

Review

Current and Emerging Adsorbent Technologies for Wastewater Treatment: Trends, Limitations, and Environmental Implications

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Citation: Younas, F.; Mustafa, A.; Farooqi, Z.U.R.; Wang, X.; Younas, S.; Mohy-Ud-Din, W.; Ashir Hameed, M.; Mohsin Abrar, M.; Maitlo, A.A.; Noreen, S.; et al. Current and Emerging Adsorbent Technologies for Wastewater Treatment: Trends, Limitations, and Environmental Implications. *Water* **2021**, *13*, 215. <https://doi.org/10.3390/w13020215>

Received: 2 December 2020
Accepted: 13 January 2021
Published: 18 January 2021

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Abstract: Wastewater generation and treatment is an ever-increasing concern in the current century due to increased urbanization and industrialization. To tackle the situation of increasing environmental hazards, numerous wastewater treatment approaches are used—i.e., physical, chemical, and biological (primary to tertiary treatment) methods. Various treatment techniques being used have the risks of producing secondary pollutants. The most promising technique is the use of different materials as adsorbents that have a higher efficacy in treating wastewater, with a minimal production of secondary pollutants. Biosorption is a key process that is highly efficient and cost-effective. This method majorly uses the adsorption process/mechanism for toxicant removal from wastewater. This review elaborates the major agricultural and non-agricultural materials-based sorbents that have been used with their possible mechanisms of pollutant removal. Moreover, this creates a better understanding of how the efficacy of these sorbents can be enhanced by modification or treatments with other substances. This review also explains the re-usability and mechanisms of the used adsorbents and/or their disposal in a safe and environmentally friendly way, along with highlighting the major research gaps and potential future research directions. Additionally, the cost benefit ratio of adsorbents is elucidated.

Keywords: adsorption; agriculture waste and peels; nanotechnology; biosorption mechanism; contaminant removal

1. Introduction

Water is an important natural resource; therefore, it must be preserved. As an important reserve for prevailing flora and fauna, it is necessary to prevent contamination via organic and inorganic pollutants. However, some technologies used for this purpose release secondary contaminants or byproducts which further pollute the environment [1,2]. Therefore, cost-effective and efficient wastewater treatment technologies are urgently needed [3,4]. Severe water scarcity is experienced throughout the world, highlighting the dire need for adequate food production throughout the year to fight hunger, deprivation, and malnutrition, requiring wastewater reuse for irrigation purposes [5]. Water reuse

through industrial wastewater recycling has gained the scientific community's interest in the last few decades. Wastewater recycling is of great advantage in farming activities because it contains an ample amount of nutrients [6], so its treatment followed by agricultural application should be conducted with great prudence to ensure it is environmentally friendly, economical, and increases agricultural output [5].

The challenge in treating wastewater is much greater than it seems. There are two primary sources of contaminants in wastewater: (i) natural, including but not limited to volcanic activity, soil erosion, and the weathering of rocks, and (ii) mineral contaminant dispersion through anthropogenic activities, waste disposal sites, urban runoff, mining, the manufacture of printed circuit boards, agricultural activities, the treatment and electroplating of metal surfaces, fuel burning, textile dyes, the manufacture of semiconductors, etc., [7,8]. Wastewater generated from agriculture, industries, and the household sector contains a varying amount of noxious inorganic (heavy metals and excessive nutrients) and organic (pigments, polyaromatic hydrocarbons, etc.) contaminants that pose serious environmental and health risks [9–12]. Among the heavy metals (potentially toxic elements or PTEs) and metalloids, PTEs belong to the group of trace elements with a density $> 4 \pm 1 \text{ g cm}^{-3}$. These include copper (Cu), mercury (Hg), cadmium (Cd), zinc (Zn), tin (Sn), iron (Fe), lead (Pb), silver (Ag), manganese (Mn), chromium (Cr), cobalt (Co), arsenic (As), aluminum (Al), and nickel (Ni) [13,14] (Supplementary Information Table S1). Due to their persistence, higher mobility, and solubility, wastewater containing these PTEs is not properly treated and discharged into freshwater resources with various environmental and health effects. Additionally, these PTEs are taken up by aquatic organisms, crops, and other plant species and make their way to the human food chain, thereby exerting negative impacts on human health [15–17].

Apart from the natural and anthropogenic sources, there are two major types of wastewater pollutants—i.e., organic and inorganic pollutants. Organic pollutants include pesticides, phenols, herbicides, petroleum, dyes, oils, biphenyls, fats, proteins, starches, and medicines, while inorganic pollutants contain chemical fertilizers, PTEs, and excessive nutrients. They cause water quality deterioration and serious environmental problems [18,19]. To reduce the environmental and health risks posed by wastewater, multiple technologies are used that are based on varying degrees of treatments, chemical reactions, and processes, such as membrane filtration, reverse osmosis, chemical precipitation, solvent extraction, oxidation, and adsorption [20–23].

Among all of the above techniques, sorption using different adsorbent materials is thought to be simpler to execute and manage and is cost-efficient [24,25]. In addition to the primary benefits, this process does not cause secondary pollution from the generation of byproducts [26–28]. Minerals and organic and inorganic materials that are commonly used as adsorbents (such as activated clay minerals, carbon, industrial byproducts, zeolite, polymer materials, bio-fuels, farming waste, etc.) have different adsorption capacities for specific pollutants' removal from wastewater [29].

Some researchers have modified adsorbent materials by combining them with other chemical and substances to enhance their capacity to adsorb pollutants. In recent developments, nanotechnology has been used in almost all areas of human interest, including environmental clean-up, soil, and natural resource management, especially wastewater treatment [30–32]. Many nano-adsorbents are used to remove organic and inorganic substances from wastewater, and, in many instances, they are even more effective than traditional adsorbents. These adsorbents involve metal oxide-based nanoparticles (NPs), carbon nanotubes (CNTs), graphite, plant nanocomposites (NCs), etc. [27,33,34]. This review aims to provide information about the major agricultural and non-agricultural material-based adsorbents. Their mechanisms of pollutant removal are also mentioned, along with possible ways to enhance their efficacy by modification or treatment with other substances. Moreover, this review also explains their prospects of re-usability and mechanisms of safe disposal.

2. Municipal and Industrial Wastewater: Sources and Reuse

Wastewater can be classified according to its sources of origin into two categories—i.e., industrial and municipal wastewater [35,36]. With the rapid increase in the population, the amount of wastewater generated has been amplified due to industrial and municipal activities [37]. The typical amount of wastewater generated through anthropological activities per day depends upon the accessibility of water, water costs, and the economic state [38]. Globally, wastewater discharge is 400 billion cubic meters/year, and it pollutes about 5.5 trillion cubic meters of water every year [39]. The type and quality of wastewater generated varies according to the region [39]. However, the current era's major problem is irrigation with wastewater [40], which causes soil and water body pollution [41]. With the increasing production of wastewater, its marginal treatment and productive use in agriculture also increase because farmers do not have alternative irrigation water sources [42].

Around 330 km³ of urban wastewater is generated worldwide every year, which could irrigate and nourish thousands of hectares of croplands. Currently, only a small fraction of wastewater is receiving treatment, and the proportion of this wastewater that is treated for irrigation purposes is nearly negligible. Food and Agriculture Organization (FAO) estimated that over 60% of all municipal sewage is treated worldwide before its reuse [43]. It is estimated that around fifty countries use wastewater to irrigate 20 million hectares of respective agricultural lands [44]. In a report from 2012, it has been summarized that around 68.5 million metric tons of municipal and industrial sewage sludge were produced, which corresponds to the Yellow River's annual stream. Additionally, according to estimates from the first census of national pollution sources, China produces approximately 108.16 billion cubic meters of sewage sludge each year (municipal 34.33 billion cubic meters and industrial 73.83 billion cubic meters) [43].

3. Environmental Implications of Wastewater Contamination

Numerous studies and efforts have been made for environmental protection against potentially toxic elements (PTEs). However, the United Nation's Sustainable Development Goals (UN-SDGs) 2030 also emphasize the promotion of good health and well-being and enhancing knowledge about resource conservation—i.e., soil, water, and the environment. The use of wastewater for irrigation purposes in those regions which lack good-quality water for this purpose is a good example of this. However, the regular use of wastewater for irrigation purposes can cause PTEs to build up in the soil and crops and cause human health effects. The main pollutants of wastewater include mineral nutrients, including nitrogen and phosphorus, PTEs, organic substances, and microorganisms [16]. Organic materials in wastewater are the main source of most pathogenic microbes such as protozoa, fungi, bacteria, and viruses that cause water-borne diseases [45].

Wastewater may also contain higher salt concentrations that accumulate in the root zone and affect soil properties, quality, and productivity. The long-term use of wastewater with high salt contents destroys the soil structure and affects soil productivity [43]. Soil salinization through wastewater irrigation has been reported frequently in the past few years [46]. It has also been stated that agricultural areas in Mexico and the United States which have been irrigated by wastewater for more than 50 years have poor soil health with a limited ability to support crop production [47–49].

The lack of proper instrumentation, facilities, and accessibility of wastewater treatment in developing countries is another reason for wastewater being used as irrigation without any treatment [50], thereby affecting approximately 245,000 km² of marine environments, fisheries, and substances ultimately entering food web [51]. In 2012, a report published by the WHO stated that around 842,000 deaths in low and middle-income countries were attributed to poor sanitation, water pollution, and insufficient essential nutrients. Such illnesses are mainly described in infants or children below 5 years of age [52]. Therefore, the world has paid great attention to wastewater treatment options, food security, and hazard assessment. Moreover, a better understanding of the prevention of waterborne diseases in most societies has also led people to utilize wastewater treatment rather than

using it without treatment. The reason for this is that wastewater treatment is a significant part of preventing and spreading waterborne and water-related diseases, environmental nuisances, eutrophication, waterlogging, salt emergence in agricultural soils, and human health effects due to the consumption of wastewater-irrigated food crops [53–56].

4. Treatment Approaches for Wastewater

Different techniques are used to remove pollutants from wastewater. Treatment processes include physical, chemical, and biological approaches [57]. Treatment practices play an important role in wastewater treatment and reuse. Some of these treatments include primary, secondary, and tertiary treatment. Primary treatments include initial screening and the sedimentation of larger particles from domestic, municipal, and industrial waste. Secondary treatment consists of aerobic and anaerobic lagoons or ponds where wastewater is treated aerobically and anaerobically. This procedure involves the microbial action, biodegradation, and bioremediation of treatable wastes. Tertiary treatment includes chemical treatment methods such as flocculation, coagulation, ion exchange, membrane filtration, and other methods. However, in some cases multiple treatment methods are employed in combination with other treatments. From the various recent water treatment technologies, adsorption remains the most effective method for removing pollutants in wastewater and water with low costs, simple operation, and simple design [58] (Table 1).

Table 1. Wastewater treatment processes and possible disadvantages.

Treatment	Disadvantage	Process Detail
Ion exchange	High cost, partial removal of certain ions	Metal ions from dilute solutions are exchanged with ions held by electrostatic forces on the exchange sites
Adsorption	Chemical regeneration requirement, fouling and adsorbent corrosion of plant, disposal of exhausted air	Molecular or atomic film is formed by the accumulation of gas or liquid solute on the surface of an adsorbent
Chemical precipitation	Sludge generation, extra operational cost for sludge	Precipitation of metal ions is achieved by the addition of coagulants, such as alum, lime, iron salts, and other organic polymer disposal
Reverse osmosis	High power consumption due to pumping pressure, the restoration of membranes	Metal ions are separated by a semi-permeable membrane at a pressure greater than internal osmotic pressure that is caused by the dissolved solids
Electrodialysis	High operational cost due to membrane fouling and energy consumption	Metal ions are separated using semi-permeable ion-selective membranes. An electrical potential between the two electrodes causes a separation of cations and anions, thus cells of concentrated and dilute salts are formed
Coagulation/flocculation	High operational cost due to chemical consumption and increased sludge volume generation	Coagulant is added to the water, which encourages colloidal material to join into small aggregates called “flocs”. Suspended matter is attracted to these flocs
Flotation	High initial capital cost, high maintenance and operation cost	Bubble attachment is used to separate solids or dispersed liquid phase

5. Organic Adsorbents for Wastewater Treatment

With the advancements in material chemistry, the focus has been shifted towards the development of cost-effective adsorbents from readily available and cheap natural resources such as fruit peel, rice husks, wheat straw, coconut and coffee waste, shells, mud, sugarcane bagasse, peat, agriculture, and domestic waste [59,60]. These wastes, along with fruit peel and other organic waste, are converted into cost-effective adsorbents through treatment with different chemical substances or drying and modifying them accordingly [61,62]. With advancements in analytical and technical approaches, wastewater treatment using organic waste is gaining much more attention (Table 2).

5.1. Forestry and Wood Waste Adsorbent

Forestry waste (such as tree twigs, branches, leaves, and bark) is accumulated in large quantities in the form of solid waste and can be used as a feedstock for manufacturing adsorbents for wastewater treatments. Polysaccharides (pectin, cellulose) and polyphenol complexes (flavonoids, tannins, lignin, terpenes) have specific functional groups in combination with hydroxyl (-OH) or carboxyl (-COOH) groups with passing ions. These wastes have a high metal ion adsorption potential through the ion-exchange or chelation process [63]. Various types of forest waste—i.e., bark, chestnut borer, sawdust, pine pectin, and pine needles—have been used as adsorbents to remove PTEs. Among these biological wastes, chestnut bur has the maximum absorption value—i.e., 16.18 mg g^{-1} —and its bark has the value of 9.31 mg g^{-1} [64]. Forestry wastes are also used to make biochar, an absorbent carbon material attained through slow pyrolysis. Biochar has the highest removal efficiency in removing PTEs—i.e., 264 mg g^{-1} —from wastewater [65]. It has been reported that the waste produced from the forest products has been used with an efficacy of more than 69%.

5.2. Agricultural Waste as an Efficient Wastewater Adsorbent

Agricultural wastes are very popular feedstocks for making adsorbents due to their availability and cost-effectiveness. Agriculture waste has been used for many purposes, as mentioned in Figure 1. They usually consist of lignin and cellulose as the main components and have -OH and -COOH groups. These groups can be combined with metal ions by providing electron pairs to form complexes. Agricultural wastes such as grape straw, tea and coffee grounds, nutshells, papaya and plant leaves, waste grains, algae, crab apple shells, rice bowls, and sunflower plants have been used by many scientists to remove PTEs such as As, Cd(II), Cr(IV), Hg, Pb, and Ni. Used tea or coffee powder is an example of farming waste that is produced in large quantities and needs little or no treatment. As with other biomass waste products, these wastes symbolize unused resources (Supplementary Information Table S2).

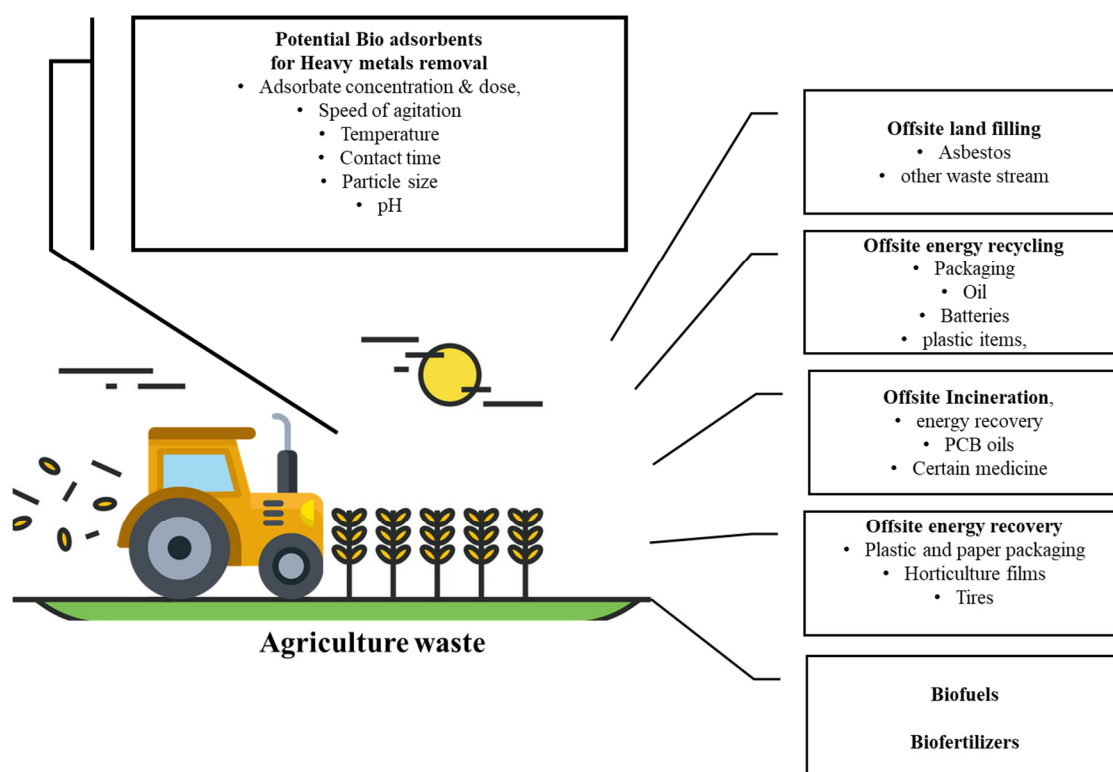


Figure 1. Agricultural waste product usage in different processes.

Agricultural waste-derived adsorbents can be modified by different chemical pre-treatments to increase the potential of functional groups, thereby increasing the adsorption capacity of adsorbents [66]. Facts have also shown that lignocellulose biomass obtained from agricultural waste-based products could be an efficient feedstock for the manufacture of carbonaceous materials such as biochar, which has a higher surface area, pore volume, and pore distribution [67].

Table 2. Treatment of industrial effluents using the biosorption process.

Type of Effluent	Metal Ions Focused (mg/L)	Other Ions/Impurities Present (mg/L)	Biosorbent	Operating Conditions	References
Battery effluent	Pb = 110		Bael leaves	Batch (pH = 2.2)	[68]
Battery manufacturing industrial effluent	Pb = 102 ± 3.6	Mn = 0.32 ± 0.02 Ni = 0.28 ± 0.01 Fe = 0.28 ± 0.01 Zn = 0.89 ± 0.05 Cr = 0.07 ± 0.01	<i>Saccharomyces cerevisiae</i>	Batch	[69]
Chrome electroplating effluent	Cr = 204	Cu = 0.29 Ni = 0.05 Fe = 0.26 Zn = 0.10	<i>Saccharomyces cerevisiae</i>	Batch	[70]
Electroplating effluent	Cr = 9.0 ± 2.0 Ni = 23.0 ± 2.0 Zn = 1.4 ± 0.2 Cu = 2.57 ± 0.04	-	<i>Saccharomyces cerevisiae</i> NCYC 1364	Multi-batch	[71]
Electroplating effluent	Ni = 5.25	Na = 115.39 Mg = 73.27 Ca = 6.52	Loofa sponge-immobilized <i>Chlorella sorokiniana</i>	Column (F = 5)	[72]
Electroplating industrial effluent	Ni = 109	Pb = 4.5 Cu = 1.3 Fe = 1.6 Cl = 289 SO ₄ = 558 Na = 101 K = 84	Crab shell	Column (pH = 7.5)	[73]
Electroplating industrial effluent	Ni = 52	Pb < 0.1 Cu < 0.1 Fe = 0.9 Cl = 230 SO ₄ = 300 Na = 76 K = 58	<i>Sargassum wightii</i>	Column (pH = 7.9)	[74]
Electroplating wastewater	Cr (VI) = 47	Cu = 0.9 Zn = 239 Fe = 8.6 Ni = 32 CN = 16.0 SO ₄ = 450 PO ₄ = 0.1	<i>Aspergillus niger</i>	Batch	[75]
Gold electroplating effluent	Au = 46	Ca = 376.7 K = 1621 Na = 1775 Cl = 3550	Immobilized <i>Cladosporium cladosporioides</i>	Batch	[76]
Gold ore mining effluent	Cu = 5.4 Zn = 0.23 Hg = 0.035 Pb = 0.34	CN = 73 Fe = 2.1 Al = 1.8	Chitosan	Batch (pH = 6)	[77]

Table 2. Cont.

Type of Effluent	Metal Ions Focused (mg/L)	Other Ions/Impurities Present (mg/L)	Biosorbent	Operating Conditions	References
ICP-OES effluent	Cr = 0.36 ± 0.11 Mn = 2.08 ± 0.07 Fe = 0.43 ± 0.17 Co = 0.26 ± 0.04 Ni = 1.47 ± 0.12 Cu = 4.81 ± 0.32 Zn = 2.31 ± 0.14 Al = 3.79 ± 0.21 Cd = 3.91 ± 0.21 Pb = 0.88 ± 0.11	Na = 118 ± 1.5 K = 17.2 ± 0.3 Ca = 7.84 ± 0.23 Mg = 4.55 ± 0.09	Hybrid Sargassum-sand	Column (pH = 4, F = 10)	[78]
Industrial effluent	Cd = 9.6 ± 0.5	Pb = 3.2 ± 0.5 Cu = 2.8 ± 0.5 Ni = 2.8 ± 0.5	Pleurotus platypus	Column (pH = 6.0, F = 5)	[79]
Laboratory effluent	Cd = 22.0; Pb = 12.0; Al = 24.1; Cu = 10.7; Zn = 21.4	-	Rice husk	Column (pH = 4.0, F = 8)	[80]
Laboratory effluent	Total metal concentration = 6 mM	-	Crab shell	Column (pH = 4.5, F = 5)	[81]
Metal plating industrial effluent	Cu = 720	Pb = 0.5 Cd = 0.005 Cr = 3.9 Zn = 2.5 Al = 26.9 Fe = 49.1 Ni = 25.0 Mn = 1.0	Gelidium sp.	Column	[82]
Metal plating industrial effluent	Cr = 108	Pb < 0.1 Cd < 0.005 Cu = 0.5 Zn < 0.15 Al < 1.0 Fe < 0.2 Ni < 0.2 Mn < 0.1	Gelidium sp.	Column	[82]
Metallurgical effluent	Zn = 88 Cd = 1.4 Mn = 11.7 Cu = 0.35	Ca = 444 Mg = 100 Na = 37	Sargassum sp.	Column (F = 25)	[83]
Storage battery industry wastewater	Pb = 3.1	Cu = 0.027 Ni = 0.005 Cr = 0.373	Rhizopus arrhizus	Batch (pH = 4.5, T = 30 A = 120)	[84]
Tannery effluent	Cr (VI) = 100 Cu = 3	-	Bacillus sp. FM	Batch (pH = 7.9, T = 37, A = 150)	[85]
Tanning industry effluent	Cr (III) = 1770	Na = 30,100	Mucor meihi	Batch (pH = 4.0)	[86]
Tanning industry wastewater	Cr (III) = 0.45	Fe = 0.14 Cl = 2340 SO ₄ = 2080 NH ₄ = 0.6 NO ₄ = 10	Cunninghamella elegans	Batch (pH = 8.2, T = 30, A = 110)	[87]

Fruit and Vegetable Peels

In most kitchen waste containers, fruit and vegetable waste and peel make up the highest proportion. Many fruit and vegetable peels are disposed of in debris or fed to livestock directly. Vegetable and fruit wastes and byproducts that are produced in significant quantities during industrial processing/secondary product manufacturing constitute a severe problem. They must be managed or recycled due to their harmful environmental impact. Fruit and vegetable peels and skins are a natural, environmentally friendly, and economical source of adsorbents that can eliminate different types of water contaminants and reduce pollution, and are therefore a renewable and promising resource [88].

Fruit shells—i.e., coconut shell—contain the dynamic functional groups of -OH and -COOH present in cellulose, hemicellulose, and pectin which are involved in PTE (Cd, Pb, As, Cr, Cu, and Ni) binding and removal [89]. Feng et al. [90] examined the efficiency of fruit shell-based adsorbents to eliminate Cu(II) from galvanic wastewater. In 50 mL of wastewater samples holding 14.33 mg L⁻¹ Cu(II) ions, the adsorption efficacy was recorded to be up to 97.1%. To evaluate the cost-effectiveness, the same process was repeated to check the adsorbent's reusability, and it was concluded that the adsorbent could be reused for the same process multiple times [91].

Santhi and Manonmani [92] reported that 6 g of adsorbent is enough to remove 90–95% Cr(VI) from the wastewater. The wastewater pollutant removal rate was increased with an increase in contact time to reach equilibrium after 120 min. The contaminant removal rate of citrus peel was recorded as 58.97% [93]. Citrus peel (*Citrus Nobilis*) was also used to remove PTEs from 10 wastewater samples taken from the battery industry in Londrina (Brazil). For all samples, the remediation rate of Cu(II), Cd(II), and Pb(II) by bio-adsorption was recorded to be up to 99.9% [94].

5.3. Peat

The use of peat in wastewater remediation has gained attention in the past few decades due to its high porosity and adsorptive capacity. Many researchers classify peat into four groups: moor peat, wood peat, herbaceous peat, and sediment peat [95]. It has the properties of being rich, cheap, and versatile and has a sturdy adsorption capability for various toxins such as PTEs and organic contaminants [96]. Unprocessed peat contains many integral constituents such as lignin; cellulose; fulvic and humic acids; and polar functional groups such as alcohols, aldehydes, ketones, carboxylic acids, and phenol hydroxides [97]. Peat also has a strong cation exchange capacity [98]. Its removal efficiency is higher for dyes when treated with acids. Peat collected from Panaga, Brunei Darussalam, showed a great affinity for the adsorption of congo red dye from wastewater.

5.4. Biochar

Agricultural waste-derived biochars have attracted greater attention among cheap and effective adsorbents for wastewater treatment [99]. They have a porous, stable structure and are an insoluble and carbon-rich solid material produced by pyrolysis (300–700 °C) under anaerobic conditions [100]. They can be produced from a broad range of agricultural and other biomass waste products such as crop straws, rice husks, yeast, sawdust, mud, kitchen waste, tea residue, and many others [101–103]. Biochar has been adopted as an efficient means of treating wastewater. Its ability to adsorb PTEs has been studied very critically in the past decade [104], showing that biochar is inexpensive, environmentally friendly, and more effective even than activated charcoal. The chemical properties of the biochar surface undergo complex and unpredictable changes during pyrolysis [105]. Although its low adsorptive efficiency of PTEs restricts its use into the field of sewage treatment, this is thought to be due to its low porosity, specific surface area, few adsorption sites, and functional groups [106].

In general, the remediation of PTEs by biochar containing aqueous solutions may be carried out by physical and chemical interaction procedures such as the complexation of the outer and inner sphere, electrostatic attractiveness, ion exchange, and surface

precipitation [107] (Figure 2). Following the results of adsorption kinetic trials and characterization experiments using Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM-eds), Fourier-transform infrared spectroscopy (FTIR), X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD), etc., the adsorbing process of biochar on PTEs commonly comprises physical adsorption, electrostatic attraction, ion-exchange, and complexation [108].

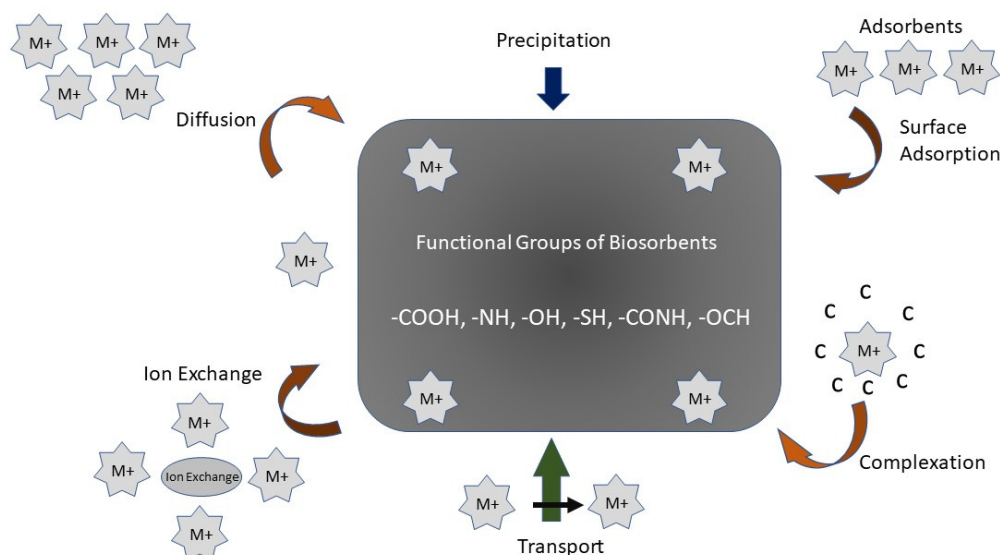


Figure 2. A schematic diagram of the mechanism of biochar/biosorbent to remediate contaminants.

The interior and surface structure of biochar comprise a blended allocation of macropores, mesopores, and micropores, whereby biochar maintains PTEs in its internal and surface pore structure. Fourier-transform infrared (FTIR) spectroscopy imaging has demonstrated that the functional groups of -COOH and -OH changed before and after adsorption because of complexation with Pb and Cd. The O-metal bond causes the electron density of O₂ to decrease, which drastically reduces the bound energy of the O₂-containing functional groups and improves its stability. The metal interaction can be interrupted by trembling in the C-C bonds [109]. Additionally, O₂ which includes functional groups emits H⁺ when ions are exchanged with PTEs, which leads to a reduction in the pH of the solution. Its surface charge properties are some of the standards used to determine the power of raw material in the electrostatic adsorbing process (Figure 2). The pH and redox potential plays a crucial role in the adsorption of PTEs in wastewater [110].

5.5. Coal Based Adsorbents

Coal is an organic material that contains different minerals. Moreover, organic materials normally make up 85–95% (wt/wt) of coal's dry biomass. Coal is a complicated sedimentary rock that mainly consists of the byproducts of plant residues and their derivatives. It is the source of carbon, although it also has different elements, such as hydrogen, oxygen, sulfur, and nitrogen. Coal and its derivatives are used not just as fuels, but as precious materials in various environmental protection processes as well. Coal is cheap and present abundantly, even some countries have numerous reserves of coal as mines. However, it has exciting properties that make it an efficient adsorptive material for removing various organic contaminants and PTEs [111].

Coal can form stable complexes with multiple PTEs because of the -COOH groups and phenol groups connected to its highly cross-linked aromatic structures. Carboxyl or hydroxyl groups can participate in ion exchange reactions [111]. Arpa et al. [112] reported that using inferior Turkish coal has the ability to efficiently remove Hg(II), Cd(II), and Pb(II) ions from mining wastewater. Karabulut et al. [113] reported that inferior Turkish coal

can also remove Cu and Zn from sewage sludge. The adsorption phenomenon appears to keep up with the Langmuir isotherm model. An analysis of crude coal and exchanged coal using FTIR showed that a significant amount of PTEs were removed and seen on the coal surface due to the development of exchange metal carboxylates. Multiple studies have also been carried out to unveil the removal of Cr at different oxidation levels from waste solutions by utilizing low-grade coal [114,115].

6. Efficacy of Industrial Waste-Derived Adsorbents for Wastewater Treatment

Industrial wastes are believed to be cost-effective adsorbents for removing PTEs from wastewater. Usually, they require no treatment to increase their adsorption capacity [116], but it is possible to improve their adsorption capacity through physical and chemical modifications or by treating them with other elements. In the past, byproducts such as fly ash, blast furnace sludge, pulp waste, lignin, red mud, and sludge have been used as effective adsorbents because they are technically suitable for removing PTEs from wastewater [117] (Table 3).

Table 3. Reported literature summary of the adsorption of metals in industrial waste.

Metals	Adsorbent	Adsorption Capacity (mg/g)	References
Zn(II)	Powered waste sludge	168	[118]
	Solid residue of olive mill products	5.40	[119]
Ni(II)	Red mud	160	[120]
	Olive stone waste	2.13	[121]
Cu(II)	Blast furnace slag	133.3	[122]
	Areca waste (AW)	1.12	[123]
Cr(VI)	Waste slurry	640	[124]
	Iron(III) hydroxide	0.47	[125]
Cr(III)	Lignin	17.97	[126]
Hg(II)	Waste slurry	560	[124]
Cd(II)	Waste slurry	15.73	[127]
	Orgabosolv lignin	1.10	[122]
Pb(II)	Lignin	1865	[128]
	Tea industry waste	2	[129]
Fe(III)	Tea industry waste	24	[129]
As(III)	Zr (IV)-loaded orange waste	130	[130]
As(V)	Zr (IV)-loaded orange waste	88.0	[130]
	Leather industry waste	26	[131]
V(V)	Waste metal sludge	24.8	[132]

6.1. Fly Ash

Industrial fly ash has significant potential to be used as an alternate to activated carbon or zeolite as an adsorbent. The chemical structure of fly ash contains a high proportion of silica (60–65%), aluminum oxide (25–30%), and magnetite (6–15%) and has a peculiar bulk density, particle size, porosity, water holding capacity, and surface area. All these properties make it an appropriate tool to use as an adsorbent material [133]. Raw fly ash modification is also performed through physical and chemical treatments to increase the adsorption capacity, thereby increasing the application rate [134].

Fly ash has been used to remove Cr and Cu from industrial wastewater. Its removal efficiency depends on the intensity, pH, and temperature. The adsorption kinetics indicate that the process is controlled by diffusion. It contains different amounts of carbon and minerals [135]. For Cr removal, fly ash and bagasse are used individually as well as in a combined form [136,137].

Banerjee et al. [138] explored the adsorption of Ni, Zn, Cr, and Hg through the application of fly ash. Compared to untreated fly ash, saturated fly ash had a greater adsorption capacity. Wastewater treatment through saturated fly ash showed lower toxicity levels of Cu, Pb, phosphate, and nitrates up to 95% as compared to untreated fly ash.

6.2. Blast Furnace Slag, Sludge and Dust

Sludge is a dry waste of the electroplating industry. It is also produced by the reaction of metal ions with calcium hydroxide and precipitation in wastewater (Figure 3). Iron and steel works produce huge quantities of granulated blast furnace slag, which is used as a filler or for the manufacturing of slag cement [116]. Adsorbents obtained from sanitation sludge by chemical pyrolysis have been used to eradicate Cd and Ni from wastewater [139]. The adsorbing capacities of Cd and Ni were computed by the Langmuir isotherm as 16 and 9 mg g⁻¹, respectively. The adsorption mechanism appeared to be the ion-exchange method. Since sewage slurry is disposed of as garbage from wastewater treatment plants, another study used distillery sludge to remove Cr from municipal and industrial wastewater [140]. The adsorption capacity was 5.7 mg g⁻¹, while a desorption study showed an up to 82% Cr removal capacity.

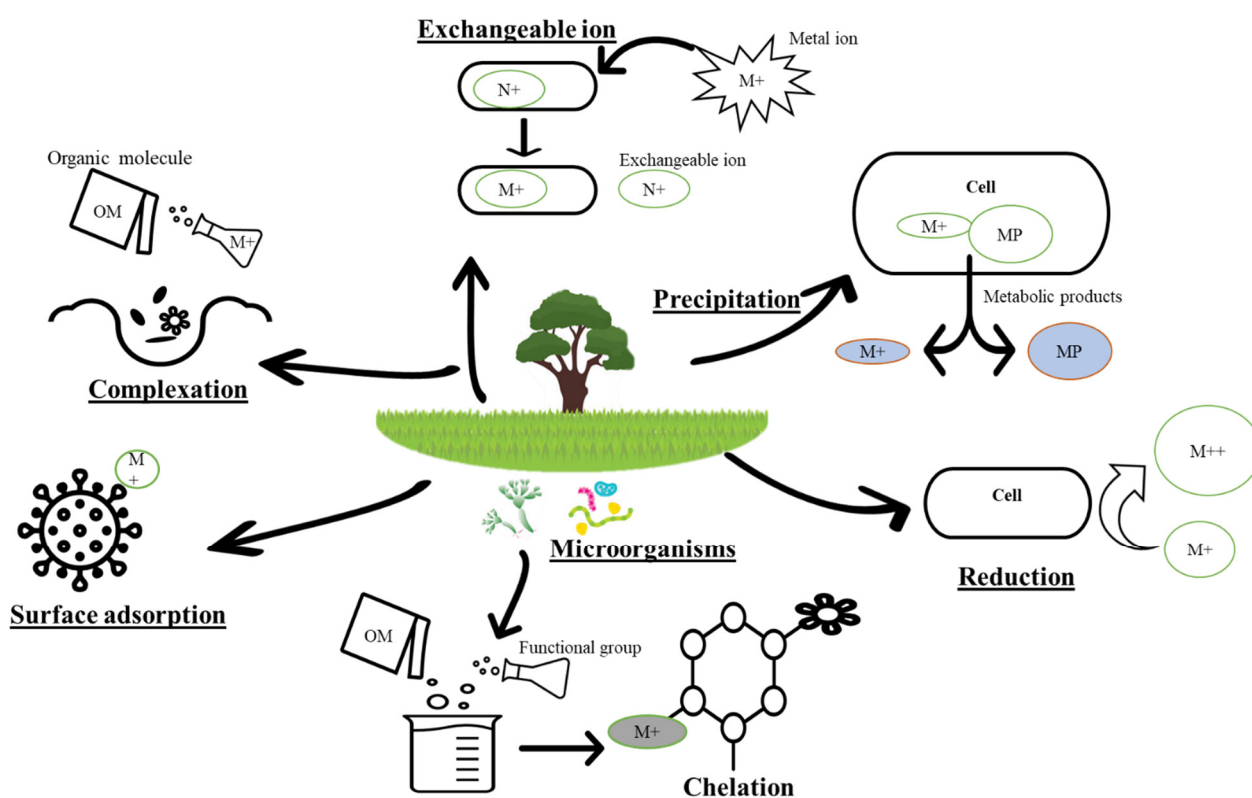


Figure 3. Possible heavy metal adsorption mechanisms.

Martín and Ruby [141] reported that blast furnace sludge, a byproduct of the steel industry, exhibited a good capacity for Pb, Zn, and Cd removal. It has a specific surface area of 27.4 m² g⁻¹ and a great affinity for Pb with an adsorption capacity of about 64.2–79.9 mg g⁻¹.

7. Marine Material-Based Adsorbents

Among these adsorbent substances, algae are a natural and economical cation exchanger with a huge surface area [142]. Brown algae cell walls usually contain alginic acid, cellulose, and sulfated polysaccharides. Therefore, carboxyl groups and sulfates are the main effective groups in these types of algal blooms. Red algae similarly comprise of cellulose, have their own role in biosorption process because of the presence of sulfated polysaccharides from galactan. The green algae cell wall contains heteropolysaccharides that provide -COOH groups and sulfate groups for the adsorption of PTEs (Figure 3). Chlorella consists of a high proportion of cell wall protein and cellulose, together with polysaccharides in the form of glycoproteins. Such substances include several functional

groups (carboxyl, amino, sulfate, and hydroxyl) and can play a key part in the adsorbing process [143,144].

8. Nanomaterials: Potential Use in Wastewater Treatment

Nanotechnology exists within the field of nanoscience. Nanomaterials are the world's tiniest structures synthesized by humans, with a magnitude of a couple of nanometers [145]. More specifically, nanoparticles (NPs) are fragments which have a structural component in a dimension of not more than 100 nm [146]. NPs are being developed in numerous forms, such as nanowires, colloids, films, quantum dots, particles, and nanotubes [147]. For wastewater treatment, highly effective, environmentally friendly, and inexpensive NPs with unique functions have been developed to purify industrial wastewater, river water, groundwater, and drinking water [148]. Due to their unique properties, they can be divided into three types: nanoadsorbents, nanocatalysts, and nanofilms [149,150]. Nano-adsorbents can be produced by utilizing atoms of such components which are chemically effective and possess a high adsorbing capability on their surfaces [151–153].

8.1. Nano-Adsorbents

The use of nano-adsorbents for wastewater treatment is a positive approach for the removal of different contaminants. The potential of nano-adsorbents has been investigated in recent years. Smaller particle sizes increase their chemical activity and adsorption capacity [154]. Because of their role in the adsorption process, nano-adsorbents are roughly divided into different groups. These include metal nanoparticles (NPs), nanostructured mixed oxides, magnetic NPs, and metal oxide NPs. In addition, the latest developments include carbon nanomaterials, carbon NPs, and carbon nanosheets. In addition, various types of silicon NPs are being used as nano-adsorptive silicon nanotubes, silicon NPs, and silicon nanosheets. In one study, nano-tones, polymer-based nanomaterials, nanofibers, and aerogels were some type of NPs that has been utilized to remove PTEs from wastewater [155].

Chemical composition, structure, solubility, shape, fractal dimension, size, and surface chemistry are factors that affect the performance of NPs used as adsorbents [156]. Chemical activities as well as particle size are two important features of NPs. In comparison to other ingredients (such as titanium dioxide and aluminum oxide on a normal scale), NPs have outstanding advantages [154]. In addition, NPs may be modified with a specific reagent to improve their pre-concentration performance for metal ions [157]. The adsorption process will depend upon the adsorption coefficient and the regeneration and distribution of pollutants in accordance with the equilibrium conditions [158]. In addition, a redox reaction with persistent inorganic pollutants facilitates the start of the transformation of the ion structure [159]. Still, some scientists agree that variations in the redox conditions affect the toxic effects of those toxins [160]. The most used NPs as adsorbents for PTEs are graphene, iron oxide, magnesium oxide, activated carbon, manganese oxide, zinc oxide, titanium oxide, and CNTs [161].

8.2. Nanocomposites in Wastewater Treatment

Nanocomposites (NCs) are usually a mixture of components (two or more) with different properties that are usually processed into a single substance with a comprehensive set of properties [162]. The key advantage of using composite materials is the ability to combine the characteristics of two materials for certain applications. Under the current circumstances, NPs have achieved popularity in different areas, such as the construction, aviation, vehicle, and biomedicine industries [163,164]. Researchers are focusing on the use of these materials in wastewater treatment [165].

It is known that NPs have a high surface area to volume ratio and can significantly improve the matrix properties (metal/polymer/ceramic) of NPs built into them to form composite materials. In recent years, nano particles (NPs) have been used to remove micropollutants due to their large surface area, adhesion properties, cost effectiveness,

antifouling properties, thermal stability, and excellent mechanical characteristics [166]. A cerium oxide NC structure was developed and has the possibility to remove carbon monoxide and contaminants from wastewater [167]. Another important strategy for the use of NCs is to magnetize CNTs with iron (zero valence) and then optimize the adsorption behavior in order to eliminate nitrates, chlorinated organic toxins, and metals from water [164,168].

8.3. Carbon Nanotubes (CNTs)

Carbon nanotubes (CNTs) are widely researched materials that can remediate PTEs and different organic contaminants from wastewater by an adsorption mechanism [169]. Yet, their inadequate disposability, problems in their separation, and tiny particle sizes are the problems with using CNT as adsorbents. In order to address these issues, researchers altered ordinary CNTs to modified CNTs such as multi-walled CNTs [170]. The modified magnetic CNTs have an elevated disposability and could easily be eliminated from wastewater or applied media with magnets [171]. Various studies have described the use of multi-walled CNTs to remove Pb, Mn [170], and Cu [172]. Gupta et al. [173] examined the adsorption capacities of treated and un-treated CNTs for Al removal. It was revealed that the coated CNTs showed a greater removal capacity than uncoated CNTs.

The surface alteration of CNTs can improve their whole adsorptive activity. Numerous researchers have reported various surface modification techniques, including acid treatment [174], metal impregnation [175], and functional molecule/group transplantation [176]. The modification of the properties of CNTs is another method to improve their efficiency for pollutant removal. This can be carried out in several ways—e.g., through plasma technology, chemical alteration, and microwaves [175]. Among these technologies, plasma technology is considered one of the most suitable because of its lower energy consumption and environmentally friendly process. Chen et al. [176] described the usefulness of modified CNTs spliced with different functional groups for the remediation of PTE-contaminated water. In addition, CNTs altered with metals/metal oxides such as MnO_2 and Al_2O_3 have shown promising results for their adsorption mechanism [173,177,178].

8.4. Graphene Based Nano-Adsorbents

Besides the utilization of NPs, NCs, and CNTs in wastewater treatment, graphene also has special properties to be used individually and in combination with other NPs. Graphene oxide (GO) is a carbon nanomaterial with a two-dimensional structure which is manufactured through the chemical oxidation of a graphite coating [179]. Due to its large surface area, high mechanical strength, low weight, flexibility, and chemical stability, GO has attracted increasing attention as an adsorbent for removing PTEs [180]. Ding et al. [181] effectively used GO in column reactors to remove PTEs from wastewater. Lee and Yang [182] modified GO with TiO_2 and applied hybrid composites to adsorb Zn, Cd, and Pb from wastewater. The adsorption capacity of the hybrid complex for Pb, Cd, and Zn reached 65.6 mg g^{-1} , 72.8 mg/g , and 88.9 mg g^{-1} , respectively. Graphene as well as other composite materials displays an extremely high removal of PTEs from wastewater [27].

8.5. Magnetic Nanocomposites

Magnetic NCs are a unique category of nanomaterials. They have core-shell nanostructures that can be quickly and easily restored by exterior magnetic fields. Functional group NPs may be grafted as well as fixed on Fe_3O_4 NPs through chemical bonding or direct deposition [183]. Raw materials which can be coated are Ag, TiO_2 , CNT, GO, Pd, and SiO_2 . Silicon oxide coating can offer a large surface area as well as echo porosity while at the same time keeping the magnetic core from erosion (Supplementary Information Table S3). After coating, the resulting magnetic NCs generally show improved adsorption capacities and have fast kinetics for the removal of pollutants such as PTEs, pigments, phenolic substances, and microorganisms [184]. In the last few years, Cui et al. [185]

synthesized a sequence of porous magnetic nanowires based on manganese that have been synthesized to remediate PTEs and various organic contaminants. A straightforward one-step solvothermal process was also proposed to produce hollow magnetic carbon spheres that are very effective in removing PTEs.

Jin et al. [110] concluded that amino acid-modified magnetic NPs can adsorb up to 94% of the bacteria cells in a pH spectrum of 4–10 within 20 min. Zhang et al. [186] developed other multifunctional magnetic NPs coated with a polyethylene core which can inactivate bacteria by penetrating the cell membrane and remove PTEs by chelation. In addition, these NCs can be regenerated by Ethylenediaminetetraacetic acid (EDTA) and NaOH and reused.

8.6. Combination of Biological-Nano Technology Processes

There are various technologies to harvest algal biomass, such as deposition, centrifugation, and air flotation, in which condensed chemicals are used as a carrier. However, these technologies cannot be used on a large scale due to their high costs [187]. In these advanced technologies, membrane technology is the utmost advantageous method for algae growth and biomass production, in which the cultivation of algae with a high density is only completed by membrane bioreactors [188]. The advantage of membrane technology is that no coagulants need to be added for membrane filtration, which promotes the reuse of filtered water and simplifies the separation of algal biomass [189]. Scientific and technical technologies on a nanoscale show that many existing problems with water quality can be solved using nanostructured catalytic membranes, nano-catalysts, nano-absorbents, nanotubes, nano-powder, and micro-molecules [190]. These are all NPs and colloids that have a significant impression on the quality of water in the treatment procedure [191]. A study has demonstrated that combining wastewater treatment processes with advanced nanotechnology can produce highly effective water treatment systems [192]. The cultivation of algae in wastewater is one of the most useful approaches for energy generation and wastewater treatment. Many types of algae show effectiveness due to the presence of PTEs [156]. Nutrients are mixed with water to form a solution which provide the essential growth conditions required for algae. In addition, algae biomass is recovered more efficiently than conventional methods without damaging the cells, and the energy requirement for the algae harvest is less than other methods [193]. Polyvinylidene fluoride, poly sulfone, and polyether sulfone membranes are widely useful because of their physiochemical stability, although the main problem is membrane material and the microbial cells between the hydrophobic mechanism and membrane contamination [194]. Research has indicated that NPs can improve hydrophilicity and decrease membrane contamination—for example, CNTs and TiO₂ [192].

The performance of microbial fuel cells can be improved by using inexpensive NCs such as nanoscale carbon in electrodes, as electrodes are mechanically stable and have a large surface area, great electrical conductivity, and good electrochemical catalyst activity [195]. Because of all the unique properties of platinum (Pt), commercial Pt cathode catalysts can therefore be replaced by CNT/Pt in microbial fuel cells [196]. To increase the adhesion of microorganisms and decrease toxicity, CNTs were also coated by numerous anemic polymers such as polyaniline and poly-pyrrole to constitute NCs. These NCs comprise in the negative charged CNTs, which are combined by electrostatic interaction with positively charged polycationic polymers as microbial fuel cells anodes [197].

9. Regeneration and Disposal of Used Adsorbents

The recycling prospective of waste biomass is a crucial criterion for the choice of adsorbent. The potential of regenerating biomass reduces the whole process costs and the dependency of the procedure on the constant supply of biomass. The success of the desorption process will depend on the type of removal mechanism used and the mechanical strength of the biomass. Since most biosorbents have an ion-exchange system for PTEs, mild to strongly acidic conditions are adequate for the desorption. The use of acids in desorption is also helpful because acid solutions are the most common waste in all industries.

Adsorbents that are used in the treatment of wastewater can also be regenerated, disposed of, or recycled. However, it should be kept in mind that the efficiency of the adsorbent should not be adversely affected by the acidic conditions. One of the most important industrial uses of desorption is to recover the contaminants (particularly valuable metals) and at the same time to regenerate the adsorbent for reuse. Actually, the efficacy of a particular biomass being as adsorbent will depend not only on its adsorption capability, although also on its ease of regeneration and recycling [198]. However, most studies have focused only on the adsorption capacity of the tested biosorbent without considering its possible reuse in industry [199]. The regeneration and discarding of waste adsorbents have been debated in detail in the past few years to certify their recycling and reduce the potential dangers of waste adsorbents [200]. In general, improper treatment of waste adsorbents loaded with As and other PTEs can cause serious environmental problems. It is therefore necessary to securely get rid of waste adsorbents like the landfills, mix them with animal manure and incorporate them in building materials [201]. In this respect, stabilization/solidification is regarded as a suitable technology for treating waste adsorbents. Therefore, this option has been widely used by almost all alternatives to dispose of toxic waste since it can transform toxic waste into less toxic through chemical, physical, or thermal procedures [202].

In general, an efficient rejuvenation process will be based upon the following principle: the desorbing adsorbent can completely restore its preliminary adsorbing performance [203]. In this context, the adsorption effectiveness of numerous adsorbents is assessed based on the reproducibility. Studies have shown that the regeneration efficacy of a composite remains unchanged even after six desorption stages [204]. Almost 90% of the adsorbents did not provide regeneration test data, especially for conventional materials. It can therefore be speculated that most conventional adsorbents have a low desorption efficiency for PTEs. Therefore, further investigation is necessary to find more sophisticated materials with a high adsorbed performance which can be directly regenerated in multiple adsorption-desorption sequences without a substantial loss of adsorption capacity.

An adsorbate can only be removed by combustion or dissolution in strong acids or bases. If low-cost biomass is used as a biosorbent for the recovery of precious metals, destructive recovery is economically viable. So far, however, the focus has been on non-destructive desorption from the stacked biosorbent. In many instances, dilute in-organic acids or bases can effectively desorb the adsorbent, although they may cause structural harm to the adsorbent himself, which leads to a decrease in the adsorbent capacity after regeneration. Similarly, as already mentioned, the pH of the solution has a significant effect on the adsorption of target pollutants. In theory, therefore, simply controlling the pH of the desorption solution should be a great way to regenerate adsorbents and to recover contaminants [205]. The benefits of adsorbent regeneration include the reusability of used adsorbents, the reduction in the costs for the disposal of metal resources, secondary waste, and the determination of the adsorption mechanisms. Various solutions, including HCl, H₂SO₄, NaOH, NaNO₃, EDTA, DRTA [206], HNO₃, NaCl, CH₃COONa, NaHCO₃ [207], have been used to regenerate the adsorbent.

Another possible method is solvent extraction (organic extraction). Particular attention should be paid to the choice of solvent and its cost effectiveness. In addition, a mixture of organic solvents and aqueous solutions can have beneficial effects in certain cases [208]. Using Fenton reagents, advanced oxidation processes for thermal re-activation, or photocatalytic processes for material regeneration and salt treatment are also possible. Nevertheless, they are not appropriate for the redevelopment of NCs for organic carriers, since oxidation can cause the decomposition of the host [208]. The expansion of new recycled approaches for NCs will make them more appealing and environmentally sustainable for industrial applications. Cobalt-laden *Ascomphyllum nodosum*, which is exposed to an acidic solution, can offer a good desorption efficiency, but leads to a significant reduction in biomass weight, which affects the next cycle of biomass performance. Therefore, these researchers tested other de-sorbents and found that 0.05 M CaCl₂ (in HCl) is an effective de-sorbent. Other methods, such as complexation, chelation, and micro-precipitation, require a broad

screening of effective de-sorbents. Facts have shown that few chemical reagents for different biomasses, including Ethylenediaminetetraacetic acid (EDTA), are effective and harmless. Under strongly acidic or strongly alkaline conditions, biomass can be stable without immobilizing the macrostructure, including crab shells, some large-scale algae industrial wastes, and agricultural wastes. Therefore, choosing a suitable biosorbent to remove metallic ions from contaminated water is a difficult task. This selection should be based on multiple criteria that have a direct impact about the performance of biosorbents in intricate matrices that commonly occur in wastewater [209].

10. Efficiency and Cost Comparison

The two most vital factors that make the adsorption process economical are the efficiency (adsorptive capacity) and the used adsorbent cost. However, as a comparison to the standard adsorbents, the adsorption capacity is relative as it differs in several factors, comprising the initial adsorbate concentration that is being tested. The cost of an adsorbent depends on several factors, including its accessibility, sources, required processing, processing conditions, recycling situations, and half-life. As fly ash is available free of cost in power plants, its use incurs only transport, laying, and rolling costs [133].

The price of fly ash from waste slag is $\$0.002 \text{ kg}^{-1}$; with the expense of transportation, chemicals, electrical energy, etc., utilized through the process, the expense of the final product is about $\$0.009 \text{ kg}^{-1}$ [210]. After fly ash, the cost of blast furnace scrap, scrap metal, carbonaceous adsorbents (fertilizer and scrap metal industrial waste) is $\leq 0.1 \text{ \$ USD per kg}$, which makes it a useful material in accordance with the conditions of cost. Activated carbon has prices that typically exceed $\$3.0$ primarily due to a mixture of two factors, adsorption capability and the cost of adsorbent.

Since many costly elements are involved, such as passive pumping and treatment plants, it is difficult to assess the cost of a credible treatment for metal-polluted wastewater. Due to changes in market demand, the quality and quantity of different wastewaters are also changing. This has led to changes in treatment costs. Therefore, complete information about the expense of wastewater treatment associated with adsorption is seldom provided in the scientific literature. Consequently, the overall cost–benefit assessment of the adsorbing process for PTE removal must be examined in more detail and suggested for future use. Labor costs vary country by country. In order to gain an accurate estimate of the operating costs of a wastewater treatment facility, pilot-scale studies must be carried out. Most of the data in this overview come from laboratory-scale studies, therefore pilot-scale experiments are required to measure the total treatment costs related to the proposed treatment. Because of their different operating conditions, it can be difficult to directly compare the treatment costs. In addition, the total treatment costs of various wastewater types depend on the procedure used in local circumstances. This could be explained by the fact that the majority of industries are situated in the commercial or industrial areas of cities. After neutralization by acid/alkali, wastewater is discharged into the sewer. The flow rate and characteristics of wastewater vary widely, and it is hard to accurately estimate treatment costs. This inconsistency in data representation makes it difficult to compare treatment technologies in terms of the cost–benefit ratio for wastewater containing PTEs and other pollutants.

11. Evaluation of the Present Scenario and Future Recommendations

Under the current circumstances, advanced water treatment technologies are urgently needed to guarantee high-quality water, reduce chemical and biological contaminants, and strengthen industrial production processes. Until now, most of the published findings involve experiments on a laboratory scale. The absence of information about pilot-scale systems is the main disadvantage of using cost-effective adsorbents as a replacement for activated carbon as well as other expensive treatment technologies. In order to promote the use of unconventional adsorbents on a large scale, further studies are required. However, nanotechnology involves the adsorption of contaminants using nanomaterials that

represents one of the best approach for advanced sewage treatment procedures. Numerous nanomaterials have been established and sewage treatment research has been successfully carried out. Among these, nano-adsorbents (based upon the oxides Fe, MnO, ZnO, MgO, CNT), photocatalysts, electrocatalysts (Pt, Pd), and nanofilms are worth discussing. In addition, these NPs can be incorporated into biological processes (algae membranes, anaerobic fermentation, microbial fuel cells) to improve the efficacy of the wastewater treatment process.

Every technology has its benefits and efficacy in removing pollutants. Nano-adsorbent can eliminate PTEs such as Cr, As, Hg, Zn, Cu, Ni, and Pb from wastewater. NP photocatalysts could be used to treat toxic contaminants, and the alteration of the catalyst material offers the possibility of using apparent areas to capture sunlight rather than expensive synthetic ultraviolet radiation.

Given that nanomaterials are not yet inexpensive compared to traditional materials (such as activated carbon), prospective research should concentrate on effective processes that only require small concentrations of nanomaterials. In addition, more work is needed to develop cost-effective methods for the synthesis of nanomaterials and test efficiencies on a large scale for successful field applications. The following research gaps need to be filled in the future:

- Cellular- and molecular-level studies are needed for a better understanding of adsorbent mechanisms in wastewater treatment.
- More work needs to be carried out on pilot-scale and field-scale experiments other than laboratory experimentation.
- Cost-effect benefits are need to be calculated before pilot-scale experiments are carried out.
- The efficiency of the discussed adsorbents, especially carbon-based adsorbents, needs to be explored in combination with other treatment approaches.
- Microbial interaction under aquatic environments should be explored.
- More work needs to be carried out on the impact of geo-environmental factors related to adsorption mechanisms.

Supplementary Materials: The following are available online at <https://www.mdpi.com/2073-4441/13/2/215/s1>: Table S1: Maximum permissible contaminant level (MCL) standards for the most hazardous heavy metals; Table S2: the adsorption capacities of some agricultural and biological wastes for heavy metals; Table S3: the efficiency of different nano-adsorbents for the removal of heavy metals from wastewater.

Author Contributions: F.Y., A.M., X.W. and Z.U.R.F. conceptualize and drafted this manuscript. S.Y., M.M.A., M.A.H. and W.M.-U.-D. helped the authors in making figures and collecting data for the tables. S.N., A.A.M. and M.M.H. worked on manuscript improvement and corrections. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not Applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: Author wants to acknowledge the Institute of Soil and Environmental Science, University of Agriculture Faisalabad, Faisalabad, Pakistan. Additionally, authors want to thank the Higher Education Commission Pakistan for providing fellowship under the International Research Support Initiative Program (IRSIP).

Conflicts of Interest: The authors declare no conflict of interest.

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