney cancer and also affect genitourinary organs among women [12]. Studies have shown that the stagnation of water in hot water tanks and polypropylene water pipes can significantly increase the concentration of various heavy metals. **Table 2** provides the chemical properties of common heavy metals.

3. Effect of the parameters of water on heavy metal removal

There are various water quality parameters that play an important role in the removal of heavy metals. The most important among them are; pH, temperature, natural organic matter (NOM), and ionic strength.

3.1 Effect of pH

The pH of the water source significantly affects the presence of heavy metals and their properties. The formation of heavy metals in water sources is primarily affected by pH. Heavy metals are cationic in nature at neutral to low pH and possess more solubility and mobility in an aqueous solution. pH also affects the surface charge, ionization state, and the concentration of ions on the functional groups of the adsorbent [18]. Several studies have reported the effect of pH on the formation of heavy metals and their removal. The stability and mobility of copper in an aqueous solution have been reported to increase with a decrease in pH value [19].

With the increase in pH, heavy metals form complexes with hydroxyl ions which in turn affect the oxidation state of the heavy metals. Heavy metals tend to form solids when the pH of the aqueous solution exceeds its neutral value and gets precipitated from the solution. The oxidation state for chromium (Cr) in its stable form is Cr(III), and it changes to Cr(IV) with the increase in pH in this oxidation state, chromium has been found to be more toxic [20]. Lower pH (<4) increases the concentration level of H^+ ions in the aqueous solution, which interferes with soluble metal ions and adsorbent surface interaction, thus reducing overall heavy metal removal [21, 22]. However, with the increasing pH (between 5 and 7), adsorption increases, and the adsorbent surface becomes more negatively charged and interacts readily with the positively-charged heavy metals [23].

The probability of removing maximum heavy metal ions by adsorption is minimal at lower pH values (<3) [24]. Furthermore, with the increase in pH, the concentration of H^+ ions is decreased, and a greater number of adsorption sites become available for heavy metal ions for adsorption, which thereby increases the heavy metal removal from water sources [18]. In the case of chromium removal, it becomes anionic in nature as the pH increases (e.g., $HCrO_4^-$, CrO_4^{2-}). Hence, the adsorption of chromium has been shown to decrease with the increasing pH of the solution. This is mainly due to the electrostatic repulsion resulting from negative surface charges on the adsorbent, inhibiting chromium adsorption [25, 26]. Overall, pH is a significant parameter that affects the behavior and removal of heavy metals from water sources.

3.2 Effect of temperature

Temperature is another important parameter affecting heavy metals' behavior and subsequent removal from water sources. Mechanisms employed for the removal of heavy metals are enhanced at higher temperatures in which surface complexation reactions and various forms of ion exchange are also included [27]. It has been reported that an increase in the removal of Cr(VI) using hull wastes is achieved with an increase in the temperature from 5 to 40°C, which is attributed to the formation of additional adsorption sites on the surface of the adsorbent [28].

The adsorption process has been observed to increase with the increase in temperature due to the increased driving force of diffusion across the boundary layer and an increased rate of diffusion within the adsorbent [29]. However, in various cases, increased temperatures have also been observed to result in a decrease in the heavy metal removal from the water sources. For example, in one experimental study, the removal of total chromium by red algae has been reported to reduce from 90 to 78% with the increase in temperature, which has been possibly observed due to the tendency for ions to remain in the aqueous phase [30]. Furthermore, researchers have also reported a reduction in the heavy metal removal, such as Pb(II) and Ni(II) with increasing temperature, which was attributed to decreased surface activity [18, 31].

An experimental study had reported an increase in the adsorption of Cr(III) and Cu(II) onto peanut shells as the temperature increased to 50°C, and a decrease in the adsorption when the temperature increased to 60°C, which has been observed due to potential damage to the adsorption sites on the peanut shells [32]. Another study evaluated a 32% decrease in the adsorption capacity of the olive cake with the increased temperature from 28 to 45°C in the removal of Cd(II) from an aqueous solution [33]. Therefore, it concluded with a fact that when estimating the effects of temperature on the removal of heavy metals from water sources, each adsorbent, and the corresponding metal ion must be explicitly evaluated in order to determine the overall impact of temperature changes on the adsorption process [13].

3.3 Effect of ionic strength

The ionic strength of the water source has also been reported to significantly affect the removal of heavy metals. It has been observed that the presence of chloride in water sources can form neutral or negatively-charged chloride complexes that have a low affinity for adsorption. These complexes are soluble and difficult to remove from water. With the increase in the formation of chloride complexes, ionic strength tends to increase due to a decrease in the removal efficiency of Cu(II) and Ni(II) [34]. Researchers have also observed that increased salinity and increased concentration of dissolved metals, such as copper, cadmium, and zinc, possess a strong correlation [35].

Interactions between heavy metals and other surfaces are strongly affected by electrostatic forces, and increased ionic strength in a solution has a significant effect on the behavior and removal of heavy metals [13]. Zhang (2011) investigated the impact of ionic strength on heavy metal removal, including Cu(II), Pb(II), and Zn(II), by using dairy manure compost and reported a decrease in the removal of heavy metals with the increase in ionic strength [36]. However, various researchers have estimated that heavy metal removal increases with the increase in ionic strength. For example, Yang et al. (2016) reported a 25% increase in the removal of As(III) and Ni(II) with the increase in ionic strength from 0.01 to 1 M Cl^- ions of the solution due to the formation of an inner-sphere surface complex [37].

3.4 Effect of natural organic matter

Natural organic matter consists of humic and fulvic acids that are formed by the decomposition of plant and animal matter [38]. Natural organic matter is a complex mixture of organic compounds and is highly reactive with heavy metals, which alter the reactivity of the heavy metals and affect their mobility, bioavailability, and toxicity [38]. The particular impact of natural organic matter on heavy metals can be difficult to discover, primarily due to the wide mixture of additional factors, such as pH, humification of a particular natural organic matter, and oxidation state of heavy metals that contribute to the manner in which natural organic matter affects heavy metal removal [19]. Arsenic has been found to form complexes with both humic and fulvic acids, which leads to an increase in the immobilization of arsenic [39].

Metals, such as copper and zinc, have been observed to form complexes with natural organic matter [35]. An experimental study has reported that the removal of Cd(II), Pb(II), and Zn(II) by mollusk shells is increased by the presence of organic matter [40]. Moreover, the presence of natural organic matter can reduce the toxic form of chromium, Cr(VI), to its less harmful and more stable form, Cr(III) [19]. However, natural organic matter can reduce arsenic from its less toxic form, As(V), to its more toxic and mobile form, As(III) [19]. These research studies estimate that natural organic matter can reasonably affect heavy metal removal from water sources.

4. Groundnut shells in heavy metal adsorption

Groundnut shell is an abundant agro-industrial waste product that is mainly obtained after the removal of groundnut seed from its pod. Groundnut shell is rich in lignin content, due to which it undergoes very slow degradation [41]. Groundnut shell plays a vital role in the adsorption of heavy metals from water sources. Groundnut shell contains cellulose, organic acids, lignin, pentosan, and tannins which aid in binding heavy metal ions [42]. Efficiency of groundnut shells in heavy metal removal from water sources has been estimated by various researchers. For example, in an experimental study, the efficacy of groundnut shells for removing heavy metals from wastewater was tested by using several parameters at a standard temperature, such as the effect of pH, contact time, and dosage of groundnut shell. It has been found that the removal efficiency of groundnut shells for copper and lead is 68.19 and 77.81%, respectively [43].

Groundnut shell has been proven to be an efficient and effective adsorbent for the removal of toxic metals from zinc and chromium-plated water in which the initial concentration of zinc and chromium has been observed to reduce to the values 0.75 and 0.85, respectively. It has been concluded with the manifestation that the ability to remove toxic metals from zinc and chromium-plated water can be effectively achieved by using a groundnut shell as the adsorbent [44].

Groundnut shell has been utilized for removing copper from a water source, using batch experiments and considering different parameters, such as pH, contact time, initial concentration of metal ion, and dosage. The efficiency of groundnut shells at pH 6 for removing copper metal from water sources has been found to be approximately 94% [45]. Groundnut shell-activated carbon can also be used for the adsorption of metal ions such as Cu (II), Zn (II), Ni (II), and Cr (IV) from the aqueous solution at different parameters, such as pH and metal ion concentration of the aqueous solution. It has been estimated that the groundnut shell activated carbon has the highest adsorption capacity for removing chromium ions than its adsorption capacity for other metal ions from water sources [46].

Groundnut shells can also be positively utilized for the removal of lead ions from the aqueous solution. The adsorption capacity of groundnut shells for lead ions has been estimated at nearly 39 mg/g. Significant removal of lead ions by groundnut shells from water sources has been observed at different temperature conditions and different pH values [18]. The maximum adsorption capacity of groundnut shells at low pH values for chromium (VI) has been estimated at around 4.3 mg/g [25].

The effective removal of chromium(III) and copper(II) using groundnut shells has been evaluated at the maximum adsorption capacities of 27.89 and 25.39 mg g-1, respectively [32]. Heavy metal removal by using groundnut husk has also been significantly observed with maximum adsorption capacities of 7.69, 10.21, and 29.11 mg g-1 for Cr(III), Cu(II), and Pb(II), respectively [47]. The maximum adsorption capacity of groundnut shells for copper, nickel, and zinc has been calculated as 5, 8.1, and11 mg/g, respectively [48]. These adsorption capacities have been increased by 40–70% by using reactive dye in chemically modifying the groundnut shells using [48].

Groundnut shells can be effectively used as a raw material for preparing activated carbon to remove chromium and nickel from dye effluents based on the contact time. It has been observed that groundnut shell-activated carbon leads to an increase in chromium and nickel adsorption with an increase in contact time [49].

The adsorption of heavy metals, such as copper, zinc, lead, manganese, and cadmium, from wastewater by groundnut shells has been observed at a concentration

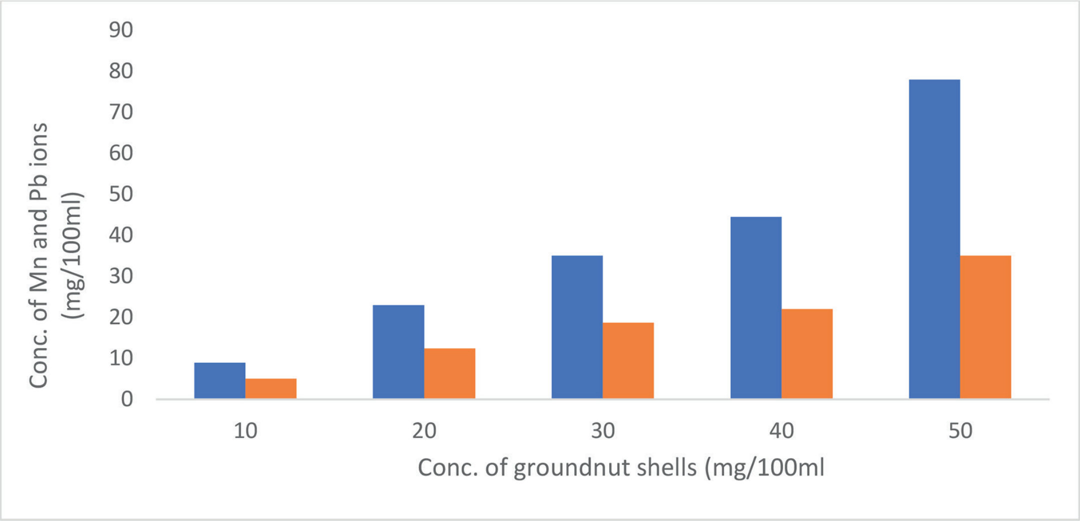

Figure 1.
Mn(II) and Pb(II) adsorption by using groundnut shells at different concentrations [50].

range of 10–50 mg/100 ml [50]. It has been evaluated that at a contact time of 100 min, maximum adsorption of groundnut shells for lead and manganese can be found. Manganese(II) and lead(II) adsorption from wastewater by using groundnut shells at different concentrations has also been observed at different concentrations at 33°C for 90 mins (**Figure 1**).

From the **Figure 1**, it is clearly observed that the adsorption of Mn(II) and Pb(II) increases with the increase in the concentration of groundnut shells [50].

5. Groundnut shells derived biochar in heavy metal adsorption

Groundnut shell-derived biochar has been used as an alternative cost-effective adsorbent to remove various heavy metals from water sources. The surface of groundnut shell biochar possesses functional groups, such as COOH and OH [51, 52]. Biochar with high efficiency can be derived from groundnut shell since no pre-treatment of biochar surfaces for adsorption is needed [53]. The production of biochar from groundnut shells by pyrolysis has been considered an eco-friendly and efficient method for the disposal of agricultural solid waste [54]. Groundnut shell biochar produced under slow and fast pyrolysis temperatures has been observed to possess a strong affinity for the removal of heavy metals from water sources. Various research experiments have been carried out to determine the adsorption efficiency of groundnut shell-derived char for different heavy metals at different parameters, such as pH of the aqueous solution, constant biochar dosage, particle size, concentration levels of heavy metal ions, constant contact time, and temperature [53].

Groundnut shell-derived biochar has been utilized for the removal of heavy metal ions from the aqueous solution at 350 ± 5°C and 700 ± 5°C. The adsorption efficiency of groundnut shell biochar for the removal of cadmium ions from the aqueous solution with concentration levels of 0.04, 0.08, and 0.20 mg/l has been estimated at 100, 99.99 and 100% at pyrolysis temperature of 350 ± 5°C, respectively. The adsorption efficiency of groundnut shell biochar for cadmium with the same concentration levels at a pyrolysis temperature of 700 ± 5°C has been estimated 100, 99.67, and 99.93%, respectively (**Table 3**). Groundnut shell biochar adsorption efficiency has been found approximately greater than 99.60% for cadmium [53]. A similar research study has reported 99.2% cadmium removal efficiency from wastewater at room temperature by using 40 g/l of adsorbent dosage, 200 mg/l of initial concentration of cadmium and pH 5 [55].

The adsorption efficiency of lead ions has been calculated 99.12% at the concentration of 0.10 mg/l, 100% each at 0.20 and 0.50 mg/l at pyrolysis temperature 350 ± 5°C, respectively, while the adsorption efficiency of lead ions in aqueous solution with same concentrations has been calculated 100% at pyrolysis temperature 700 ± 5°C was 100% (**Table 3**). The removal efficiency of lead ions in a mono-component system by groundnut biochar has been found to be higher than 99.1% [53].

The removal efficiency of mercury ions in the aqueous solution with a concentration of 0.10, 0.20, and 0.50 mg/l at 350 ± 5°C has been calculated 99.99, 100 and 100%, respectively (**Table 3**), while removal efficiency of mercury by groundnut shell biochar with same concentration levels of mercury ions has been estimated 100% at high pyrolysis temperature (**Table 3**). The removal efficiency of Hg^{2+} by low and high pyrolysis temperatures in mono-component systems with different maximum contamination limits has been reported to be almost 100% [53].

It has been found that groundnut shell-derived biochar at 400°C exhibits the highest capability for the removal of heavy metals from water sources because of its high specific surface area and rich functional groups [54]. The physicochemical properties of groundnut shell biochar prepared at varying pyrolysis temperature is shown in **Table 4**. With the temperature elevation from 350 to 400°C, the surface area and total pore volume has been observed to increase from 3.77 to 6.45 m^2/g and from 0.0097 to 0.0161 cm^3/g, respectively [54]. However, as the temperature increases more than 400°C, surface area and pore volume have been found to decrease. This phenomenon has been attributed to too-high temperatures that accelerated the deformation and collapse of micropores in biochar. Therefore, groundnut shell biochar at 400°C has the highest surface area and pore volume [56]. With the increase in temperature from 350 to 600°C, the pH value of the produced biochar has been observed to increase from 9.11 to 10.35, and the biochar yield has declined from 47.9 to 33.6%. The rise in pH has been observed by the decomposition of organic acid and carbonate into alkali

Metal ions	**Initial concentration (mg/l)**	**350 ± 5°C**	**700 ± 5°C**
		Adsorption efficiency (%)	
Cd^{2+}	0.04	100	100
	0.08	99.99	99.67
	0.20	100	99.93
Pb^{2+}	0.10	99.12	100
	0.20	100	100
	0.50	100	100
Hg^{2+}	0.10	99.99	100
	0.20	100	100
	0.50	100	100

Table 3.
Adsorption efficiency of groundnut shell biochar for Cd^{2+}, Pb^{2+}, Hg^{2+} ions [53].

Pyrolysis temperature (°C)	Surface area (m^2/g)	Total pore volume (cm^3/g)	pH	Yield (%)
350	3.77	0.0097	9.11	47.9
400	6.45	0.0161	9.34	41.4
500	4.78	0.0156	9.58	36.3
600	4.36	0.0153	10.35	33.6

Table 4.
The physicochemical properties of groundnut shell biochar prepared at varying pyrolysis temperatures [54].

salts. Therefore, it is beneficial for the heavy metal ions to precipitate in an alkaline environment [54].

The initial concentration of the metal ions in an aqueous solution has been considered to play an important role in the removal of metal ions in aqueous solution by the groundnut shell biochar. For example., the removal of Pb(II) ions in an aqueous solution has been reported to decrease with the increasing initial concentration of Pb(II) ions from 100 to 400 mg/l [57]. The experimental results are shown in **Figure 2**.

Similarly, pH of the solution is considered as one of the most important parameters in the removal of heavy metals from aqueous solutions. For example, the effect of pH on the adsorption of Pb(II) ions on the biochar derived from the groundnut shell has been carried out by changing the initial pH of the solution from 2 to 6. The removal of Pb(II) ions has been evaluated to increase with the increasing pH. The removal of Pb(II) ions by the groundnut shell biochar has been found to be very low at a pH value of 2.0. The increase in pH has been reported to promote the removal of heavy metals [54]. The effect of pH on the removal of Pb(II) ions from the aqueous solution by groundnut shell biochar is shown in **Figure 3**.

From **Figure 3**, it can be concluded that the pH plays a vital role in the removal of Pb(II) ions from the aqueous solution by groundnut shell-derived biochar. With an

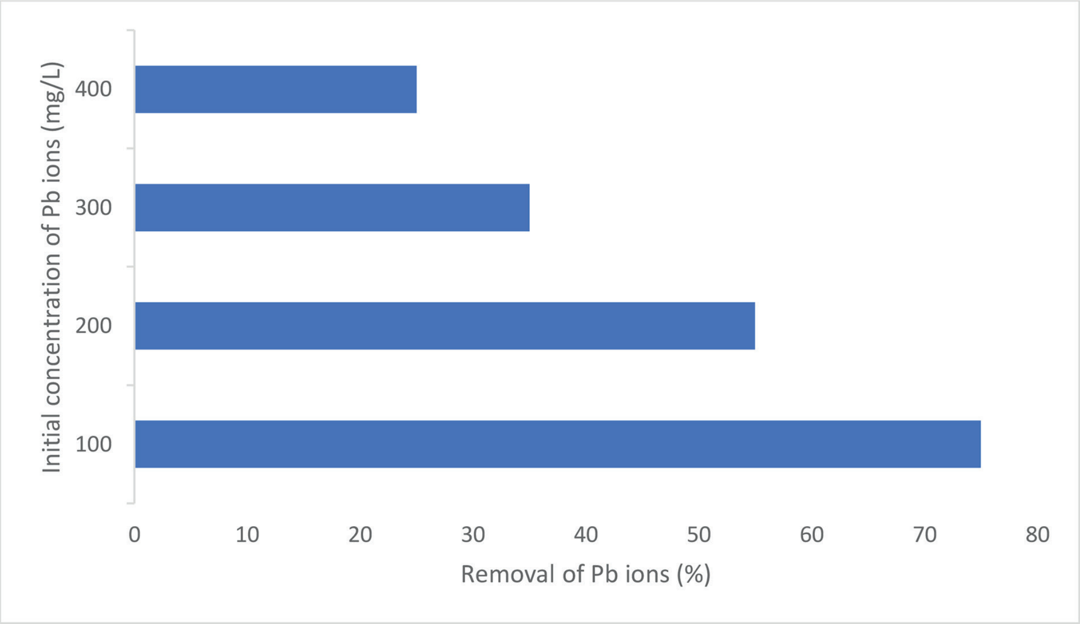

Figure 2.
Effect of initial concentration of the Pb(II) ions in aqueous solution on the removal of lead ions by the groundnut shell biochar.

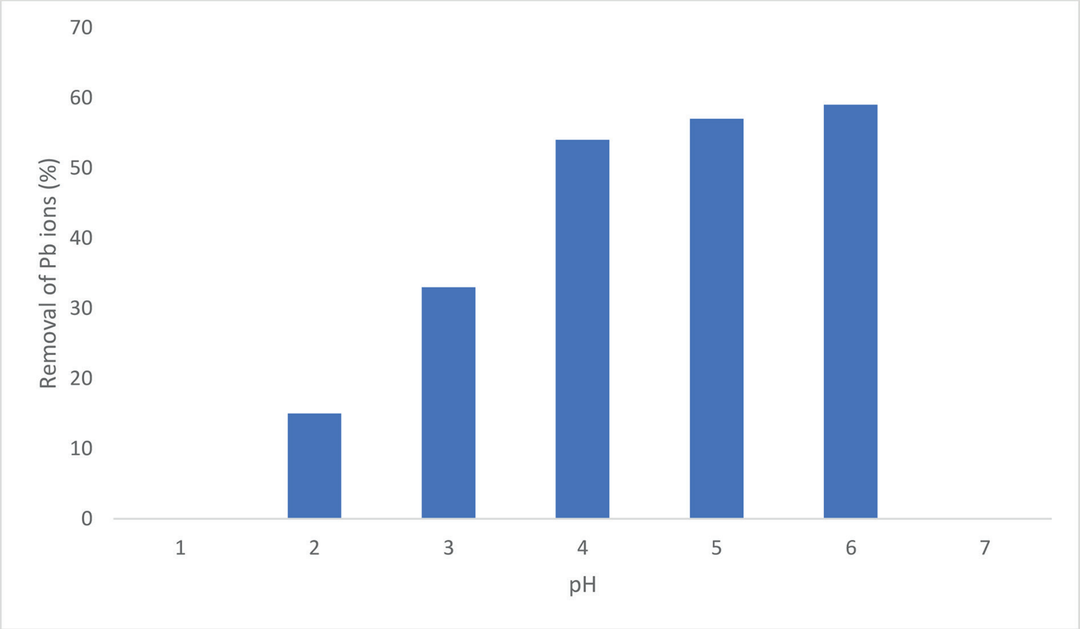

Figure 3.
Effect of pH on the removal of Pb(II) ions from the aqueous solution by groundnut shell biochar [54].

increase in the pH value, the covered H_3O^+ leaves the biochar surface and makes the sites available to Pb(II) ions [57]. Some studies have also reported that pH-dependent surface charges play a crucial role in controlling the surface adsorption of heavy metal ions through electrostatic interactions [58].

6. Conclusions

In developing countries, the increase in water scarcity and pollution significantly leads to less accessibility to clean drinking water. Heavy metal contamination in drinking water sources is a growing concern in the present era. Moreover, there is a lack of wastewater treatment methods in developing countries that would remove heavy metals from wastewater. As a result, various research studies have been conducted to investigate the use of low-cost adsorbents to remove heavy metals from water sources.

Groundnut shell is generally considered an agro-industrial waste; millions of tons of its quantity are produced every year as a leftover. Groundnut shell is rich in lignin, due to which these shells undergo slow degradation in a natural environment. Groundnut shell biomass has a wide range of applications. Groundnut shells can be converted into a valuable bio-product that can be efficiently and cost-effectively utilized in heavy metal removal from water sources. The chapter highlights the results of several scientific studies illustrating the adsorption efficiency of groundnut shells and biochar derived from the shell for the removal of various heavy metals from wastewater.

Various research studies have reported that groundnut shells can effectively remove heavy metals from water sources. The effectiveness of groundnut shells and biochar produced from groundnut shells at removing heavy metals depends on various parameters, such as pH, ionic strength, temperature, natural organic matter, initial concentration of heavy metal ions, etc. Groundnut shell-derived biochar exhibits a strong affinity for heavy metals in water sources at slow and fast pyrolysis temperatures.

Acknowledgements

The authors acknowledge the support of the Department of Post Harvest Engineering and Technology, Aligarh Muslim University, Aligarh.

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