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Review

Lignocellulose-Based Biosorbents for the Removal of Contaminants of Emerging Concern (CECs) from Water: A Review

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Abstract: Contaminants of emerging concern (CECs) are chemicals or materials that are not under current regulation but there are increasing concerns about their possible occurrence in the environment because of their potential threat to human and environmental health, with wastewater perceived as their primary source. Although various techniques for their removal from water have been studied, it should be emphasized that the choice should also consider the use of resources and energy within the removal processes, which must be minimized to avoid additional carbon footprints and environmental impact. In this context, the use of biomass-based sorbents might represent a cost-effective and environmentally friendly approach for the removal of CECs from water because they are based on preferably local renewable resources with lower negative impacts on the global carbon cycle through greenhouse gas emissions than the conventional nonrenewable ones. This paper provides an overview of the studies dealing with the application of such so-called biosorbents for the removal of CECs from water and discusses the use of their different forms: sorbents after a minimal pretreatment of the original lignocellulosic biomass; sorbents extracted from lignocellulosic biomass and/or modified; and biochar-based sorbents obtained after thermochemical conversion of biomass. It explains possible modifications of biosorbents and discusses the efficiency of various biosorbents for the removal of selected emerging compounds that belong to the classes of pharmaceuticals, personal care products, and pesticides and compares the adsorption capacities, kinetic models, and mechanisms reported in the relevant literature. Biochar-based sorption has been studied more often if compared to other considered biosorbents. In some cases, removal efficiencies of contaminants greater than 90% were achieved, but nonetheless a wide range of efficiencies for different CECs indicates that for successful simultaneous multicompound removal, a combination of different processes seems to be a more appropriate approach than the stand-alone use of biosorbents. Finally, this review discusses the reasons behind the limited commercial application of the considered biosorbents and provides directions for possible further research, in particular the use of spent biosorbents from a perspective of circular systems.

Keywords: biomaterials; contaminants of emerging concern (CECs); biosorption

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1. Introduction

The development of industry and the increase in population affect the quality of water resources and lead to increased water consumption. High-quality water remains

the most vital natural resource. As such, water must be treated as a resource of public interest and used rationally, diversely, and repeatedly. Various factors such as population growth, urbanization, and global climate change are having a major impact on reducing water as a resource, and it is expected that over time it will become a scarce commodity in many areas. Furthermore, the problem of increasing environmental pollution and public health issues has encouraged research in this area. Most water used in settlements or industries ends up as wastewater that is discharged (treated or untreated) into natural recipients (receiving water bodies) and then reused since often the receiving waters also serve as a source from which water is drawn for use. Thus, not only do contaminants from wastewater enter (and harm) the natural environment, but they also appear in water used as a resource, making it even more complex and expensive to treat. In this way, the quality of fresh water is increasingly connected to the quality of wastewater. Considering all the above, it is necessary to take all measures to protect water from pollution and optimize water consumption.

Contaminants of emerging concern (CECs) have been recognized as chemicals that pose a significant risk to the aquatic environment or a risk transmitted by them [1,2]. Many chemicals are grouped under the term CECs; the most important groups are pharmaceuticals, personal care products, microplastics, pesticides, per- and polyfluoroalkyl substances, and their potential transformation products. Many of them are commonly used in industry and households [3], but these pollutants are still not regulated and are not included in regular monitoring [4]. In addition, conventional wastewater treatment plants are often not efficient enough to remove them, so additional wastewater treatment is required after the conventional treatment process is complete. Although various techniques to treat them have been studied and developed, CECs still represent a serious threat to human health and the ecosystem [5], primarily due to a wide range of simultaneously present compounds that significantly vary in their physico-chemical properties and occur at low concentrations.

When evaluating a process such as the removal of CECs to solve environmental problems, economic, social, and environmental considerations must be taken into account as well as determining which of the processes studied are appropriate for a particular application. Different treatment technologies have been investigated for the removal of CECs from water and wastewater [6–12], and some showed very good efficiency. Ozonation, for example, provides more than 90% removal of examined substances such as carbamazepine, fexofenadine, tolyltriazole, cetirizine, caffeine, and suisobenzone [13]. Membrane filtration processes [10,14] and advanced oxidation processes (AOPs) [15,16] showed outstanding performances. Zhang et al. [17] reported results for the removal efficiency of carbamazepine, metoprolol, ciprofloxacin, and trimethoprim with E-UV/Cl_2 and found that after treatment, the concentrations of all the selected pollutants were below detection limits. However, alternative methods such as advanced oxidation processes and membrane-based processes might be costly and sometimes result in secondary pollution because of oxidation by-product formation. They are commonly found to involve high investment and maintenance costs [18], while sorption-based technologies are regarded as more straightforward and safer approaches than other water treatment technologies because of the minimal use of chemicals and low energy expenditure [19] without the formation of undesirable by-products [18]. Activated carbon is a widely used sorbent in commercial water treatment facilities and is also known for the efficient removal of organic matter, including micropollutants such as various classes of CECs [20]. However, commercial activated carbon is costly and sometimes linked to fossil-based (nonrenewable) carbon. Other less traditional sorbent materials include ion exchange resins, zeolites, and metal–organic frameworks [20] as well as calcium lead hexaferrite photocatalysts [21], molecularly imprinted polymers, and mesoporous organosilica (MPOS) [22], but their production costs are high and still limited to lab-scale application results [18]. Hence, in order to find low-cost and locally available solutions for water treatment, "green" carbonaceous materials obtained from different biomass sources (including biowaste) have also been investigated as innovative and cost-effective technologies for removal of CECs.

Thus, the aim of this paper is to provide an overview of the current state of the application of these so-called biosorbents and discuss the potential of biosorptive treatment of water and wastewater as a cost-effective approach in the removal of CECs.

2. Biosorption

There is no single, universally accepted definition of biosorption because it encompasses several mechanisms (e.g., adsorption, absorption, surface complexation, ion exchange, and precipitation), and multiple factors can contribute to the entire process, including the adsorbate (the substance to be sorbed), used biosorbent, environmental factors, and (in the case of living microorganisms as biosorbents) even the metabolic processes taking place. However, the prefix "bio" implies that a biological entity (e.g., lignocellulosic or microbial biomass) is involved. Therefore, the simple definition of biosorption is that it is a physicochemical process for removing substances from a solution using biological materials [23]. Michalak et al. [24] provided an even simpler definition of biosorption by stating that biosorption is a subcategory of adsorption when the adsorbent is of biological origin. Based on the method of operation, adsorption processes can be divided into batch and continuous (Figure 1). The batch method of performing adsorption implies a closed system that contains a defined amount of adsorbent in contact with the adsorbate solution. In contrast, dynamic adsorption is a process in an open system in which the adsorbate solution continuously passes through a column in which the adsorbent is placed. Batch systems are predominantly used at the level of laboratory tests, which have the purposes of defining physical and chemical characteristics of the adsorption process as well as gaining insight into adsorption mechanisms, while continuous systems enable the evaluation of adsorption for applications with larger capacities for water treatment.

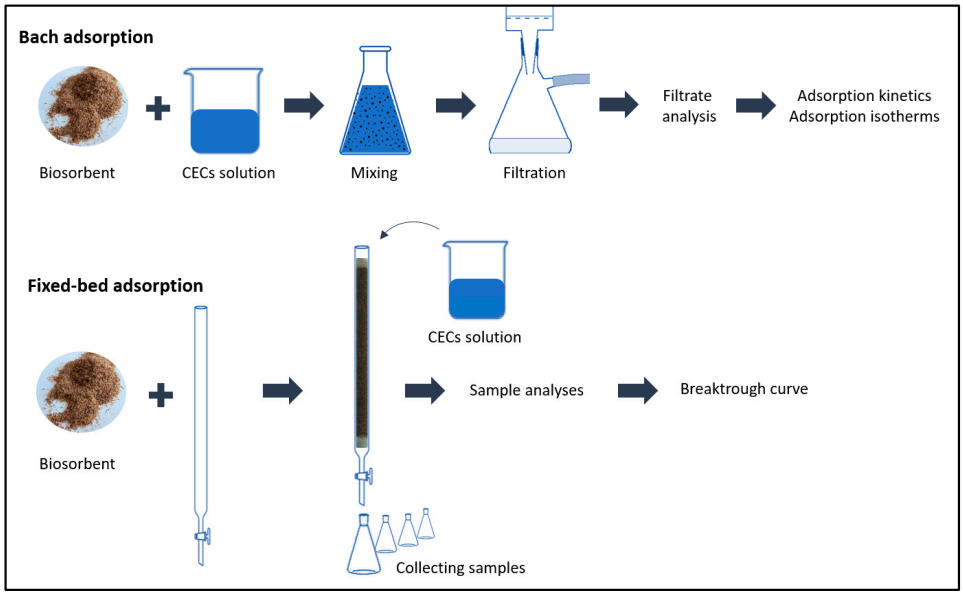

Figure 1. Schematic representation of batch and continuous adsorption methods.

The idea of using biomass-based materials as adsorbents for wastewater treatment arose from the need to find adsorbents that were cheaper than activated carbon. Despite the high efficiency and ease of use of activated carbon, its price is often high and there are also additional costs for regeneration. An adsorbent is considered inexpensive if it requires little or no processing, occurs naturally in large quantities, or is available in large quantities throughout the year [25,26]; e.g., as a production residue (by-product or industrial waste).

Biomass is a broad term that includes various types of organic materials such as microbial biomass (bacteria, fungi, and algae) and lignocellulosic biomass/waste (from forestry, the wood industry, the agri-food industry, horticulture, households, etc.), but also materials of biological origin that do not fall under the latter two categories (e.g., bones,

feathers, eggshells, and wool). Almost all biological materials have some affinity for metal ions, various organic matter, or even emerging contaminants, so the types of biomass that can potentially be used for biosorption are numerous.

Okoro et al. [27] presented the recent trends in the use of biosorbents for the removal of organic contaminants from wastewater. Adsorption with biomass-based adsorbents has proven to be a useful method due to its efficiency, simplicity, ease of application, and cost-effectiveness. Furthermore, it avoids the use of toxic solvents and minimizes degradation [28]. The main advantage of the adsorption process for the treatment of water containing emerging contaminants is that it does not produce by-products, which can be more hazardous than the original compounds [29]. In addition, researchers around the world are focusing on environmentally friendly (green) and sustainable materials that are inexpensive, do not produce hazardous by-products when used, and maximize the efficiency of pollutant uptake from water [30]. Various types of biomass that have been used as adsorbents to date fit this description.

When testing an adsorbent, the following considerations should be made:

- Evaluating the potential for using a particular adsorbent for the adsorption of a specific adsorbate;
- Determining the optimum operating conditions under which the use of an adsorbent will be the most effective;
- Insights into the adsorption mechanism, the understanding of which will enable a more efficient application of a particular adsorbent.

3. Lignocellulose-Based Biosorbents: Types and Preparation Methods

Lignocellulosic materials have been extensively studied as low-cost adsorbents in recent years and have been shown to be efficient in removing structurally diverse pollutants. In general, lignocellulosic biomass originates from cultivated, residual, or waste materials from agriculture, forestry, and industries based on these two sectors, as well as from urban zones. A recent study [31] showed how agricultural wastes, for example, can be used as biosorbents and have the potential to help solve pollution problems. Any reuse of production residues; i.e., by-products or waste materials, increases the profitability of raw material cultivation and reduces the cost of waste management, giving them a higher value as secondary raw materials. This study showed that lignocellulosic biomass could be used: (i) as an adsorbent in its original form with appropriate processing (drying and grinding) to obtain a suitable structure; (ii) as a chemically modified biomass-based adsorbent; or (iii) as a thermochemically modified biomass in the form of a carbon-based sorbent (biochar). Based on the results reviewed, the authors concluded that the use of biosorbents for purification of industrial and municipal wastewaters is an attractive solution for reducing environmental pollution by treating one waste (wastewater) with another waste (biowaste used as biosorbents).

Before the selected biomass is used for adsorption, it must be prepared or pretreated in a specific way (Figure 2). As mentioned above, if biomass is used as a raw material, it needs to be processed; i.e., chopped, ground, washed if necessary, dried, and sieved to obtain the appropriate fractions. Washing can be performed with water, an acid, or a base depending on the biomass type and composition [20]. In addition, biomass can be used to produce materials such as hydrochar and biochar [32–34] or to isolate lignin [35–37], which can also be considered an adsorbent.

The physical and chemical properties of biosorbents; i.e., the physical and chemical characteristics of their surface, have a great influence on their sorptive performance and also determine the mechanism of adsorption and their potential application [38]. The characteristics of a good adsorbent with a high adsorption capacity include a large surface area, a small volume, stability (both chemical and thermal), a high porosity, and mechanical strength [38]. Binding forces within the particles of the sorbent are also important characteristics on which the adsorption efficiency depends [39,40]. The key features for determining the adsorption properties of lignocellulosic materials include the chemical composition

(the most important), functional groups, surface area (which is often negligible), porosity, and surface morphology [41,42].

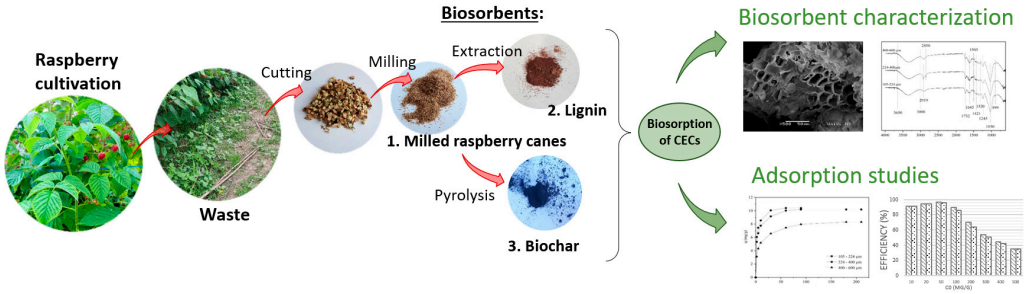

Figure 2. Schematic representation of possible biomass pretreatment processes used for biosorbent production using an example of raspberry plant waste.

The three main components of lignocellulosic biomass are cellulose, lignin, and hemicellulose. Cellulose (30–50%) is a polymer that consists of β-D-glucopyranose sugar units, the average chain of which has a high degree of polymerization (between 9000 and 10,000 units). About 65% of cellulose is crystalline and highly oriented with no access to water and other solvents, while the rest consists of less oriented chains associated with hemicellulose (20–40%) and lignin (15–25%). The chemical composition of some lignocellulosic materials is shown in Table 1.

Table 1. Average chemical composition of some typical lignocellulosic materials.

Lignocellulosic Material	Component (%)			
	Cellulose	Lignin	Hemicellulose	Reference
Rice straw	25–35	10–15	20–30	[43]
Barley straw	30–35	14–15	24–29	[43]
Sugarcane bagasse	32–44	19–24	25–35	[43]
Sugarbeet bagasse	22–30	3–4	24–32	[44]
Brewers' spent grain	20	28	28–30	[45]
Bamboo	26–43	21–31	15–26	[45]
Orange peel	12	2	15	[46]
Banana peel	12	10	26	[46]
Corncob	35–45	5–15	35–45	[43]
Hardwood	40–55	20–25	25–40	[43]
Softwood	40–50	25–35	25–35	[43]
Nut shell and stone	25–35	30–40	25–30	[43]

A very important factor in the binding of pollutants to lignocellulose is the presence of functional groups in their chemical composition such as hydroxyl, carboxyl, silanol, etc., which are responsible for binding of organic pollutants from the aquatic environment and binding forces within the particles of the sorbent [39,40]. A summary of the properties of typical lignocellulosic biosorbents is shown in Table 2.

Table 2. Summary of the properties of typical lignocellulosic biosorbents [41].

Chemical composition	Cellulose 7–73%, hemicellulose 6–33%, lignin 2–33%, ash 1–17%
Functional groups	Hydroxyl, carbonyl, silanol, alkyne, aromatic rings, keto and aldehyde groups, lactones
Surface area	Raw form: 3.14 to 25.97 m^2/g Modified form: 566 m^2/g

In recent decades, lignin has attracted considerable attention as an adsorbent for various types of inorganic and organic pollutants in water because of its unique physico-

chemical properties, biocompatibility, low cost, availability, and possession of active sites for pollutant binding. Lignin, which is the second most abundant biopolymer in nature, is obtained from pulp and paper production during the extraction of cellulose [47]. Lignin surrounds cellulose and hemicellulose and provides strength to plant cell walls. It is characterized by an amorphous structure and a high resistance to chemical and biological degradation [48]. The term lignin is used to describe a group of aromatic polymers formed by the oxidative coupling of 4-hydroxyphenylpropanoids [49]. Hydroxycinnamyl alcohols (or monolignols), coniferyl alcohol, sinapyl alcohol, and lower amounts of p-coumaryl alcohol are the major building components of lignin [50]. With its large surface area and porosity, lignin or lignin-rich biomass is a very interesting feedstock for activated carbon production [51,52]. Lignin is believed to be the main storage medium for organic pollutants; the more lignin, the higher the affinity to poorly degradable organic pollutants [41,53].

Modification of Biosorbents

As mentioned earlier, biomaterials can be used as biosorbents directly, in natural form, or in modified form [30]. The main disadvantage of using lignocellulosic waste materials as biosorbents in their natural form is often a much lower adsorption capacity for some substances compared to conventional adsorbents or the modified forms of these waste materials. In addition, the possible release of organic compounds present in natural materials leads to secondary pollution and an increase in the value of the COD/BOD of treated wastewater [54]. Modifications can improve the general adsorption properties of the natural form of biomaterials; i.e., increase the surface area [55] and the number and diversity of functional groups [56]; improve their uptake capacity [41,57,58]; and also improve particle size, pore size, pore volume and morphology, stability, etc.

Different types of modifications can be made to improve adsorbent performance; these include chemical and physical activation or incorporation with different materials. Chemical modification includes the use of chemicals such as acids, salts, or alkaline compounds, while physical modification can be conducted with steam, CO_2, or air [59] and has a smaller impact on biosorbent porosity compared to chemical activation [60]. This means that more micropores and small mesopores can be formed by chemical activators. A recent study [61] showed that chemically modified biobased activated carbons have much larger surface area than those physically activated.

In his review of chemically modified biosorbents for the removal of pharmaceuticals from water, Adewuyi [42] described some of the modifications, including extraction with water, treatment with alkalis (e.g., NaOH, K_2CO_3) or acids (e.g., H_2SO_4, H_3PO_4), impregnation with metal salts (e.g., $ZnCl_2$), surface grafting (e.g., polyethylenimine), and pyrolysis followed by treatment with organic solvents. In addition, the author confirmed that chemical modification of some biosorbents such as solid plant and animal wastes, microorganisms, and biocomposites improves their capacity to remove pharmaceuticals from water. Biocomposites have also shown great promise compared to other biosorbents because they contain two or more specific materials that are grouped together to produce a new material with better characteristics than the individual components. Most of the reported adsorption processes followed the pseudo-second-order model and can be described by Langmuir's isothermal model.

Wang et al. [52] reported different modification methods of lignin for increasing oxygen-containing groups in order to improve the adsorption capacity of lignin-based adsorbents. For example, Zhang et al. [62] compared the modification of lignin carbon materials with three different activation agents (KOH, $KHCO_3$, and K_2CO_3) and found that KOH was the best activation agent for lignin sulfonate and that the surface area of the obtained lignin-based carbon material was 2770 m^2/g. On the other hand, $KHCO_3$ was the best activator for alkali lignin. The surface area of the alkali lignin carbon material was up to 2084 m^2/g.

Coldebella et al. [63] compared the efficiency of raw and modified biosorbents for the removal of atrazine from water. The results showed that the thermally treated adsorbent

possessed a significantly higher capacity (7.47 mg/g) and surface area (70.54 m^2/g) than the unmodified material (0.63 mg/g and 3.72 m^2/g). An improvement in adsorbent properties after modification was reported by Portinho et al. [64]. The results showed that the tested adsorbent exhibited a better adsorption capacity after modification with phosphoric acid compared to the unmodified material even though the raw material had a larger surface area. This indicated that not only the surface area affects adsorbent performances. The nature and composition of active sites (functional groups) on the adsorbent surface have an important influence on adsorption efficiency. In this case, acid activation led to an increase in the number of oxygenated surface groups that favored caffeine adsorption.

Adsorption processes using biochar and activated carbon have been widely applied for the removal of many diverse wastewater contaminants [65–67]. Biochar, which is a type of coal, is a stable carbon material obtained via the pyrolysis of biomass in an oxygen-deficient or oxygen-free atmosphere [68]. The benefits of converting biomass into biochar are the elimination of carbon from the geochemical cycle and a reduction in greenhouse gas emissions [5]. Activated carbon, on the other hand, is obtained by activating (bio)char, which can be done via chemical or physical modification (depending on the desired surface properties). Physical activation is usually achieved with steam or carbon dioxide, while chemical modification is achieved with zinc chloride, phosphoric acid, potassium or sodium hydroxide, and potassium or sodium carbonate [69].

The production of biochar and activated carbon from wastes and by-products has attracted considerable attention because the availability of cheap precursors is necessary for the economic feasibility of its large-scale production [70]. Various materials can be used as raw materials for their production: not only lignocellulosic biomasses such as residues and waste from agriculture and forestry-based sectors, but also different industrial by-products and some unconventional materials such as municipal solid waste, food residues, animal bones, etc. Biochar and activated "green" carbon have been recognized as prospective materials for water remediation due to their strong adsorption capabilities [71].

4. Biosorptive Removal of CECs from Water

Most of the presented results were obtained via the batch adsorption process, while a small number of papers addressed column adsorption [72–74] because continuous systems are more complex and require a larger volume of solution [75]. The latest research on a fixed-bed adsorption column with powdered activated carbon for the removal of carbamazepine and sildenafil was reported by Delgado et al. [76]. The experiments were first conducted in laboratory conditions and then at the pilot plant. The results showed that at the laboratory scale, the removal efficiency for sildenafil was higher than 90%, while carbamazepine was marked as a limiting pollutant to reach the saturation of the column. On the other hand, the pilot plant study showed excellent performances with removal efficiencies for both examined pollutants above 90%. In the future, the focus should be on testing column systems for the biosorption of CECs in order to transfer this technology to an industrial scale.

Different biomaterials such as lignin from black liquor, tea waste, olive stones, eucalyptus bark, corn cob, bamboo chips, bagasse fly ash, olive mill waste, etc., have been investigated for the removal of CECs from water [77–83].

Okasha and Ibrahim [84] reported the results of testing the possibility of utilization of some biobased materials (almond shell, date stone, olive leaves, black tea residual, and coffee grounds) as low-cost, effective, and locally available resources for the elimination of phenol present in aqueous solutions. This study showed that the examined materials could be effectively used as adsorbents. Good results were presented by Achak et al. [85] when using banana peels as a promising material for the removal of phenolic compounds from olive mill wastewaters—the peels showed a high adsorption capacity (689 mg/g).

Two other biosorbents—rice bran ash (RBA) and biomass of brown algae (*Cystoseira indica*)—were used for the elimination of phenolic compounds (phenol (Ph), 2-chlorophenol (2-CP), and 4-chlorophenol (4-CP)) from contaminated water, and the results were com-

pared with those for commercial granular activated carbon (GAC) (Table 3) [86]. The adsorption process was very fast for all examined sorbents. The results showed that the examined biosorbents (especially RBA) were very effective in the removal of phenolic compounds from aqueous solutions compared to the commercial GAC.

A recent study [87] indicated that the leaves of the *Ziziphus* tree may be a promising adsorbent for efficient phenol removal from wastewater.

Due to its chemical composition and presence of carboxyl and hydroxyl groups, granulated cork has proved to be a good biosorbent for the removal of different CECs (ibuprofen, carbamazepine, clofibric acid, ofloxacin, phenol, 2-chlorophenol, 2-nitrophenol, 2,4-dichlorophenol, pentachlorophenol carbamazepine, naproxen, ketoprofen, diclofenac, triclosan, and methyl paraben) from water and wastewater [88–91].

Considering that pharmaceuticals and personal care products (PPCPs) are constantly entering natural waters and that their presence in the environment is of increasing concern, special attention must be paid to their treatment. An overview of PPCPs' adsorption capacities with different natural/unmodified and modified biomaterials can be found in Table 3.

Table 3. PPCPs' adsorption capacities for different natural/unmodified and modified biomaterials.

Biosorbent	Adsorbant	Adsorption Conditions	Kinetic Model	Mechanism	Isotherms	Q_{max} (mg/g)	Ref.
Grape stalk	Paracetamol	m = 3.3–33.3 g/L, C = 20 mg/L, pH = 6, T = 298 K	PFO	π-stacking interactions and hydrogen bonding	Langmuir	2.18	[92]
Macro-algae (*F.vesiculosus*)	Trimethoprim	m = 2 g/L, C = 0.1–400 mg/L, pH = 7	PSO	Electrostatic interactions	Langmuir	71.4	[93]
Wood chippings	Trimethoprim	m = 2 g/L, C = 0.1–400 mg/L, pH = 7	PSO	Electrostatic interactions	Freundlich	8.33	[93]
Date palm leaflet AC	Ciprofloxacin	m = 2 g/L, C = 50–300 mg/L, pH = 6, T = 318 K	PSO	-	Langmuir	133.3	[94]
Corn cob AC	Chlortetracycline	m = 10 g/L, C = 200 mg/L, pH = 5, T = 298.15 K	PSO	-	Freundlich	12.39	[95]
Sugarcane bagasse AC	Chlortetracycline	m = 10 g/L, C = 200 mg/L, pH = 4, T = 298.15 K	PSO	-	Freundlich	16.96	[95]
Macadamia nutshell AC	Tetracycline	m = 1 g/L, C = 250–800 mg/L, pH = 3	Elovich	-	Temkin	455.33	[96]
Alfalfa BC (*M. sativa* L.)	Tetracycline	m = 0.1 g/L, C = 10–100 mg/L, pH = 5, T = 298.15 K	Elovich	hydrogen bonding, electrostatic and surface complexation interactions	Temkin	372.31	[97]
Tea waste AC	Sulfamethazine	m = 1 g/L, C = 250–800 mg/L, pH = 3, T = 298 K	-	π– π EDA interactions	Langmuir/ Freundlich	33.81	[98]
Pomegranate wood AC	Amoxicilin	m = 0.8 g/L, C = 0–50 mg/L, T = 298 K	PSO	Electrostatic interactions	Langmuir	437	[99]
Eucaliptus wood BC	Sulfamethazine	m = 0.08 g/L, C = 0.25–20 mg/L, pH = 4–4.25, T = 298.15 K	PSO	π–π interactions	Langmuir	20.71	[100]
Eucaliptus wood BC	Chloramphenicol	m = 0.08 g/L, C = 0.25–20 mg/L, pH = 4–4.25, T = 298.15 K	PSO	π–π interactions, hydrogen bonds	Freundlich	21.35	[100]
Red pine BC (*P.massoniana*)	Sulfamethoxazole	m = 0.01–0.015 g/L, pH = 6	-	π–π EDA interaction π	Freundlich	1.9	[101]
Red pine BC (*P.massoniana*)	Sulfapyridine	m = 0.01–0.015 g/L, pH = 6	-	π–π EDA interaction	Freundlich	1.5	[101]

Note: m—dosage of biosorbent; C-concentration of adsorbate; Q_{max}—adsorption capacity; PFO—pseudo-first-order model; PSO—pseudo-second-order model.

The latest research concerning the removal of pharmaceuticals from a water solution was reported by Malesic-Eleftheriadou et al. [102]. The authors examined the possibility of the use of activated carbon biosorbents synthesized from orange peels for the removal of a mixture of five pharmaceuticals (diclofenac (DCF), ibuprofen (IBF), ketoprofen (KPF), salicylic acid (SAL), and paracetamol (PAR)) from water. Three different biosorbents were

synthesized from orange peel AC with different activation agents via pyrolysis at 450 and 650 °C: ORPs-H_3PO_4(450), ORPs-H_3PO_4(650), and ORPs-KOH (450). The results showed that ORPs-H_3PO_4(650) had the best removal efficiencies of 99% (DCF), 94% (KPF), 91% (SAL), 79% (IBF), and 76% (PAR).

Paunovic et al. [103] investigated the adsorption of a pharmaceutical compound (naproxen) onto biochar prepared from waste plum kernels activated with microwave treatment. The tested material showed very good properties and a high adsorption capacity for the removal of pharmaceuticals from wastewater.

Ibuprofen is one of the most widely available and sold anti-inflammatory drugs, and its amount in water bodies ranges from 10 to 169 µg/L. Bello et al. [104] investigated the ability of bean husk as an adsorbent to remove ibuprofen from an aqueous solution. The results showed that bean husk was an efficient adsorbent with a maximum monolayer adsorption capacity of 50 mg/g at 50 °C. Similar results were presented in [105] when using bamboo waste as an adsorbent.

Zhou et al. [58] showed the results of ibuprofen removal on activated carbon (AC) obtained from various agricultural wastes (Table 4).

Table 4. Process parameters of ibuprofen removal for different biochars.

Adsorbent	Process Variables					Q_{max} (mg/g)
	m (g/L)	C (mg/L)	t (min)	pH	T(K)	
Cork AC	0.67	20–120	240	2	303	378.10
Siris seed pod AC	0.67	20–120	240	2–4	303	378.10
Olive waste cake AC	0.33–1.43	10.04	1560	4.1	298	12.60

Another important study of ibuprofen removal was reported by Finn et al. [62] with activated carbons obtained from different materials (bitumen, lignit, coconut, and wood-based biomass). What is interesting about the obtained results is that despite significant differences in the surface areas and pH values of the contact solutions, the removal percentages of most of the tested biosorbents were similar (~55%). The best removal efficiency for ibuprofen (75%) was obtained with a coconut-based activated carbon (AC-Coco-2).

Kozyatnyk et al. [106] reported a study in which biochar from wheat straw, softwood, and peach stone was tested for the removal of six CECs (caffeine, chloramphenicol, carbamazepine, bisphenol A, diclofenac, and triclosan). For all tested contaminants, the efficiency of peach stone biochar was 2 to 10 times lower than that of wheat straw and softwood biochar. The highest efficiency was reported for triclosan and bisphenol A using softwood biochar.

The use of pesticides in agriculture, crop protection, and animal health have resulted in environmental pollution that poses serious risks to the ecosystem and human health whether through direct exposure or through food and drinking water. Alarming amounts of pesticides (some of which are persistent, toxic, mutagenic, and carcinogenic) have been detected in water, air, soil, and food and biological materials. The contamination of water systems with pesticides has been of great concern in recent years [73]. In a study reported by Rojas et al. [107], the focus was on the adsorption behavior of four pesticides (atrazine, alachlor, endosulfan sulfate, and trifluralin) on different biomaterials. The results showed that the maximum removal efficiency (73.9%) for atrazine, alachlor, and endosulfan sulfate was achieved by using rice husk as an adsorbent. At the same time, a composite of sewage sludge and sunflower seed hulls was better for the removal of trifluralin. Table 5 shows the adsorption capacities of the different biomaterials for the removal of organic pesticides and herbicides from water.

Table 5. Organic pesticide and herbicide adsorption capacities for raw and modified biosorbents.

Adsorbate	Adsorbent	Q_{max}(mg/g)	Kinetic Model	Isotherm	Reference
Oxamyl	AC from apricot stone	147.05	PSO	Langmuir	[108]
Dieldrin	Olive stone (acid treated)	23.74	PSO	Langmuir	[109]
Endrin	Olive stone (acid treated)	43.71	PSO	Langmuir	[109]
2,4-Dichlorophenoxy propanoic acid (2,4-DP)	Apple shell	40.08	-	Langmuir	[110]
4-Chloro-2-methyl phenoxy acetic acid	Coffee wastes	340	PSO	Langmuir	[111]
Methyl parathion	Rice bran Rice husk	113.59 ± 2.62 101.94 ± 2.33	Lagergren Morris-Weber	Freundlich	[112]
2,4-Dichlorophenoxyacetic acid	*Merremia vitifolia*	66.93	PFO, PSO	Langmuir	[113]
Isoproturon	Lignocellulosic substrate	61.8	PSO	Freundlich	[74]
Propazine	Vegetable AC Mineral AC Coconut AC	25.62 25.05 27.15	-	Frumkin	[114]

The most recent research [115] reported results of removal efficiencies of selected biosorbents (powdered dead roots from the invasive species *E. crassipes*, *P. stratiotes*, and *F. japonica*; coffee grounds; and green tea grounds) for the removal of oxybenzone, octocrylene, lindane, diuron, chlordecone, and ametrin. Biosorbents obtained from the root powders showed very good efficiencies for the removal of chlordecone, oxybenzone, octocrylene, and diuron. Spent coffee grounds and green tea grounds showed excellent properties for the removal of chlordecone, oxybenzone, octocrylene, and lindane. The percentage of removal for all mentioned pollutants was between 89 and 90%, and there was a complete removal of chlordecone. The removal efficiency of ametrine was lower and did not exceed 50%.

Increasingly strict laws that regulate the disposal and utilization of sewage sludge as well as the increase in its generation and lack of proper disposal routes has prompted an urge for alternative uses of sewage sludge. One particularly promising option is the conversion of sewage sludge into adsorbents [116], which has the potential to add value to the sludge. Recently, biochar derived from sewage sludge (SSBC) was identified as a promising alternative to AC in terms of the circular economy [117]. Krasucka et al. [118] evaluated the adsorption efficiency and mechanism of two drugs (lopinavir (LOP) and ritonavir (RIT)) for three different sewage sludges derived from municipal wastewater treatment plants. The results showed that RIT and LOP could be adsorbed by sewage sludges, and the obtained values for Kd (the adsorption distribution coefficient, which is defined as the ratio of adsorption capacity and equilibrium concentration of adsorbate) were quite high (2076–3449 L/kg), indicating the high affinity of the tested sludges for the selected antiviral drugs. Even in a solution containing both drugs, the Kd values and sorption efficiency were high (close to 70%).

Paper mill sludge can also be used as an adsorbent for the removal of emerging contaminants from water. Jaria et al. [29] reported the results of fluoxetine removal from water by three adsorbents obtained from paper mill sludge. It was found that the characteristics and adsorption capacities of the tested adsorbents differed noticeably depending on the process used for their preparation. For example, results for the adsorption of fluoxetine-HCl onto three carbons named PS800-10KOH, PS800-10NaOH, and PS800-10$ZnCl_2$ showed that the maximum adsorption capacity was 191.6 ± 4.8, 136.6 ± 9.6, and 28.4 ± 0.3 mg/g, respectively.

Although significant progress in the understanding of binding mechanisms and a rapid increase in scientific papers dealing with the modeling and optimization of biosorption processes have been seen, the commercial application of this technique has been limited. One of the main reasons for this is that most research has focused on improving the performance and increasing the pollutants' removal rates with biosorbents, while thorough

technical, economic, and environmental assessment of biosorption techniques is missing; i.e., there is a lack of studies on biosorbent application in pollutant removal coupled with techno-economic and life cycle analyses. Moreover, biosorption removal studies for CECs have mainly considered a limited number of preselected contaminants in model solutions based on lab-quality water, while the real water samples generally contain numerous CECs of varying properties representing a complex and heterogeneous matrix that requires advanced analytical approaches based on expensive instruments (ultra-high-performance liquid chromatography coupled to high-resolution mass spectrometry) and highly experienced analysts for the wide-range detection of CECs [119]. Besides this analytical challenge in studying the biosorptive removal of CECs occurring in real water samples, dissolved salts and organic matter as well as solid particles present in fresh or seawater inevitably affect the sorbent's surface properties and need to be further studied.

Additionally, little information is available on the regeneration and/or disposal strategies for exhausted biosorbents [120,121]. The cost-effective application of an adsorbent depends on the possibility of regenerating and reusing the spent adsorbents. It is economically viable to regenerate adsorbents on a large scale, especially if they are not from naturally abundant materials [121]. There are various regeneration techniques (e.g., thermal, chemical, and microbial, among others), and the choice depends on the nature of bonds established between adsorbate and adsorbent; nevertheless, it seems that a combination of techniques might provide a better regeneration efficiency than a single one [121]. On the other hand, for low-cost adsorbents based on biomass, regeneration may not be economically acceptable; however, like during the biosorption process, they are converted from nonhazardous into hazardous materials, and attention must be paid to treat the spent sorbents before the disposal to avoid leaching of the adsorbed pollutants into the environment. Landfilling might also lead to increasing deposits [121], so the possibility of reusing the exhausted biosorbents in other systems should be studied in this way to be in compliance with the circular models of waste management [122]. Based on literature-based examples of the possibilities for the valorization of exhausted biosorbents loaded with heavy metals [120,123–126], it may be hypothesized in a similar manner that spent biosorbents used for the removal of CECs can be incorporated into a material such as rubber, similar to some previous studies on blending biochar with carbon black as filler for styrene–butadiene rubber [127,128]. Knowing that biochar can be considered an active catalyst for the pyrolysis of biomass or tire waste, this might be another route for the spent-biochar-based sorbent utilisation [122,129]. Gasification is a process of converting carbonaceous materials at high temperatures, so spent biosorbents such as biochar might be gasified into a flammable gaseous mixture (consisting primarily of hydrogen, carbon monoxide, and methane); this plays a very important role in the chemical industry for production of hydrocarbons, hydrogen, and other chemicals [130]. Combustion might be considered as an ultimate way of using the spent biosorbents for energy recovery (heat and/or power) and a reduction in waste volume. At the high temperatures of pyrolysis/gasification/combustion, thermal degradation of both lignocellulosic- or biochar-based adsorbents and the adsorbed organics occurs simultaneously with numerous reactions such as reduction, cracking, reforming, polymerization, etc. [121]. The most recent study [131] reported that the decomposition of a wide range of per- and polyfluoroalkyl substances in diverse organic waste during industrial-scale pyrolysis led to more than 96.9% removal of these CECs, resulting in less than 3% of their emission through the gas phase. This finding confirmed that the thermochemical route might play an important role as a spent biosorbent waste-handling option. Nevertheless, any of the post-treatment options for biosorbents need to be evaluated in a way that does not negatively impact the environment.

5. Conclusions

Removal of CECs from water via sorption has the advantages over the other advanced technologies of being simpler and safer with significant concentration factors [19]. The use of biosorbents has additional benefits due to their renewable origin and low costs,

particularly if they originated from biowaste, the application of which also contributes to waste minimization. Although biosorbents have been extensively studied in recent years, there is still a vast range of materials that are locally available in large quantities that have not yet been tested, so research in this area is still required. The main challenge is to select the appropriate biomass from a large number of promising and inexpensive biomass sources, and efficiency, availability, and price are very important selection factors. According to the presented results, the most commonly investigated methods for the removal of CECs from water is adsorption onto biochar, which showed very good efficiency and adsorption properties. Modified and raw-biomass-based materials have also been intensively investigated and have proven to be useful, effective, and low-cost adsorbents.

Nevertheless, none of the biosorbents can offer complete or high enough efficiency in the removal of different classes of CECs occurring in water. This is possibly contrary to the existing popular notion that carbonaceous sorbents almost completely remove organic compounds in water, but knowing that properties of CECs vary significantly (with many very polar compounds among them), it seems that a combination of biosorbents with other removal approaches would be more efficient, particularly in the case of real water samples consisting of a wide range of CECs at low levels. Hence, further studies focused on multisorbents and hybrid processes in the real water system may result in higher overall efficiencies while also taking into account the number of CECs efficiently removed. It must be emphasized that studies with real water samples will indicate more objectively the removal efficiencies for CECs in the presence of intrinsically occurring organic compounds, solid particles, and/or dissolved salts, which all impact the sorbent's surface properties and are critical to the removal process.

However, the analysis of CECs in real water samples requires very expensive advanced analytical approaches supported by dedicated software tools for screening numerous compounds at low levels, which means that a rather limited number of such studies might be reliably conducted at conditions existing in the environment. In addition, the management of saturated biosorbents is also a challenge, and finding an economical and environmentally friendly way to deal with this—including the problem of leaching or emission of the contaminants, the spent sorbent reuse, and/or regeneration while respecting the principals of sustainability and circular models—must fall within the scope of future research.

Thus, we finally concluded that to achieve comprehensive and identifiable results in the short to medium term that lead to long-term solutions for preventing and mitigating environmental and health problems caused by CECs, it is necessary to improve the efficiency of existing treatment technologies and develop new and/or integrated processes for their removal. Therefore, extensive research is still needed to find optimal, cost-effective, environmentally friendly, and efficient technologies for removing diverse emerging contaminants from real water resources.

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