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Valorization of organic waste as biosorbents for wastewater treatment

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Abstract

With increasing population and waste generation, organic waste disposal has presented an unprecedented challenge. Valorization of the waste for wastewater treatment emerges as a feasible way to recycle or upcycle the waste. Agricultural and non-agricultural waste has been successfully converted into biosorbents to remove various pollutants. Through reviewing 126 papers, this review aims to provide a comprehensive overview of the effectiveness and mechanisms of biosorbents from organic waste in adsorbing different organic and inorganic waste. Most recent studies have focused on using biosorbents to remove dyes, pharmaceuticals, and heavy metals. The biosorbents were synthesized primarily through drying and pulverization, or pyrolysis. Some biosorbents have been chemically treated with acids, alkalis, or salts to increase their surface functional groups. Furthermore, different biomass materials have also been combined to synthesize biocomposite sorbents. The extraction of lignocellulose and chitin from biomass as biosorbents is also common. Biosorption occurs via chemisorption and physisorption, with the former more prevalent among organic pollutants and the latter among inorganic pollutants. The Langmuir isotherm model, which indicates monolayer sorption, and the pseudo-second-order kinetic model, which implies chemisorption as rate-limiting, best describe most of the biosorption data. Biosorption is governed mainly by pH, temperature, initial pollutant concentrations, dosage and size of biosorbents, and contact time. This review highlights the need to standardize optimization procedures and develop cost-effective and scalable biosorption systems. It highlights the potential of biosorbents, especially biochar, as potential substitutes for activated carbon in the column adsorption process of tertiary wastewater treatment.

Keywords: Biosorption, dyes, heavy metals, pyrolysis, Langmuir isotherm, valorization

INTRODUCTION

Organic waste is a term used to describe biodegradable materials that originate from plant or animal sources. This category includes a wide variety of items such as uneaten food, coffee grounds, fruit and vegetable peels, eggshells, yard waste, fallen leaves, branches, weeds, cut flowers, pet food, livestock manure, untreated wood, and paper or packaging stained with food [Figure 1][1]. Annually, the world produces 2.01 billion tonnes of municipal solid waste, with a conservative estimate suggesting that at least 33% of this is not managed in an environmentally friendly way. Despite making up only 16% of the global population, high-income countries are responsible for about 34%, or 683 million tonnes, of the world's waste[2]. By 2050, global waste is projected to increase to 3.40 billion tonnes, which is more than twice the expected population growth during the same period. The amount of waste produced in low-income countries is predicted to rise more than threefold by 2050. Middle- and low-income countries contribute 53% and 57% of food and green waste, respectively, with the proportion of organic waste increasing as the level of economic development decreases[2].

Each year, an estimated 11.2 billion tonnes of solid waste is collected globally. The decomposition of the organic portion of solid waste contributes to about 5 percent of global greenhouse gas emissions[3]. It produces a substantial amount of methane, a powerful greenhouse gas that plays a significant role in climate change. Organic waste can also contain substances such as antibiotics, chemicals, and pathogens that can infiltrate the soil and groundwater as the waste decomposes[4]. The appropriate handling of organic waste is vital for the sustainability of our environment. Valorization of organic waste into products with commercial values has been probed as the global shift toward the circular economy gains momentum[5,6]. Organic waste can be valorized through conversion into raw materials for energy sources or incorporation into finished goods. The latter is usually achieved by recycling and upcycling, which involve salvaging and reprocessing the waste into new, useful items[4]. Composting and aerobic digestion could convert organic waste into organic fertilizers and soil additives that add nutrients to the soil, as well as biogas, serving as a renewable energy source[7]. Organic waste contains a variety of essential phytochemicals, antioxidants, enzymes, and other bioactive compounds that can be extracted. Crop residues and by-products from food can provide the materials for manufacturing biofuels, bioplastics, and other biomaterials[8].

Organic waste can also be valorized as biosorbents. Biosorbents are materials of biological origin that can be used for sorption, particularly in removing contaminants from wastewater. Biosorption is generally a passive process that does not require energy[9]. The capacity of a biosorbent to eliminate contaminants relies on its kinetic equilibrium and surface characteristics. Since organic waste is made of cellular structures, contaminants are adsorbed to these structures when organic waste is employed as biosorbents[10]. Biosorbents play a crucial role in environmental cleanup efforts and are seen as a cost-effective solution for extracting toxic heavy metals from industrial wastewater, contributing to environmental remediation. They are capable of effectively eliminating toxic heavy metal ions, such as mercury, arsenic, lead, cadmium, and chromium, from polluted water[9]. Biosorbents can also remove other pollutants, such as dyes, pesticides, phenolic compounds, and fossil fuels. In wastewater treatment, these sorbents can be packed in an adsorption column where effluent with contaminants is fed into the column from the top[11]. The biosorbents adsorb the contaminants, and the clean effluent exits the column at the bottom. This process can be reversed to flush out the contaminants adsorbed. The biosorbents can then be reused or discarded and replaced[11].

Despite the rising interest in valorizing organic waste as biosorbents, the existing review is fragmented. Karim *et al.* reviewed the potential use of fibrous food waste for the biosorption of heavy metals. However, fibrous food waste is only a type of organic waste[12]. Though heavy metals have been the primary focus of

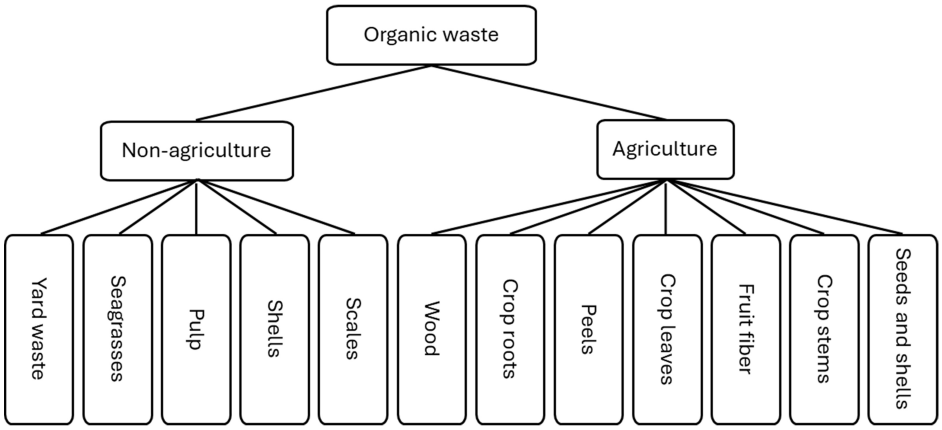

Figure 1. Examples of agricultural and non-agricultural organic waste. Agricultural organic waste originates from agricultural activities such as crop and fruit farming, while non-agricultural organic waste comes from food waste, yard waste, and various organic materials from natural habitats.

studies related to biosorbents, other types of pollutants in wastewater deserve attention. Okoro *et al.* were interested in reviewing the use of nanomaterial-based biosorbents to eliminate certain organic pollutants in wastewater. These nano-biosorbents comprise algae, bacteria, chitosan/chitin, and fungi[13]. Except for chitosan, which can be extracted from seafood waste, particularly the shells of crustaceans, other biosorbents are not considered organic waste. Additionally, seafood waste is one of the many different types of organic waste generated. The use of agricultural waste to produce biosorbents has garnered much review attention[14-16]. Agricultural waste, typically including waste from farms, poultry houses, and slaughterhouses, is a major constituent of organic waste but not its entirety. Organic waste also encompasses non-agricultural waste, such as food waste, yard waste, wood waste, and paper waste. Moreover, there is more interest in reviewing the ability of agricultural waste-derived biosorbents to remove heavy metals than other pollutants[15-17]. Park *et al.* gave an overview of the history, raw materials, and mechanisms of biosorption, as well as the synthesis of biosorbents. The overview did not examine the different types of organic waste that can be used for biosorbent synthesis and their specific adsorption performances and mechanisms in aqueous environments. Additionally, the overview was published over 10 years ago[18].

This review, therefore, aims to provide more comprehensive coverage of a wider array of organic waste for the biosorption of different pollutants from wastewater. Specifically, it aims to examine the efficiency and mechanisms of biosorbents derived from different types of organic waste in removing various pollutants from wastewater. It contributes to a better understanding of the feasibility of various biosorbents in treating wastewater while highlighting the latest innovations in their applications for wastewater treatment and future research directions.

METHODS

This review employed a narrative approach to comprehensively present the biosorbents derived from a wide array of organic waste for wastewater treatment. The literature search was conducted using databases primarily consisting of Web of Science, Scopus, and ScienceDirect. The keywords used in the search include adsorbent, biosorbent, organic, waste, water, wastewater, treatment, and household. To refine the search, the keywords were combined into key phrases such as organic adsorbent for water treatment, biosorbent for wastewater treatment, adsorbent from household waste, and adsorbent from organic waste. A total of 249 articles were retrieved from the databases and were screened using these criteria: (1) the articles must have been published in the past 10 years, with those in the past 5 years prioritized; (2) they must be related to

adsorbents from organic waste; (3) they must include the use of the biosorbents to treat various wastewater contaminants, ideally with the efficiency and mechanism specified. The screening resulted in 126 articles to be included in this review.

AGRICULTURE-DERIVED ORGANIC WASTE

Agricultural waste crops that have undergone physical and chemical treatments can be utilized to eliminate contaminants from wastewater. The adsorption capacity of the biosorbents is influenced by their chemical composition and physical properties, such as density, porosity, particle size, specific surface area, and the size and distribution of pores[19]. These factors determine the ability of the biosorbents to adsorb a wide variety of ions and molecules[9]. Through appropriate functionalization, the selectivity of the biosorbents can be "engineered". This enables customization of their affinity toward the intended pollutants and allows for their use in multiple applications[20]. For instance, acid modification of walnut shells was found to improve their biosorption of metals through the enhancement of sulfur-containing functional groups on their surfaces that facilitate complexation with metals[21] [Figure 2]. Agricultural waste may consist of crop waste, such as corn straw, rice and wheat, and sugar cane, as well as fruit waste, such as peels, pits, and shells. These types of waste are usually rich in cellulose, hemicellulose, and lignin, making them good candidates for activated carbon production [Figure 1][16].

Biosorbents can be physically synthesized from dried pulverized organic waste or chemically by converting organic waste to activated carbon [Figure 3]. The feasibility and adsorption efficiency of activated carbon production depends on the carbon content of the waste and the carbonization process[11]. The carbonization procedure frequently involves a combination of thermochemical and/or chemical methods to produce cost-effective materials with high adsorption capabilities [Figure 3]. The production of activated carbon primarily involves pyrolysis, a thermochemical method conducted at elevated temperatures or combined methods that include the use of chemical substances and a somewhat reduced operating temperature [Figure 3][23]. At times, pyrolysis becomes the sole technique for carbonizing raw materials, especially when the biomass contains harmful compounds. However, if a combined carbonization approach is employed, intricate chemical reactions can take place effectively. Typically, the most desirable properties are achieved by activating with bases, acids, and mineral acid salts[24].

The thermochemical method is essentially a thermal treatment, which involves heating at temperatures ranging from 827 to 1,027 °C using air, steam, or supercritical CO_2. Meanwhile, the combined method requires both thermochemical and chemical procedures[25]. While pyrolysis results in less environmental pollution and has a lesser environmental impact, biosorbents with superior adsorption characteristics are typically produced through chemical activation[25]. Comparatively, the physical method of producing biosorbent is simpler, often involving drying, crushing, and pulverizing of biomass or subjecting the biomass to ultrasound treatment [Figure 3]. This method can be combined with chemical treatments, as with pyrolysis, to enhance the biosorbent's functional groups [Figure 3][16].

Adsorption of organic pollutants

Wastewater contains a myriad of organic pollutants, including pesticides, pharmaceutical and personal care products, polycyclic aromatic hydrocarbons (PAHs), dyes, and microplastics. Dyes are significant contaminants for wastewater from textile, paper-making, leather, and cosmetic industries, though their presence in municipal wastewater is not generally significant. Furthermore, they have received substantial attention in studies on applying biosorbents for organic pollutant removal[10,14,26]. They are, thus, included in the list of organic pollutants in this review. Some of the organic pollutants are persistent organic pollutants[27]. These are artificial organic chemical compounds that are either intentionally or

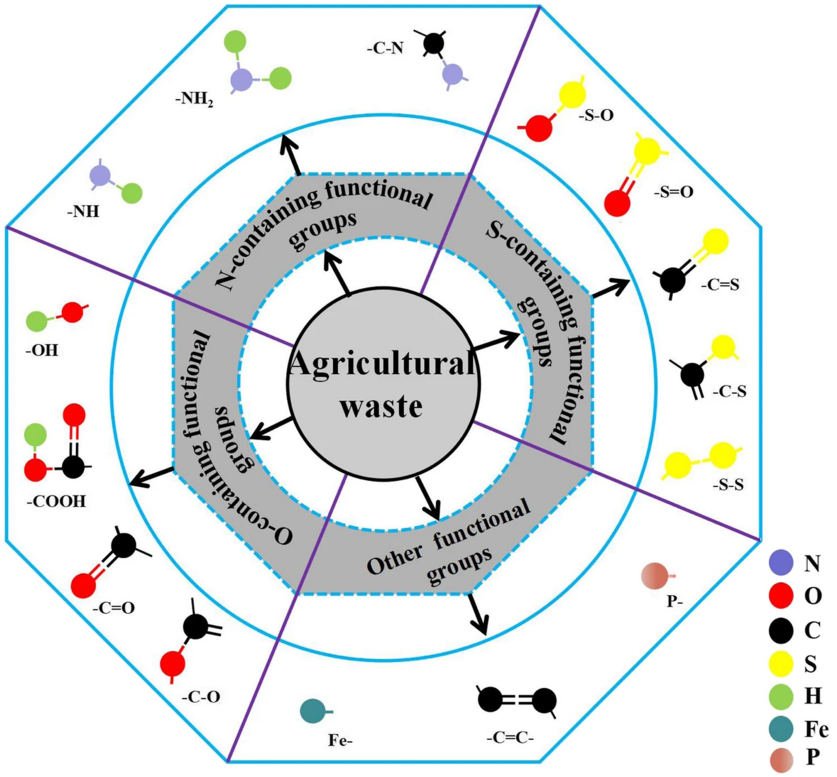

Figure 2. Various functional groups on the surface of biosorbents[22].

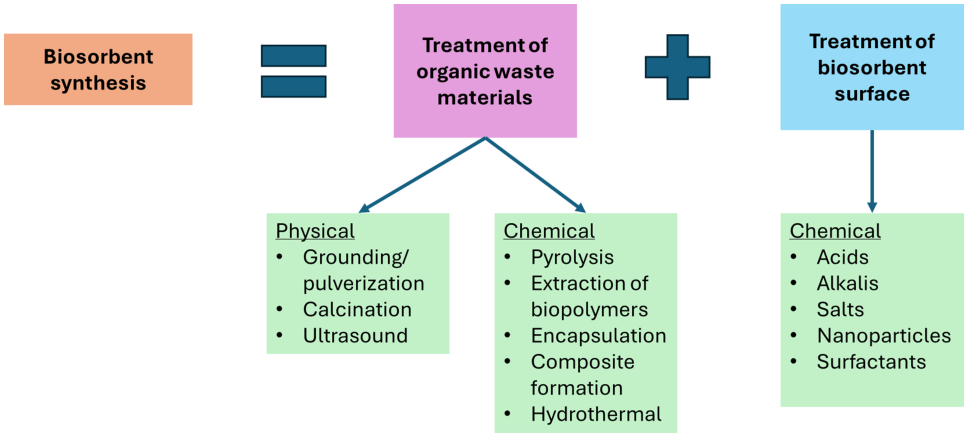

Figure 3. Synthesis of biosorbents. Organic waste can be physically or chemically treated to obtain the biosorbent materials, which are subsequently subjected to surface treatments or modifications to increase the adsorption capacity.

unintentionally produced. They pose a significant environmental threat due to their toxicity, persistence, ability to be transported over long distances, bioaccumulation, and potential harmful effects on living organisms[26]. The United States Environmental Protection Agency (USEPA) has defined the organic priority pollutants, which comprise multiple substances such as halogenated hydrocarbons, organochlorine pesticides, monocyclic aromatics, phthalate acid esters, PAHs, polychlorinated biphenyls, *etc.*[28]. Organic micropollutants are another group of organic pollutants characterized by their small sizes and low environmental concentrations. They could be persistent or non-persistent. Microplastics, polychlorinated

biphenyls, and certain pharmaceutical products are examples of persistent organic micropollutants[29]. These pollutants can be removed from wastewater via adsorption, with activated carbon being the most common sorbent.

Biosorbents from husks and shells

Biosorbents can serve as a sustainable alternative to activated carbon. As aforementioned, there is a wide variety of agriculture-derived waste types, such as husks, shells, peels, and stone fruits. Husks and shells are plentiful agricultural by-products that possess significant stability, extensive specific surface area, and good mechanical strength. They are primarily made up of lignin and polysaccharides[11]. Numerous studies have demonstrated the effectiveness of sorbents derived from these materials under experimental conditions for the biosorption of metal ions and organic compounds comprising bisphenol A, carbamazepine, diclofenac, naproxen, quinoline, and tetracycline[14]. They have also been used to reduce chemical oxygen demand (COD) and remove dyes. The capacity of crude and modified coconut shells, as well as waste coconut fiber, to remove various pollutants from wastewater samples has been studied. These pollutants include Congo Red (98.9%), Rhodamine-B (99.2%), phenol (64%), and diamond green (85%)[30-33]. Studies found that these biosorbents performed well in treating complex samples such as landfill leachate and industrial effluents, where multiple pollutants can be eliminated concurrently[34,35] [Table 1]. When landfill leachate samples were subjected to 5- to 25-time dilution, the efficiency of removing COD and dye using rice husk increased significantly[34] [Table 1].

However, a study also showed that the efficiency of removing certain pharmaceuticals like carbamazepine and diclofenac from both ultrapure water and wastewater effluents was lower in real samples compared to simulated ones. This lower efficiency could be due to the presence of solid substances and soluble organic compounds in wastewater, which give rise to a matrix effect, evident when the presence of numerous components in a sample affects the accuracy of measuring the analyte concentration[50,51]. Another study demonstrated the effective removal of crystal violet (97%) using *Camellia oleifera* shells modified with citric acid. The removal was due mainly to surface adsorption, intraparticle diffusion, and chemical interactions between the dye and the surface functional groups of the biosorbent[36]. Furthermore, it has been highlighted that rice husk can be used in constructed wetlands augmented with microorganisms to remove textile dyes from industrial wastewater. The efficiency of removing Black-5 azo dye using the biosorbent varied with the pH of the pollutant solution, achieving elimination efficiencies of 84%, 52%, and 55% at pH 4, 7, and 10, respectively[37]. This indicates the potential of using biosorbents together with other strategies to improve pollutant removal efficiency.

Biosorbents from fruit and vegetable peels

Additionally, fruit and vegetable peels are popular alternatives for biosorbents. Examples of common waste peels used for this purpose include avocados, bananas, lemons, oranges, pomegranates, and potatoes. These waste peels are commonly used for biosorption probably because they are widely consumed globally[14]. Due to their availability, they provide a cost-effective option for biosorbent production. Research has shown that raw banana peels and orange peels have a wide range of practical uses in treating wastewater. For instance, raw banana peels have been found to be effective in removing various organic contaminants such as PAHs, phenol, benzoic acid, salicylic acid, and Rhodamine-B dye from both simulated and real wastewater samples, with efficiencies more than 90%, except for phenol (60%) and benzoic acid (88.7%)[38,40,52,53].

Similarly, orange peel has displayed its potential in removing methylene blue and reducing total dissolved solids (TDS) and COD in real wastewater samples at efficiencies of 90%, 92.3%, and 88.8%, respectively[41] [Table 1]. In all these applications, the main interaction explaining the effectiveness of these materials has

Table 1. Adsorption performance of biosorbents from agricultural waste for organic pollutants

Biosorbent	Contaminant	Initial pollutant concentration	pH	Temperature	Reaction time	Sorption capacity	Removal (%)	Ref.
Sulfuric acid-activated coconut husk	Rhodamine-B	200-1,000 mg/L	7	30-50 °C	120 min	16,666.7 mg/g	99.2%	[31]
Coconut shell	Phenol	50-250 mg/L	NA	25 °C	2 h	18.1 mg/g	64%	[32]
Coconut shell	Brilliant green dye	10-100 mg/L	2-12 (optimal 8)	NA	100 min	NA	85%	[33]
H_3PO_4-activated rice husk	COD, color, NH_4^+	880 mg/L COD; 18 mg/L NH_4^+	NA	30 °C	24 h	NA	80% COD; 90% color; 100% NH_4^+	[34]
Citric acid-modified *Camellia oleifera* shell	Crystal violet	50-300 mg/L	7	20-40 °C	5 h	141.8 mg/g (40 °C)	97%	[36]
Rice husk mixed with soil at a 1:1 ratio	Black-5 azo dye	100 mg/L	4	25 °C	48 h	NA	84%	[37]
Banana peel	Naphthalene, fluorene, phenanthrene	20 mg/L	7	25 °C	1.33 h	333.3 mg/g naphthalene; 285.7 mg/g fluorene; 217.4 mg/g phenanthrene	95% naphthalene; 98% fluorene; 98% phenanthrene	[38]
Pomegranate peel	C.I. reactive yellow	125 mg/L	2	25 °C	1-100 min	209.7 mg/g (at 53.9 min)	75%	[39]
NaOH-treated avocado peel	Alcian blue; brilliant blue; neutral red; methylene blue	5-200 mg/L	2-12	30 °C	0-25 h	31 mg/g Alcian blue; 3.2 mg/g brilliant blue; 6.9 mg/g neutral red; 62.1 mg/g methylene blue	99.6% Alcian blue; 99.5% brilliant blue; 99.8% neutral red; 99.7% methylene blue	[40]
HNO_3-activated orange peel	Methylene blue	5-60 mg/L	NA	25 °C	5-120 min	NA	No mixing: 84.2% (5 min); 88.7 (120 min) With mixing: 90.2% (5 min); 94.2% (120 min)	[41]
Raw orange peel (A); orange peel modified with instant controlled pressure drop (B); orange peel modified with NaOH and instant controlled pressure drop (C); orange peel modified with NaOH, citric acid, and instant controlled pressure drop (D)	RR-272; phenol	RR-272 (50-800 mg/L); phenol (0-2,000 mg/L)	pH 5 (RR-272); pH 7 (phenol)	25 °C	NA	RR-272: 2.1 mg/g (A); 15.2 mg/g (B); 12.2 mg/g (C); 39.8 mg/g (D) Phenol: 4.6 mg/g (A); 17.8 mg/g (B); 34.3 mg/g (C); 14.9 mg/g (D)	640% (RR-272, raw orange peel); 1,812% (phenol, raw orange peel) (The percentage refers to adsorption capacity in comparison to orange peel)	[42]
Raw cactus peel	BR46	200 mg/L	6	25 °C	180 min	82.6 mg/g	NA	[43]
Barley straw with H_3PO_4 and microwave treatments	Norfloxacin	10-100 mg/L	7.03	25-55 °C	1-168 h	349 mg/g (25 °C); 359 mg/g (35 °C); 387 mg/g (45 °C); 441 mg/g (55 °C)	95.5% (25 °C); 93.3% (35 °C); 95.4% (45 °C)	[44]
Corncob and sugarcane bagasse biochar	Chlortetracycline	200 mg/L	3-7	25 °C	48 h	36.1 mg/g (pH 7, corncob); 25.7 mg/g (pH 7, sugarcane bagasse)	59.6% (pH 5, corncob); 84,5% (pH 4, sugarcane bagasse)	[45]
Citric acid functionalized magnetic graphene oxide coated corn straw (CA-mGO; CA-mGO_5CS; CA-mGO_{10}CS)	Methylene blue	1,000 mg/L	8	25 °C	3 h	276.5 mg/g (CA-mGO); 300.3 mg/g (GA-mGO_{10} CS); 315.5 mg/g (GA-mGO_5CS)	77% (CA-mGO); 82.4% (CA-mGO_{10}CS); 84% (CA-mGO_5CS)	[46]

Rice straw and *Phanerochaete chrysosporium*	Methylene blue	400 mg/L	5	37 °C	24 h (straw); 12 days (straw + *P. chrysosporium*	140.4 mg/g	88% (28.4% biodegraded and 59.6% adsorbed)	[47]
Mango stones treated with iron oxide magnetic nanoparticles	Chloroquine; sertraline	20 mg/L	5-6	43 °C	24 h	49.4 mg/g (chloroquine); 64.8 mg/g (sertraline)	86% (contaminant mixture)	[48]
Algerian date stone	Methylene blue	10-100 mg/L	7	25-55 °C	10-120 min	6.7 mg/g (25°C)	98.8% (60 min)	[49]

NA: COD: chemical oxygen demand; RR-272: reactive red 272.

been identified as chemisorption, where the contaminant molecules form strong chemical bonds with the surface of the biosorbents. A study showed that using NaOH and citric acid to modify orange peel in moderate conditions, along with thermal activation, greatly increased the efficiency of phenol biosorption by over 18 times and enhanced the removal of reactive-red dye (RR-272) by more than 6 times, indicating that chemical modification could potentially enhance the efficiency of biosorption[42] [Table 1]. Specifically, NaOH modifies and improves the structural properties of biosorbents, increasing their surface area and porosity, while citric acid can introduce more functional groups onto the surface of biosorbents. Akkari *et al.* utilized dried ground raw cactus fruit peel as an inexpensive and eco-friendly sorbent to eliminate basic red 46 (BR46) from water-based solutions. They found that the maximum adsorption capacity was 82.58 mg/g. The biosorption isotherm was better represented by the Freundlich model, while the kinetic biosorption data were well described by the pseudo-second-order model. The Freundlich model describes the adsorption process on heterogeneous surfaces, which is not limited to a monolayer. The pseudo-second-order kinetics suggest chemisorption as a likely mechanism. Moreover, their reuse study demonstrated that the biosorbent could be regenerated after three successive cycles using HCl as a chemical reagent. As with other studies, the biosorption process was spontaneous and exothermic[43] [Table 1].

Biosorbents from straws and corn cobs

Rice, wheat, and barley straws, as well as corn cobs, are frequently utilized as biosorbents. They are obtained from the fibrous remains of the plant body and are rich in lignin, cellulose, and hemicellulose. These agricultural residues have sparked considerable interest due to being valuable by-products of the plant cultivation process. They are notable not only for their abundant supply but also for their cost-effectiveness and widespread availability[54]. Barley straw was treated with phosphoric acid and exposed to microwave radiation to create a modified adsorbent. 0.5 mg of the modified barley straw was then tested for its effectiveness in removing 100 mg/L of norfloxacin at a neutral pH. The results showed a sorption capacity of 349 mg/g at 25 °C, 359 mg/g at 35 °C, 387 mg/g at 45 °C, and 441 mg/g at 55 °C[44] [Table 1]. Two types of agricultural waste, corn cob and sugar cane, were utilized to remove the organic pollutant chlortetracycline. The reported removal efficiency for both biosorbents was over 90% at an optimal reaction time of 20 h[45] [Table 1]. Corn straw modified with citric acid and graphene oxide proved effective in adsorbing at least 80% of methylene blue at pH 8[46] [Table 1].

Using biosorption in conjunction with biodegradation provides another efficient option for eliminating methylene blue from wastewater. For example, when a mixture of 1.0 g of rice straw and a 5 mL suspension of the white rot fungus (*Phanerochaete chrysosporium*) spore was used, 88% of methylene blue with an

initial concentration of 400 mg/L was removed at pH 5.0. This removal process involved 28.4% of the total removed dye being broken down by *P. chrysosporium*, while the remainder was subjected to biosorption by fungal cells and biodegraded rice straw[47] [Table 1]. Similarly, a study conducted with a combination of rice straw and *Bacillus subtilis* showed at least 90% of methylene blue with a starting concentration of 400 mg/L was removed by 0.5 g of rice straw mixed with 0.2 mL of spore suspensions. The pH of the process was maintained at 7. The presence of both *P. chrysosporium* and *B. subtilis* led to an increase in the specific surface area of the straw and the generation of more functional groups, resulting in a considerable enhancement in removing methylene blue. Additionally, microbial biodegradation played a crucial role in the overall dye-removal process[55].

Biosorbents from stone fruits

Biosorbents can also be produced from stone fruits, particularly the seeds of fruits such as avocados, apricots, and peaches, due to their properties. Like other biosorbents derived from crops, fruit stones have high cellulose, hemicelluloses, and lignin content. The surface hydroxyl groups on these biosorbents can facilitate the introduction of various functional groups to enhance biosorption [Figure 2][10]. A study utilized oven-dried ground mango stones treated with iron oxide magnetic nanoparticles (MS-Fe_3O_4) to eliminate chloroquine and sertraline from polluted water[48] [Table 1]. Both contaminants required an optimal adsorbent dosage of 0.01 g. The pH level significantly affected their adsorption on MS-Fe_3O_4, with the most effective outcomes observed within a pH range of 5-6. Analysis of kinetic data suggested that the pseudo-second-order model provided the best fit, with equilibrium attained at 16 and 4 h for chloroquine and sertraline, respectively. Isotherm studies indicated maximum adsorptive capacities of 49.42 mg/g for chloroquine and 64.79 mg/g for sertraline at 318 K, highlighting the favorable impact of elevated temperatures. The Sips model was observed to explain the adsorption more precisely. It combines elements from the Langmuir and Freundlich isotherms and is particularly useful for fitting adsorption data in cases where the adsorption surface is not uniform. It is, therefore, appropriate for systems with both homogenous and heterogenous adsorption sites at low and high adsorbate concentrations, respectively[48]. In another study, Algerian date pits were examined for their utility as a biosorbent to eliminate methylene blue from water solutions. The Freundlich model was determined to provide the most accurate representation of the adsorption isotherm. Upon conducting a kinetic study, it was observed that the pseudo-second-order model effectively described the adsorption process[49] [Table 1]. However, most of the studies on the use of stone fruits as biosorbents center on the biosorption of metal ions. This is further illustrated in the subsequent section.

Biochar and organically derived activated carbon

In addition to drying and pulverization, biosorbents can also be synthesized through carbonization to produce biochar or activated carbon. Carbonization can be performed on a wide variety of organic waste, and like non-carbonized biosorbents, they can be chemically treated to enhance sorption and recyclability[56]. An investigation was conducted to evaluate the effectiveness of using activated carbon derived from coconut shells to remove pentachlorophenol added to aqueous solutions passing through a column. The biosorbent exhibited a notable sorption capacity of 36.82 mg/g, particularly noteworthy for its efficacy under neutral pH and relatively gentle and economically feasible conditions, involving a 6-hour duration of contact and a temperature of 37 °C[57]. Naghdi *et al.* highlighted the effectiveness of activated carbon derived from pine wood in removing carbamazepine, which is commonly found in surface waters, even at very low concentrations ranging from 0.5 to 20 μg/L. This significant adsorption capacity was observed across a broad pH range of 3.0 to 8.0. The study also revealed that nanobiochar exhibited an adsorption capacity of 0.074 μg/g after 3 h of contact time. Furthermore, the results indicated that increasing the pH from 3 to 8 led to an improvement in the adsorption efficiency[58]. In a spiked sample containing an

initial methylene blue and Rhodamine B concentration of 1,000 mg/L at neutral pH, the cationic dyes were effectively captured using 20 mg of porous carbon derived from banana peels. This carbon material exhibited impressive sorption capacities of 744.39 mg/g for methylene blue and 520.29 mg/g for Rhodamine B, showcasing its potential for efficient dye sequestration[59]. In a study, carbonized yam peels were used to create a magnetic adsorbent to remove 2,4-dichlorophenoxyacetic acid (2,4-D). The activated carbon was modified using magnetite nanoparticles. The results showed higher adsorption at high 2,4-D concentration, low pH, and room temperature. The process was found to be exothermic and spontaneous. The magnetized activated carbon can be easily separated from water, and it retained 60% of its adsorption capacity after five cycles[56].

Implications

A wide variety of agricultural waste has been demonstrated to be potential adsorbents for various organic pollutants, particularly dyes and pharmaceuticals, and in some cases, COD in general. This waste often contains functional groups such as hydroxyl, carboxyl, and amino groups, which can facilitate the adsorption of a wide range of contaminants [Figure 2]. Using this waste aligns with circular economy principles, as it reduces waste, promotes resource recovery, and minimizes reliance on non-renewable resources[60]. The perishable nature of agricultural waste does introduce some challenges but generally does not significantly limit its potential as an adsorbent. In fact, with proper processing and treatment, perishable agricultural waste can be effectively transformed into stable, high-performance adsorbents[60]. Perishable agricultural waste can be converted into biochar or activated carbon through pyrolysis or carbonization. It can also be dried and pulverized. These processes transform short-lived organic materials into valuable, long-lasting adsorbents.

However, these biosorbents have varied biosorption efficiencies under different conditions and when applied to different pollutants. In practical use, owing to the different physicochemical characteristics of wastewater, optimization experiments are crucial to increase the performance of these biosorbents [Figure 4]. Biosorption has shown the flexibility of integrating with other methods, such as biodegradation, for better performance. Biosorbents can be synthesized physically, thermochemically, or via both methods, with different potential chemical treatments to enhance their surface functional groups for better performance. In most instances, biosorption of organic pollutants is characterized by the Langmuir or Freundlich isotherms and the pseudo-second-order kinetics, with chemisorption being more predominant.

Adsorption of inorganic pollutants

There are comparatively more studies conducted on the adsorption of inorganic pollutants, particularly metal ions, using biosorbents from agricultural waste. The remarkable performance of these biosorbents has been confirmed through their ability to eliminate over 90% of metals, including Cu^{2+}, Cd^{2+}, Pb^{2+}, and Zn^{2+} [Table 2][61,62]. Among the tested metals, Pb^{2+} appears to be the most examined[71,63]. The primary mechanism driving biosorption is physisorption.

Biosorbents from husks and shells

A study compared the effectiveness of using 0.5 g/L of dried and ground almond, hazelnut, peanut, pistachio, and almond shells to extract Cd, Pb, and Hg from polluted water. Hazelnut shells demonstrated the highest potential for this purpose when tested with single and multiple contaminants in mineral water [Table 2][63]. Variations in the effectiveness of the different shells were attributed to the varying proportions of their cellulose, hemicellulose, and lignin. The presence of a complex heavy metal mixture in the water caused a decrease in Cd removal, likely due to reduced electrostatic interaction and complexation with anions such as Cl^-. When all three contaminants were present in the water, they competed with each other,

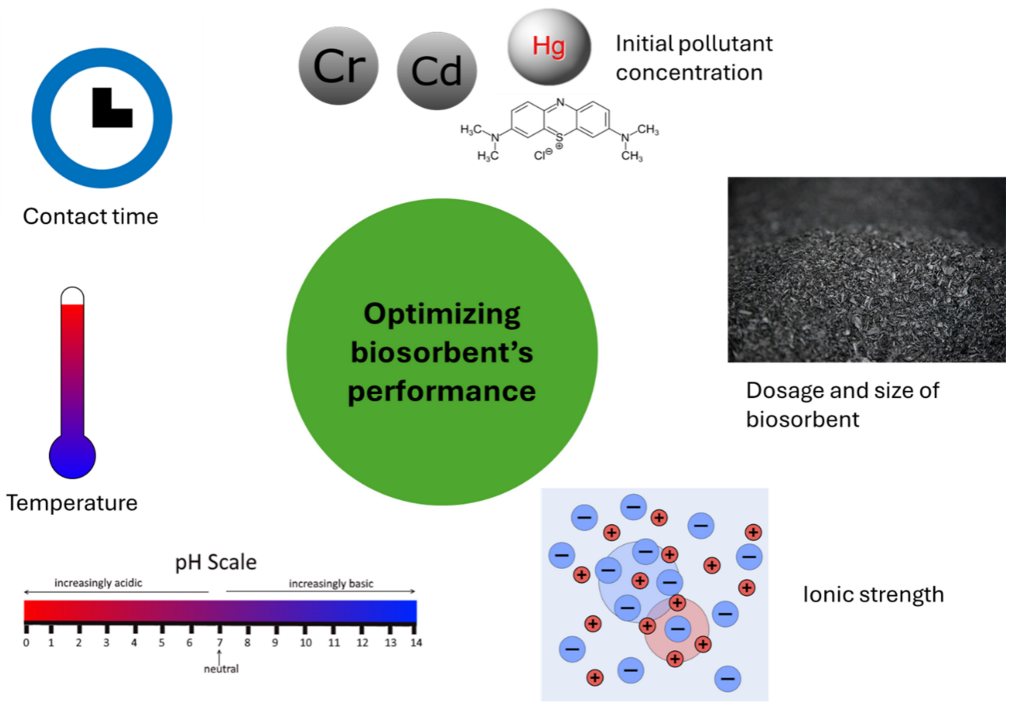

Figure 4. Optimization of a biosorbent's performance typically takes into account the temperature, pH, and ionic strength of the reaction medium, the initial pollutant concentration, the dosage and size of the biosorbent, and the contact time between the sorbate and biosorbent.

with lead demonstrating a greater affinity for the sorbent than mercury[63].

Walnut shell powder was subjected to chemical modification using sodium hydroxide and citric acid to create modified biosorbents with numerous pores serving as binding centers[64]. Analysis of Cr^{6+} adsorption at pH 2 using isotherm and kinetic models revealed that the adsorption behavior of Cr^{6+} was effectively explained by the Langmuir model, indicating the formation of a monolayer of the adsorbate on the surface of the biosorbents. The citric acid-modified biosorbent exhibited the highest maximum adsorption capacity for Cr^{6+} at 75.26 mg/g, followed by the sodium hydroxide-modified biosorbent at 69.56 mg/g, and walnut shell powder at 64.82 mg/g [Table 2][64]. Sodium hydroxide-treated dried and pulverized rice hulls were employed for the concentration, extraction, and reclamation of metal ions in both batch and column operations. The treatment notably enhanced the effectiveness of removing various metal ions[72]. The process was rapid for several ions (Ba, Co, Cd, Cu, Ni, Sr, and Zn), showing promise for efficiently removing low-level metal ions at high throughput. The primary removal mechanism is presumed to be the electrostatic attraction between the rice hulls and the metal ions. A pH level of 5 was found to be optimal for the biosorption of most metal ions[72]. This forms a significant difference from the adsorption of organic pollutants using husks and shells, which often relies on interactions such as van der Waals forces, hydrogen bonding, and hydrophobic interactions. However, polar organic pollutants also exhibit electrostatic interactions with biosorbents. Like organic pollutants, chemical modifications tend to improve the adsorption efficiency of biosorbents for metal ions.

Biosorbents from fruit and vegetable peels

The removal of Cd^{2+}, Cr^{3+}, and Pb^{2+} ions from real industrial wastewater at pH 6 using banana peel resulted in high biosorption efficiencies. Specifically, the removal efficiencies were 97.77% for Cd^{2+}, 98.9% for Cr^{3+},

Table 2. Adsorption performance of biosorbents from agricultural waste for inorganic pollutants

Biosorbent	Contaminant	Initial pollutant concentration	pH	Temperature	Reaction time	Sorption capacity	Removal (%)	Ref.
Rice husks	Cu^{2+}	100-500 mg/L	2-7	25 °C	240 min	12.8 mg/g	100% (4 g biosorbent, 100 mg/L Cu^{2+})	[61]
Risk husks treated with CS_2 to enhance sulfur-bearing groups	Cd^{2+}	250 mg/L	7	15 °C	120 min	138.9 mg/g	90%	[62]
Peanut, hazelnut, pistachio, walnut and almond shells	Cd, Pb, and Hg in single-element solutions	2 μmol/L	6.5-7	22 °C	48 h	Hazelnut shell (408-449 μg/g Cd; 693-767 μg/g Pb; 656-731 μg/g Hg)	Peanut shell (97% Cd, 92% Pb, 65% Hg); hazelnut shell (98% Cd, 97% Pb, 90% Hg); pistachio shell (88% Cd, 91% Pb, 77% Hg); walnut shell (93% Cd, 72% Pb, 77% Hg); almond shell (81% Cd, 89% Pb, 79% Hg)	[63]
Walnut shell powder modified with alkali and citric acid	Cr^{6+}	20-120 mg/L	2	25 °C	40 min	64.82 mg/g (unmodified); 69.56 mg/g (alkali-modified); 75.25 mg/g (citric acid-modified)	73.4% (unmodified); 69% (alkali-modified); 63.8% (citric-acid-modified)	[64]
Banana peel	Cd^{2+}, Cr^{3+}, Pb^{2+}	Cd^{2+} (0.0003 mg/L); Cr^{3+} (0.05 mg/L); Pb^{2+} (0.1 mg/L)	6	25 °C	60 min	Cd^{2+} (2.23 mg/g); Cr^{3+} (1.1 mg/g); Pb^{2+} (3.1 mg/g)	Cd^{2+} (97.8%); Cr^{3+} (98.9%); Pb^{2+} (96.9%)	[65]
Banana peel	Th, U	U (55.8 mg/L); Th (18.8 mg/L)	4.1	25 °C	25 h	U (8.99 mg/g); Th (0.0362 mg/g)	U (70%); Th (100%)	[66]
Orange peel and *Aspergillus niger*	Cd^{2+}, Pb^{2+}	50-250 mg/L	5-6	30 °C	300 min	Simulated sample: Cd^{2+} (370.4 mg/g, pH = 5); Pb^{2+} (286 mg/g, pH = 6)	Simulated sample: Cd^{2+} (96%, 300 min); Pb^{2+} (90%. 300 min) Competitive sorption: Cd^{2+}or Pb^{2+} (65%, 60 min)	[67]
Unmodified and citric acid-treated *Byrsonima crassifolia* endocarp	Cd^{2+}, Ni^{2+}	25-100 mg/L	5	30 °C	24 h	Unmodified in single solution: Cd^{2+} (1.71 mg/g); Ni^{2+} (0.95 mg/g) Modified in single solution: Cd^{2+} (3.10 mg/g); Ni^{2+} (2.45 mg/g)	All biosorbents in binary solutions: Cd^{2+} (7.5-75%); Ni^{2+} (8.5%-97.4%)	[68]
Peach stone modified by citric acid	Cd^{2+}, Cu^{2+}, Pb^{2+}	6-120 mg/L	2-7	30 °C	180 min	Cd^{2+} (37.5 mg/g), Cu^{2+} (32.2 mg/g), Pb^{2+} (118.8 mg/g)	Cd^{2+} (89.6%), Cu^{2+} (97.2%), Pb^{2+} (93.4%)	[69]
Avocado seeds	As^{3+}	5-30 mg/L	6	25-40 °C	120 min	93.75 mg/g	65% at 20 mg/L As^{3+}	[70]

and 96.9% for Pb^{2+} [Table 2][65]. It is worth noting that raw banana peels were found to have the same purification efficiency when used as a separation medium for thorium and uranium in laboratory and mine wastewater samples from South Africa [Table 2][66]. Orange peel has shown great potential for various applications, including the removal of metal ions such as Cu^{2+}, Fe^{2+}, and Mn^{2+}[73]. In a study, a hybrid sorbent consisting of orange peel and the fungus *Aspergillus niger* demonstrated a 65% elimination efficiency in the simultaneous removal of Cd^{2+} and Pb^{2+} ions from real water samples with a pH of 5.5. This remarkable performance was achieved using just 0.5 mg of the hybrid sorbent [Table 2][67]. A study utilized banana, orange, and potato peels to create biosorbent beads for removing heavy metals from a solution. The beads showed higher uptake of As and Pb at neutral pH but decreased uptake of Cd, Cu, Hg,

and Ni at higher pH. At equilibrium, the beads could remove up to 92%, 87%, 84%, and 90% of Cd, Cu, Hg, and Ni, respectively. The adsorption was primarily physisorption, following the pseudo-first-order model. Banana peel and orange peel beads were more efficient than potato peel beads due to their higher surface heterogeneity[74]. Another study used melon peel as a biosorbent to effectively remove Cu, Cd, and Pb from water. The NaOH-modified melon peel biosorbent showed increased functional groups and surface area, which are important for heavy metal adsorption. The Langmuir isotherm model fitted the heavy metal biosorption better. Maximum capacities for adsorbing a single metal (Cu, Cd, and Pb) were 77.76, 76.16, and 191.93 mg/g, respectively[75].

Biosorbents from straws

As for crop-derived sorbents, three types of rice straw-based biosorbents - raw rice straw, alkali-treated rice straw, and rice straw biochar - were analyzed for their ability to adsorb Cd^{2+} and Zn^{2+}. Raw rice straw showed the highest adsorption, with 20.9% for Cd^{2+} and 8.4% for Zn^{2+}[76]. The optimum pH for Cd^{2+} adsorption was 5 for all three adsorbents, while the optimum pH for Zn^{2+} was 4. The adsorption capacity best fitted the Freundlich isotherm for all adsorbents, indicating a heterogeneous property and a favorable adsorption process[76]. In a recent study, an environmentally friendly straw-based adsorbent was developed via acrylamide and citric acid surface modification to remove $Cr_2O_7^{2-}$ and Cu^{2+} from water. The adsorption capacities of the modified biosorbent for $Cr_2O_7^{2-}$ and Cu^{2+} in a mixed system saw significant increases (196% and 151%, respectively), attributed to a collaborative effect through electrostatic attraction. Both the functional groups and adsorbed pollutants served as adsorption sites[77].

Since crop-based biosorbents are rich in lignin, hemicellulose, and cellulose, studies have also been conducted to extract these materials for biosorption. Yu *et al.* investigated the performance and stability of carboxyl group-modified straws and their cellulose, lignin, and hemicellulose extracts for removing Pb^{2+}[78]. It was revealed that modification significantly improved both the adsorption capacity and stability of the materials, with modified hemicellulose showing the highest adsorption capacity, followed by both modified lignin and straw and, finally, modified cellulose. The optimal pH range for Pb^{2+} adsorption was found to be between 4.0 and 5.0, with modified cotton and rape straw being more stable than modified maize straw within this range[78]. Mohammadabadi and Javanbakht used ultrasonic and gelation-solidification methods to extract lignin to synthesize a biocomposite as a biosorbent for lead[79]. The biocomposite beads were effective in lead removal from water under various conditions. Its maximum adsorption capacity was reported to be 344.83 mg/g at an initial lead concentration of 210 mg/L, a temperature of 328 K, and a pH level of 5. The study also investigated the adsorption kinetics and isotherms and found that they followed the pseudo-second-order kinetic model, as well as the Langmuir and Temkin isotherms[79]. This aligns with frequently reported Langmuir isotherm in other biosorption studies, though other isotherm models were also reported at times.

Biosorbents from stone fruits

As with organic pollutants, attempts have been made to synthesize biosorbents for inorganic pollutants from seeds and fruit stones. Monroy-Figueroa studied the effectiveness of using natural and chemically modified *Byrsonima crassifolia* endocarp for removing Cd^{2+} and Ni^{2+} ions[68] [Table 2]. Treating the biomass with citric acid improved its ability to adsorb these ions under competitive conditions. The citric acid modification increased the functional groups on the biosorbent's surface through reactions with lignocellulosic materials. The process involves an ion-exchange mechanism, potentially involving surface carboxylic, hydroxyl, phenolic groups, and Ca^{2+} replacement. This is supported by the findings that the main elements found in the biomass were C, O, and Ca^{2+}[68]. The peach stone modified with citric acid has been found to effectively remove Cu^{2+} at a sorption capacity of 118.8 mg/g, Pb^{2+} at 93.4 mg/g, and Cd^{2+} at

89.6 mg/g from solutions. This was observed under conditions of an initial metal ion concentration of 200 mg/L, pH 5, and an adsorbent mass of 0.1 g [Table 2][69]. Additionally, when avocado pits were modified with citric, sulfuric, or tartaric acid, they exhibited enhanced sorption capacities between 3.3 and 21.8 mg/g for Cd^{2+}, Cu^{2+}, Ni^{2+}, Pb^{2+}, Zn^{2+}, as compared to untreated avocado seeds, with sorption capacities between 2.5 and 5.6 mg/g[70].

While acids, whether mineral or organic, are commonly used to functionalize the surfaces of biosorbents, basic functionalization has also been reported. For instance, Šoštarić *et al.* utilized apricot stone waste, which was treated with a 1.0 mol/L NaOH solution, to remove Cu^{2+}, Pb^{2+}, and Zn^{2+} (with an initial concentration of 400 mg/L) from a solution at a pH of 5.0[80]. The process involved using 0.5 g of biosorbent, resulting in the elimination of 81% of Cu^{2+}, 87% of Pb^{2+}, and 97% of Zn^{2+}[80]. A recent study used jackfruit seed powder to efficiently remove As^{5+}, Cd^{2+}, and Cr^{6+} from water. The study optimized various conditions and found that the powder worked best at an adsorbent weight of 0.5 g, initial metal concentrations of 40 μg/L (As^{5+}), 30 mg/L (Cd^{2+}), and 30 mg/L (Cr^{6+}), temperatures between 25 and 30 °C, and a pH range of 7-7.5[81]. The results aligned well with Langmuir (As^{5+}, Cd^{2+}, Cr^{6+}), Freundlich (As^{5+}, Cr^{6+}), and Dubinin-Radushkevich (Cd^{2+}, Cr^{6+}) isotherm models, indicating that heavy metal biosorption may not be limited to a specific isotherm model[81]. Nonetheless, the Langmuir isotherm has been more consistently mentioned than other isotherms. A study used dried and powdered *Phyllanthus emblica* fruit stones to remove Pb^{2+} and Cd^{2+} from water through adsorption. The biosorbent removed 80% of both heavy metal ions within 60 min at pH 6. The process was determined to be endothermic, spontaneous, and feasible[82].

Biochar and organically derived activated carbon

Like organic pollutants, biosorbents are commonly subjected to chemical modifications, particularly treatments with acids and alkalis, to increase their sorption capacities. The lignocellulosic components of agricultural waste have been extracted for the biosorption of inorganic pollutants. Thermochemical treatment has also been employed to synthesize biosorbents in the form of biochar for inorganic pollutant removal. A type of magnetic biochar derived from banana peels exhibited high biosorption potential for Cu^{2+} (75.9 mg/g), Hg^{2+} (83.4 mg/g), and Zn^{2+} (72.8 mg/g) in individual systems at pH 6 within 3 h[83]. However, in multi-component systems, Hg^{2+} significantly inhibited the biosorption of Cu^{2+} and Zn^{2+}. Additionally, the biochar showed higher biosorption affinity toward Hg^{2+} compared to Cu^{2+} and Zn^{2+}[83]. Dieme *et al.* evaluated the effectiveness of activated charcoal composed of a combination of cashew shells, millet stalks, and rice pomace[84]. This charcoal was enhanced by a coating of Fe^{3+} ions to increase its chemical reactivity. The study focused on its ability to remove oxyanion As (V), and the results showed that the charcoal had an adsorption capacity ranging from 0.236 to 0.301 mg/g[84]. Erabee *et al.* studied the performance of activated biochar from coconut shells, modified with $KMnO_4$, in adsorbing ammonia nitrogen (NH_3–N) and S^{2-} ions from leachate[85]. They found that the positively charged surface of the sorbent influenced the efficient binding of NH_3–N and S^{2-}, leading to biosorption capacities of 0.1979 and 0.0065 mg/g, respectively[85].

Wang *et al.* (2019) explored the pyrolysis of pinewood biomass and natural hematite mixtures at different temperatures (300, 450, and 600 °C) under a nitrogen (N_2) environment to create nanocomposites, designated as P1, P2, and P3, respectively[86]. The findings indicated that P1 could adsorb up to 0.0194 mg/g of Cd^{2+} and 0.0228 mg/g of Cu^{2+}, P2 could adsorb up to 0.0155 mg/g Cd^{2+} and 0.0109 mg/g Cu^{2+}, and P3, 0.0146 mg/g Cd^{2+}, and 0.0125 mg/g Cu^{2+}. It was observed that increasing the pH from 3 to 5 enhanced the sorption of both Cd^{2+} and Cu^{2+}, while higher ionic strength resulted in decreased Cd^{2+} sorption. Notably, Cd^{2+} sorption was reduced by over four times in the presence of Cu^{2+} in a binary system[86]. While the study indicates the effect of pyrolysis temperature on the adsorption capacity of a biosorbent, it also shows that

individual pollutants in a multi-component system could mutually affect the respective adsorption efficiencies. Furthermore, fifteen types of *Caragana korshinskii* biochar were synthesized by pyrolysis at varying temperatures and times in an oxygen-free environment[87]. The results indicated that the pyrolysis at 650 °C for 3 h yielded biochar with the best performance. The maximum adsorption capacities were 220.94 mg/g for Pb^{2+} and 42.43 mg/g for Cd^{2+}. The amounts of biochar producing the optimal adsorption were 3 g/L for Pb^{2+} and 2.2 g/L for Cd^{2+}[87]. At pH ranges of 3-6 and 6-7.5, adsorption was optimum for Pb^{2+} and Cd^{2+}, respectively. The adsorption fitted the Langmuir and Freundlich isotherm models, as well as the pseudo-second-order kinetic model. The adsorption mechanisms were characterized by cationic-π interactions, electrostatic adsorption, intraparticle diffusion, ion exchange, physisorption, and precipitation, among others[87].

Implications

Similar to organic pollutants, the optimal conditions for biosorption of inorganic pollutants considerably vary, depending on the types of biosorbents and pollutants, thus prompting optimization studies specific to wastewater types [Figure 4]. Most of the studies on inorganic biosorption have been conducted on heavy metals due mainly to their toxic effects and environmental concerns. The biosorbents can be tailored via chemical treatments, pyrolysis at different temperatures, combining to form composites, and extraction of specific materials such as lignocellulosic components from biomass. While the Langmuir and Freundlich isotherms generally describe the adsorption behavior, the former is more commonly mentioned. Physisorption, particularly electrostatic interactions, is the predominant sorption mechanism. However, that does not rule out chemisorption, especially surface complexation, which is also common between inorganic pollutants and biosorbents. The presence of other metal ions has been reported to affect the adsorption of certain metal ions in binary or multi-component systems, confirming that a biosorbent may not be equally effective in removing different inorganic species from wastewater. This is also the largest limitation of biosorbent.

The studies reviewed highlight the potential matrix effect in real wastewater due to the complex mixture of substances therein that can influence the action and efficiency of biosorbents. This could be due to competitive adsorption, where different contaminants in wastewater can compete for the same binding sites on biosorbents, and the presence of organic matter, which can coat the biosorbent surface, blocking active sites or changing the surface charge[88]. In addition, high concentrations of dissolved ions can impact biosorbent performance, resulting in "screening effects", which diminish the electrostatic interactions between biosorbents and charged contaminants, especially sorption involving ion exchange. Additionally, when multiple contaminants are present, biosorbents may exhibit varied affinities toward different substances[63]. For instance, Oladipo *et al.* reported that Cu^{2+} and Zn^{2+} biosorption was substantially hampered in the presence of Hg^{2+} in a multi-component aqueous system, while Wang *et al.* observed a reduction of Cd^{2+} biosorption in a binary system[83,86]. Additionally, Aranda-García *et al.* found that the biosorption efficiency of Ni^{2+} by a biosorbent from oak acorn shell decreased as the ionic strength in the solution rose from 2 to 2,000 mM. Higher ionic strength likely introduces competing ions, particularly divalent ions like Ca^{2+} and Mg^{2+}, which have a greater negative effect on Ni^{2+} uptake than monovalent ions such as Na^{+} and K^{+}[89].

The presence of matrix effects in wastewater highlights a major drawback of the current designs of biosorption studies, which often employ single-contaminant systems. These systems may not represent the multi-component nature of real wastewater. Pertile *et al.* pointed out that a NaOH-activated biosorbent from cones at a concentration of 0.1 mol/L adsorbed 96% Ni and 19% Zn from the wastewater of a neutralization station containing other heavy metals over a 20-min treatment. Despite these significant

reductions, the treated water did not conform to the required emission limits for safe discharge into the sewer system[90]. This limits the practical applications of biosorbents and underscores the importance of their modifications and optimization for enhanced performance.

NON-AGRICULTURE-DERIVED ORGANIC WASTE

Though agricultural activities generate a myriad of organic waste, this waste can also come from non-agricultural sources. For instance, household waste containing food scraps and yard waste is a significant source of organic waste. Industrial and municipal solid waste, such as pulp, paper, and textile materials, is also rich in organic matter [Figure 1]. These types of waste have the potential to be converted into biosorbents. Nonetheless, studies on the production of biosorbents from non-agricultural organic waste are fewer than those on biosorbents from agricultural waste. The methods for converting these types of waste into biosorbents are the same as those for agricultural waste, often involving physical, chemical, and thermochemical methods, with or without chemical treatments.

Adsorption of organic pollutants

Huge amounts of fish scales are discarded as solid waste from seafood processing plants and fish markets. This biowaste can be used as a biosorbent in the treatment of wastewater. Devasena *et al.* investigated the use of waste fish scales as a biosorbent to treat pollutants in seafood processing plant wastewater[91]. The treatment showed over 70% removal of nitrite, phosphate, and biological oxygen demand (BOD), and over 95% removal of ammonia and COD. The best adsorption for BOD, COD, and ammonia was attained when a biosorbent dosage of 1 g/100 mL was used, with a contact time of 90 min. The adsorption was characterized by the Langmuir and Freundlich isotherm models[91]. A study examined the process of removing methylene blue from a water solution using fish scales modified with Fe_3O_4 nanoparticles[92]. The biosorbent's structure contained collagen fibers, apatite crystals, and nano-magnetite particles. Methylene blue adsorbed onto the biosorbent through physisorption, which was best characterized with the Sips isotherm equation. Its maximum adsorption capacities estimated with the Langmuir and Sips models were 68.72 and 60.87 mg/g, respectively. Its adsorption kinetics fitted the pseudo-second-order model[92]. Neves *et al.* examined the impact of NaCl and surfactant on the removal of reactive blue 5G dye using unmodified fish scales[93]. Using phenomenological modeling, the researchers found that despite the nonporous nature of the fish scales, they displayed a notable adsorption capacity of approximately 291 mg/g. This high adsorption capacity is attributed to the strong affinity between the adsorbent and the adsorbate, as well as the adsorption of dye aggregates onto the biosorbent. The addition of NaCl and a surfactant raised the sorption capacity to 291 and 299 mg/g, respectively[93]. These findings highlight fish scales as a viable biosorbent.

Food waste containing pollutants is a major concern for disposal. Traditional methods for handling food waste are not effective when the waste contains harmful chemicals such as As^{3+}. Vakili *et al.* recommend using an alkali pre-treatment process to decontaminate the waste and reutilize it as an adsorbent for wastewater treatment[94]. The research findings indicate that treating the contaminated food waste with 0.8 M NaOH at 60 °C for 4 h successfully removes As^{3+} with an impressive removal efficiency of 99.8%. The alkali-treated waste displayed a strong adsorption capacity for methylene blue, achieving an adsorption capacity as high as 534.6 mg/g when the pH was 9 and the adsorbent dosage was 40 g/L[94]. The adsorption aligned with the intraparticle diffusion and Temkin models. Additionally, the adsorbent can be reused for not less than 4 cycles without losing its efficiency in removing methylene blue substantially, thus highlighting its practical potential[94]. Attempts were made to utilize brewery residue for organic pollutant removal. A study investigated the use of brewery residue (*Saccharomyces pastorianus*) to remove methylene blue from water[95]. The findings showed that immobilizing *Saccharomyces pastorianus* in sodium alginate

resulted in effective dye removal. Microencapsulation of the residue, a pH of 9, a biosorbent dose of 5.28 g/L, and a contact time of approximately 100 min resulted in the most effective sorption[95]. The Langmuir model best represented the biosorption isotherm. The maximum biosorption capacity estimated with the model was 188.7 mg/g at room temperature. The biosorption mechanism was chemical or physical in nature and was found to be influenced by biosorbent particle size[95].

Attempts to use the shells of marine organisms to adsorb organic pollutants show varying results. The potential of using the shells of *Corbicula fluminea* as a natural sorbent for treating toxic olive oil mill waste was studied[96]. The results showed that the shells were not effective in adsorbing chemicals from mill waste, particularly aromatic structures, after a 14-day treatment. An increase in pH and COD levels of the mill waste and spectroscopy data implied that the shell dissolved partially[96]. In a different study, snail shell dust from *Bellamya bengalensis* was utilized to treat industrial wastewater[97]. The study examined variations in multiple water quality parameters, including BOD, COD, dissolved oxygen, pH, and the levels of inorganic ions, after the treatment. The results indicated that with a 15 g/L application rate over 4 days, all parameters except dissolved oxygen displayed a significant decrease in concentration, in contrast to the findings of the previous study using the shells of *Corbicula fluminea*[97]. A study used shrimp shell chitin to remove COD and polyphenols from olive mill wastewater[98]. The optimized conditions for maximum removal were found to occur at a pH of 12, an adsorbent concentration of 10 g/L, a contact time of 24 h, and a stirring speed of 420 rpm. The maximum adsorption capacity for polyphenols reached 69.47%, while the maximum COD removal achieved 43%, contrary to another study on olive mill wastewater decontamination using *Corbicula* shells[96,98]. Kinetic studies showed that the pseudo-second-order model provided a better fit for the adsorption behavior than the intraparticle diffusion and pseudo-first-order models[98]. This implies that the types of shells used could have opposing or different effects on the same kind of wastewater. Chitin could be the main material in shells that confers its adsorbent characteristics[11]. Generally, when shells are pulverized to a smaller particle size, the surface area increases, and more chitin is exposed. The findings from an adsorption study revealed that finer waste oyster shell powder (average 8.15 μm) adsorbed Congo red and methylene blue to a substantially greater extent, with an increase of 48.3% and 62.5%, respectively, compared to the ordinary shell powder (average 27.75 μm). Notably, the adsorbent can remove more Congo red than methylene blue, because of the presence of cationic chitin[99].

There has been increasing interest in using *Eichhornia crassipes*, an aquatic weed, to remove synthetic dyes due to its wide availability. Saufi *et al.* explored the potential of the dried leaves and roots of *E. crassipes* in removing Rhodamine B[100]. Their findings indicated that the leaves had a greater capacity for adsorbing Rhodamine B compared to the roots. Additionally, they observed that increasing the initial dye concentrations and the duration of contact between the biosorbent and dye solution led to enhanced removal of the dye by both the roots and leaves. The adsorption data were effectively represented by the pseudo-second-order kinetic model and the Langmuir isotherm model[100]. In a study by Kulkarni *et al.*, the biosorption capacity of dried roots of *E. crassipes* for crystal violet dye was examined[101]. It was reported that higher temperatures intensify the bond between the dye molecules and the bonding sites within the roots of *E. crassipes* (when sprayed). The adsorption behavior was tested with the Langmuir and Freundlich isotherm models. The authors concluded that the Freundlich model fitted the biosorption equilibrium data better. Additionally, they found that the pseudo-second-order model offered the best representation of the biosorption kinetic data[101]. Patil *et al.* studied the use of live roots of *E. crassipes* to remove crystal violet from water[102]. They found that the Freundlich isotherm model best described the biosorption process, indicating multilayer biosorption on an energetically heterogeneous surface. They also observed that the pseudo-second-order model was most suitable for describing the adsorption kinetics[102].

The pseudo-second-order model seems to primarily govern the kinetics of organic pollutant biosorption using various non-agricultural biosorbents. While the Langmuir isotherm predominantly describes the adsorption behavior, other isotherms, such as Freundlich and Sips, are also frequently mentioned. Other than the variability caused by different biosorbents and pollutants, this review highlights the possibility of magnetizing the biosorbents to facilitate their separation, and immobilizing the microbial consortia in certain waste such as brewery waste for biosorption. In addition, some biosorbents may not be suitable for certain types of wastewater and may worsen contamination. This prompts the careful selection of biosorbents according to the purpose for optimal performance.

Adsorption of inorganic pollutants

A study revealed that mussel shells can effectively remove ammonium from water. Optimal adsorption was observed to occur over 90 min using 700 mg/25 mL of ground mussel shells, with an initial ammonium concentration of 40 mg/L and a pH level of 8[103]. The maximum adsorption capacity was 2.33 mg/g, with an efficiency of 46.6%. The adsorption process was well described by the Freundlich isotherm model and both pseudo-first-order and pseudo-second-order kinetics[103]. Esmaeili *et al.* (2020) successfully employed the calcined shell of the *Solamen vaillanti* snail for the extraction of Cu^{2+}, Co^{2+}, and Pb^{2+} from a solution[104]. The optimal conditions for the removal process were observed at a pH of 5 for Co^{2+} and Pb^{2+} and a pH of 4 for Cu^{2+}. Optimal adsorption occurred at a temperature of 25 °C, with an initial metal ion concentration of 10 mg/L and an adsorbent dose of 2 g/L, over a contact time of 60 min. Moreover, the Langmuir model indicated the highest adsorption capacities for Pb^{2+}, Co^{2+}, and Cu^{2+} to be 26.04, 29.41, and 33.55 mg/g, respectively[104]. Similar to the extraction of lignocellulosic materials from crop residues as biosorbents, specific shell components can also be extracted for the synthesis biosorbents. Ali *et al.* conducted a study where they used chitosan nanoparticles derived from shrimp shells for the adsorption of Fe^{2+} and Mn^{2+} from wastewater[105]. Through batch experiments, it was found that the maximum adsorption capacity for Fe^{2+} was 99.8% at 116.2 mg/g, and for Mn^{2+} was 95.3% at 74.1 mg/g. The pH levels of 4 and 5 were optimal for Fe^{2+} and Mn^{2+} adsorption, respectively, where the initial ion concentration was 40 mL/g. Optimal adsorption occurred over a contact time of 100 min with an adsorbent dose of 10 g/L[105]. As with other studies[103,104], the adsorption was best described by the pseudo-second-order and the Langmuir models.

Foroutan *et al.* explored the process of extracting Pb^{2+} from an aqueous solution using Vanami shrimp peel[106]. They achieved an impressive removal efficiency of 98% by optimizing various factors, including maintaining a pH of 6, a temperature of 30 °C, a contact time of 130 min, employing an adsorbent dose of 5 g/L, and starting with a Pb^{2+} concentration of 60 mg/L. Utilizing the Langmuir model, they were able to determine the maximum adsorption capacity, measuring at 24.331 mg/g[106]. In a study using the Taguchi method, oyster shell powder was used to remove Cd^{2+} from water[107]. Optimal removal was recorded when the biosorbent was calcined at 900 °C, and the reaction pH was 10. Biosorbent calcination at 900 °C or higher temperatures led to a larger surface area. The study also found pH to contribute most significantly to the adsorption performance of the biosorbent, followed by temperature, pollutant concentration, biosorbent dosage, and contact time[107]. This study also highlights the potential of thermochemical methods, in this case calcination, in synthesizing or improving the performance of biosorbents.

Other than organic pollutants, fish scales also show potential for removing inorganic pollutants. A study focused on using waste fish scales as a biosorbent to remove Cr^{3+} metal ions through batch experiments[108]. The highest removal of Cr^{3+}, at 99.8%, occurred using 0.8 g of the biosorbent, a solution pH of 5, and a contact time of 90 min, with an initial Cr^{3+} concentration of 150 mg/L. The Langmuir isotherm model best described the experimental data, with a maximum adsorption capacity of 18.3 mg/g. Additionally, the adsorption kinetics were observed to follow the pseudo-first and second models well[108]. Conversion of fish scales to biochar through a thermochemical approach has been conducted. Devasena *et al.* focused on using

waste fish scales to create biosorbents for treating pollutants in fish processing wastewater[91]. The findings showed that fish scale biochar effectively reduced BOD by 87% and COD by 84% within 150 min. The ideal pH range was identified as 6-8, and the experimental results aligned well with the Langmuir isotherm and pseudo-second-order kinetic models[91]. Fish scale biochar was also used in another study aiming to assess its effectiveness in removing toxic Cd^{2+} from contaminated water. Key design parameters and biosorption performance were investigated through batch and fixed-bed experiments. Batch experiments revealed that the maximum adsorption capacity of the biosorbent was 99.14 mg/g, and it was monolayer (Langmuir) in nature. The removal efficiency was 95.4% under a batch mode and 99% under a dynamic flow mode. The biosorbent retained a biosorption capacity of more than 78 mg/g and a recovery rate of 99% after 10 sorption/desorption cycles. Cd^{2+} was found to be immobilized by the –NH, –OH, and carboxyl groups[109].

In a study, food waste was combined with iron to yield Fe-loaded food waste biochar, which was then examined for its capacity to adsorb phosphate from water[110]. The study involved adjusting the temperature, pyrolysis time, and iron concentrations. The findings demonstrated that the biochar effectively captured phosphate, with the adsorption process explained by pseudo-second-order and Elovich kinetic models. The Freundlich and Redlich–Peterson isotherm models provided a more precise representation of the phosphate adsorption onto the biochar compared to the Langmuir isotherm model[110]. In another study, a similar iron-infused food waste biochar was employed to adsorb Se^{6+} from water[111]. The study revealed that the duration of pyrolysis did not significantly affect the Se^{6+} adsorption capacity of the biosorbent, but the temperature and Fe concentration were influential factors. Pyrolysis over 3.5 h, at 495.0 °C, with 0.44 M Fe added, resulted in optimal adsorption[111]. Se^{6+} adsorption best fitted the pseudo-second-order and pseudo-first-order kinetic models. Additionally, the Freundlich model was a better fit for the equilibrium adsorption data compared to the Langmuir model. The biosorbent can adsorb Se^{6+} at a capacity as high as 11.7 mg/g[111].

Though not as prevalent as other organic waste, dead or loose seaweeds floating in water or accumulating on beaches can constitute a portion of the waste. The potential of seaweeds as biosorbents has been explored. In a study, the ability of *Sargassum tenerrimum* to remove Cr^{6+} was examined through batch experiments[112]. Biosorption was observed to be maximum at 25 °C and pH 2, with a biosorbent particle size of 0.6 mm. The Langmuir isotherm model satisfactorily described the adsorption behavior and gave a maximum adsorption capacity of 37.7 mg/g dry biomass. Employing the biosorbent for batch treatment of real tannery wastewater under optimal conditions revealed a Cr^{6+} removal efficiency of around 88%[112]. Al-Homaidan *et al.* (2018) investigated the use of three types of green algae, namely *Enteromorpha intestinalis*, *Microspora amoena*, and *Cladophora glomerata* as biosorbents to remove Cr^{6+} from the water via batch experiments[113]. They found that *C. glomerata* had the highest removal efficiency of up to 66.6% at pH 2.0 and 45 °C over 60 min when a dosage of 1.0 g dried algal cells/100 mL and a starting Cr^{6+} level of 20 mg/L were used. They also determined that the Freundlich model provided a better fit for the equilibrium data[113]. Additionally, a biosorbent derived from *Jania adhaerens* was employed to adsorb Cu^{2+} from water[114]. The kinetics data were suitably described using the pseudo-second-order model. The Langmuir model best represented the adsorption isotherm and demonstrated the highest adsorption of 67 mg/g. The biosorption was facilitated by amine, carboxyl, and sulfonate functional groups [Figure 2]. A main mechanism underlying the biosorption was identified to be ion exchange[114].

Again, here, the Langmuir and the pseudo-second-order models satisfactorily describe the isotherm and kinetics of most of the studies reviewed. A study on various factors influencing biosorption found the influence of pH to be the strongest[107]. The presence of salts and surfactants in the aqueous solutions can also affect or increase biosorption efficiency, implying that modifying wastewater could be an option to improve biosorption.

DISCUSSIONS

Adsorption has gained significant attention compared to other wastewater treatment methods because of its effectiveness in removing various contaminants, cost-effectiveness, and simplicity. In particular, biosorbents have been highlighted for their potential, as they are often derived from agricultural by-products, making them more affordable and sustainable than activated carbon, another commonly used adsorbent[115]. They are biodegradable and some can be regenerated for reuse, simplifying disposal and management. However, biosorbents may suffer variable performance, influenced by the type of biosorbent used, the nature of the pollutants, and environmental conditions. Furthermore, they may have lower adsorption capacities than activated carbon[15,116]. A study comparing the adsorption capabilities of commercial adsorbents and biosorbents for fluoxetine in water found that among commercial options, granulated activated carbon (233.5 mg/g) topped the list, followed by zeolite 13× (32.11 mg/g) and zeolite 4A (21.86 mg/g). For low-cost biosorbents, the order was spent coffee grounds (14.31 mg/g), then pine bark (6.53 mg/g), and cork waste (4.74 mg/g)[117]. This indicates the superior adsorption capacities of commercial adsorbents. However, a cost analysis showed that commercial adsorbents had a higher cost per gram of fluoxetine removed, with zeolite 4A costing 6.85 €/g, zeolite 13× costing 3.13 €/g, and granulated activated carbon costing 1.07 €/g. In comparison, low-cost biosorbents offered lower costs: pine bark was 0.92 €/g, cork waste was 0.41 €/g, and spent coffee grounds were the least expensive at 0.16 €/g[117]. Biosorbents were generally more cost-effective for the same amount of contaminant removed.

The variability of biosorbents is also demonstrated in this review, where the adsorption capacities of different biosorbents vary with different pollutants, as shown in Table 3. Avocado peel stands out as an effective biosorbent for dyes, while H_3PO_4-activated rice husk could remove all ammonium from water. Rice husk also eliminated Cu^{2+} in an experiment. Hazelnut shells and banana peels perform excellently as adsorbents of multiple heavy metals. Chitosan nanoparticles, Vannamei shrimp peels, fish scales, and fish scale biochar are also good adsorbents of metal ions [Table 3]. Chitosan is derived from the exoskeletons of crustaceans, confirming the potential of shrimp peels as a biosorbent. Additionally, the abundance of these waste types makes them good candidates for biosorbent production. For instance, approximately 8.4 million tonnes of avocados were produced worldwide in 2021, and with the peel accounting for about 10%-15% of an avocado's total weight, it is estimated that 840,000 to 1.26 million tonnes of avocado peel waste is produced annually. The annual generation of risk husk is estimated at 100 million tonnes, while that of banana peel is approximated at 36 to 48 million tonnes[118]. Valorizing these waste materials as low-cost biosorbents could substantially reduce the volume of organic waste bound for landfills. Turning these waste materials into biosorbents substantially extends their lives, and this process typically requires only drying to reduce the moisture content without the need for extensive and energy-consuming preservation procedures, such as freezing.

In some instances, the carbonization of the biosorbent materials increases their performance. A study comparing the effectiveness of *Euryale ferox* Salisbury seed coat and its activated carbon in removing Pb^{2+} ions revealed that the activated carbon achieved an impressive removal efficiency of 99.9% for Pb^{2+}, while the seed coat itself only managed to remove 89.5% of the ions at neutral pH[119]. Another study reported that biochar was notably more efficient than woodchip in reducing COD, total organic carbon, and fecal indicator bacteria. The reasons for this could be its higher porosity, larger surface area, and greater chemical stability after carbonization[120]. Additionally, biochar can be customized through additional treatment to target specific pollutants. Different feedstocks can be mixed to produce biochar, further alleviating the concern for raw material supply for biosorbent production[121]. Mixing feedstocks may enhance functional group diversity, which can improve the adsorption of a wider range of contaminants from wastewater.

Table 3. Best-performing biosorbent with more than 90% removal efficiency based on pollutant type

Pollutant	Biosorbent	Removal (%)
Rhodamine-B	Sulfuric acid-activated coconut husk	99.2%
NH_4^+	H_3PO_4-activated rice husk	100%
Crystal violet	Citric acid-modified *Camellia oleifera* shell	97%
Naphthalene Fluorene Phenanthrene	Banana peels	95% 98% 98%
Alcian blue Brilliant blue Neutral red Methylene blue	Avocado peels	99.6% 99.5% 99.8% 99.7%
Norfloxacin	Barley straw with H_3PO_4 and microwave treatments	95.5%
Cu^{2+}	Rice husks	100%
Cd Pb Hg	Hazelnut shells	98% 97% 90%
Cr^{2+} Cr^{3+} Pb^{2+} Th	Banana peels	97.8% 98.9% 96.9% 100%
As^{3+}	Food waste treated with NaOH	99.8%
Fe^{2+} Mn^{2+}	Chitosan nanoparticles	99.8% 95.3%
Pb^{2+}	Vanami shrimp peels	98%
Cr^{3+}	Fish scales	99.8%
Cd^{2+}	Fish scale biochar	99%

Carbonization to produce biochar incurs additional costs. An analysis of life cycle environmental and economic performance performed by Alhashimi and Aktas revealed that biochar presents a more favorable energy demand and a reduced global warming potential compared to activated carbon. Specifically, biochar has an energy requirement of 6.1 MJ/kg, while activated carbon's demand is significantly higher at 97 MJ/kg[122]. Regarding greenhouse gas emissions, biochar generates an average of -0.9 kg CO_2eq/kg, in stark contrast to activated carbon's emissions of 6.6 kg CO_2eq/kg. When assessing heavy metal adsorption as the primary function, biochar typically exhibits lower environmental impacts, displaying a significant difference of about one order of magnitude between the two materials. This conclusion holds even after accounting for the environmental effects associated with transporting biochar over long distances[122]. Additionally, the cost associated with using biochar to adsorb chromium and zinc is lower than that for activated carbon. For lead and copper, however, the adsorption costs were found to be similar for both materials[122].

In actual wastewater treatment, biosorbents, including biochar, can be applied in column biosorption in tertiary treatment, where wastewater is passed through a column packed with biosorbents. As the water flows through, pollutants are adsorbed onto the materials. This system is suitable for continuously treating large volumes of wastewater in industrial or municipal settings[123]. Biosorbents can be used in stabilization ponds or filtration systems to improve the removal of suspended solids and pollutants[124]. In some cases, more biochar is needed than activated carbon to treat the same amount of wastewater, primarily due to differences in their adsorption capacities and characteristics, thus resulting in more waste biosorbent generated. This waste could be reduced by enhancing the reusability of the biosorbents or channeling it for anaerobic digestion to recover energy while reducing waste volume[15]. Its uses for composting and soil amendment are controversial due to the presence of contaminants and the reentry of these contaminants into the environment. For the same reason, the use of activated sludge has been controversial. It is a

regulated waste in countries such as China, Germany, and the Netherlands[125]. Thompson *et al.* also stated that biochar from biosolids performed worse environmentally than powdered activated carbon and wood biochar because of the energy requirement for drying biosolids and the additional adsorbent needed[126]. Furthermore, due to its inorganic content, it has not been included in this review. It is noteworthy that the matrix effect of wastewater could limit the effectiveness of biosorbent, as discussed in Section "Implications".

CONCLUSION

A large inventory of organic waste has shown the potential of being converted into biosorbents. In fact, most studies show that organic waste, particularly agricultural waste, can be used for biosorption, and the efficiency can be improved with optimization of pH, temperature, biosorbent dosage, and contact time according to the physicochemical properties of the wastewater treated. Modifications, such as pulverization, chemical treatments, pyrolysis, calcination, immobilization, and composite production, can also be made to the biosorbents, whether individually or in combination. Despite their versatility and flexibility, certain barriers to their wide application must be addressed. Their major limitations are (1) biosorbents may experience decreased efficiency after several regeneration cycles, and their regeneration could be complex and costly; (2) biosorbents may lack specificity in their actions and may be suitable when it comes to removing a specific contaminant from the wastewater; (3) used biosorbents contribute to secondary waste that needs to be treated or disposed of carefully; (4) the performance of biosorbents vary considerably for different types of pollutants under different operational conditions; and (5) utilization of biosorbents may require specialized technology and equipment.

To overcome these barriers, it is recommended that (1) the efficiency and cost-effectiveness of regeneration techniques, often including chemical, thermal, or biological approaches, can be enhanced to improve the recyclability of biosorbents; (2) more studies can be dedicated to functionalization and modification of biosorbents to increase their performance in removing certain pollutants; (3) the biosorption process can be optimized to reduce chemical and energy requirements, hence operational costs; (4) the optimization procedures are standardized by addressing the major factors influencing biosorption, and this can be facilitated with artificial intelligence; (5) user-friendly and scalable biosorption systems can be developed, for instance, by designing biosorption modules that can be added to the existing wastewater treatment facilities. This includes the development of column technologies to pack the biosorbents. This review highlights the potential use of biosorbents, especially biochar, as a substitute for high-cost, energy-intensive activated carbon in the column adsorption process of tertiary wastewater treatment or filtration systems of small-scale water treatment. It also sheds light on the possibility of combining different feedstocks for biochar production to ensure an adequate supply of raw materials. It contributes to the circular economy by turning waste into resources. In relation to the recommendations, future studies could focus on improving the regeneration potential of biosorbents, the synthesis of biosorbents with better performance, and their applications in pilot- or full-scale wastewater treatment, as well as the use of artificial intelligence for optimization of the treatment process.

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Authors' contributions

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