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Sorption Activity of Plant Biosorbents in Wastewater Treatment

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ABSTRACT

Wastewater plays a crucial role in deteriorating water quality and can significantly affect human health and ecosystems if discharged without proper treatment. Among available treatment methods, adsorption is often considered an effective, relatively inexpensive, and environmentally friendly purification technique, but its efficiency depends on the sorbents used. The use of low-cost biosorbents with high adsorption capacity is widely studied. These include various biomaterials such as microalgae, cyanobacteria, fungi, and plant materials. The utilization of different biosorbents derived from plant waste, such as Paulownia wood, aspen, hickory, Ziziphus bark, peach tree shavings, as well as grasses such as red fescue and reed, and Sargassum algae in natural and modified forms, is a crucial research direction. Such studies highlight the potential to address waste issues by repurposing it as biosorbents. Several studies have examined the ability of different biosorbents to treat wastewater and suggested that

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the physicochemical properties of the material's surface, such as specific surface area, pore size, and pore volume, play a decisive role in adsorption capacity. A quantitative analysis of plant-based biosorbents will significantly aid in developing water treatment systems and achieving optimal adsorption through modifications of their physicochemical properties. Furthermore, the analysis will help understand the relative importance of each physicochemical property in determining adsorption capacity, thereby contributing to the implementation of treatment methods targeting specific pollutants.

Keywords: Biosorption; Plant Sorbents; Adsorption Capacity; Wastewater

1. Introduction

The extraction and processing of minerals, as well as energy production, generate significant amounts of contaminated effluents. Since ancient times, solid industrial and agricultural wastes have been processed into fertilizers or burned, while liquid wastes have been dumped into rivers or other bodies of water. This path led to the pollution of small and large rivers and the environment in general. The process of decomposition of organic substances in water bodies is accompanied by a decrease in the concentration of oxygen dissolved in water, which is necessary for the life of aquatic fauna [1]. The need to protect the water basin has led to a ban on the discharge of wastewater into water reservoirs and the development of technologies for their purification from pollution. However, the development of treatment technologies has lagged behind and still lags behind the pace of industrial development and the increase in the amount of wastewater. The intensification of agriculture, technological progress in industry have led to disproportions in the environment, deformation of the established balance of the ecological system and deterioration of the ecological situation in all spheres of human activity. Industrial enterprises pollute the atmosphere with gaseous and solid emissions, water reservoirs – with wastewater containing a large amount of harmful to very toxic substances, from which fauna and flora suffer. These substances enter human food through plants and animals. The chemicalization of agricultural production also has a negative impact on soil, air, water and food pollution. Therefore, there are areas where the ecological situation has grown disastrous. A great danger to the environment is caused by highly concentrated waste from food industry enterprises – alcohol, sugar, biochemical plants, and livestock farms located in regions where the population uses underground water sources [2,3].

For example, bioethanol plants are in the category of highly concentrated organic pollution due to distillery stillage and the yield of raw post-alcohol stillage to 12-15 times larger than the capacity of alcohol {4]. The lack of effective and economic methods for their purification leads to the accumulation of wastewater in artificial reservoirs, pollution of the environment, soil and groundwater with harmful substances and the formation of greenhouse gases for more than 6–7 million m^3 per year.

The environmental situation in the world can be characterized as follows:

(1) **Agriculture:**

Erosion and compaction of soil, pollution with chemicals, spread of weeds, and reduction in humus content.

(2) **Bodies of Water:**

Chemical pollution, reduction of fish stocks, and alteration of aquatic fauna and flora.

(3) **Biosphere:**

Loss of certain species of animals and plants, primarily due to deforestation and pollution of reservoirs with chemicals.

(4) **Humans:**

Incidence of diseases, genetic changes, and challenges in economic activity.

(5) **Atmosphere and Climate:**

Atmospheric pollution with gases such as SO_2, NaO, and CO_2; acid rain (pH 4.5–5.7); destruction of the ozone layer due to freon and N_2O; and increased ultraviolet radiation.

Summing up the above and other negative consequences of anthropogenic activity, it should be noted the need and relevance of taking urgent measures to protect the environment. In this complex problem, scientific and technical solutions for wastewater treatment and sludge treatment are of great importance [5,6].

Since the cost and operational requirements of treatment methods can be excessively high [7], wastewater is often discharged into filtration fields (FF). However, despite the apparent limitations of these lands, FF serves as a major source of toxic elements for living organisms [8]. Thus, there is a need for an economically viable, sustainable, and integrated approach to treating industrial effluents. The biochemical method, which is based on the ability of microorganisms to consume various soluble organic and inorganic compounds contained in wastewater, has been widely used in its treatment and in the treatment of sediments. The biochemical method is carried out under aerobically and anaerobically [9]. Anaerobic processes are used in purification practice less widely than aerobic ones. Recently, interest has been growing in the methane digestion of concentrated carbon-containing substrates by anaerobic microorganisms. For wastewater from food enterprises, the use of methane digestion is quite promising, since it allows achieving a more favorable C/N ratio for subsequent aerobic treatment, obtaining energy raw materials, and maintaining a sufficient temperature for the subsequent aerobic process in winter [10]. Methane fermentation is the process of decomposition of organic substances to final products, mainly methane and carbon dioxide, because of the vital activity of the microorganism's complex in anaerobic conditions. According to the data presented in [11], anaerobic fermentation reduced the content of organic matter in the post-alcohol stillage by 9 times. Additional treatment with aerobic bacteria reduced COD by 98% of the initial value and the content of phosphorus and total nitrogen by 20 and 9 times, respectively [11].

Biosorption using biomass is an alternative to existing wastewater treatment technologies as it utilizes the ability of dead or denatured biomass to remove pollutants from aqueous solutions [12,13–16]. Removal efficiencies across various water quality parameters revealed the series connection as the most effective treatment, achieving exceptional removal efficiencies for critical parameters such as fat, oil, and grease, total suspended solids, and turbidity. The Mixture treatment demonstrated synergistic effects, surpassing individual treatments in removing contaminants such as Arsenic, Biochemical Oxygen Demand, cadmium, Chemical Oxygen Demand, fluorides, ammonia nitrogen, and surfactants.

Food industry waste can be used after appropriate wastewater treatment. Thus, water purification from cadmium (II), lead (II) and chromium (II) ions using cashew nut shells has been studied [17]. Date palm fruit seeds are biosorbents for water purification from chromium (VI) ions [18]. Rice husks after treatment with nitric acid, as well as banana and orange peels can be used as biosorbents for the removal of manganese ions from water [19,20]. Ebrahimi et al. studied the effect of chemically modified wheat straw on water purification from arsenic ions [21]. Beech sawdust and wheat straw stalks can be used as biosorbents for the removal of lead ions from aqueous solutions [22,23]. Black tea powder after treatment with steam and sodium hydroxide acts as a biosorbent with an adsorption capacity of 43 g/kg and can be used to purify water from copper (II) ions after 10 minutes of exposure [24].

The structure of dried macroalgae includes a significant number of functional groups, which explains their sorption properties [25]. Depending on the substances that make up the cell membranes of green algae, their adsorption capacity also varies. Thus, brown algae include carboxyl groups of alginates, which have high adsorption properties. While char algae or Charophyceae combine the features of algae and higher plants [26]. Their cell walls consist of polymers that have a significant affinity for cellulose, hemicellulose, lignin, due to which metal ions dissolved in water bind to the functional groups of algae through electrostatic attraction, ion exchange with "light" metal ions or complexation processes [27]. Macroalgae and other biosorbents, such as activated carbon, interact well with cations dissolved in water and are relatively ineffective in treating oxyanions or non-metal oxides, which are the most common components of wastewater [28]. However, depending on the types of wastewater pollutants, it is possible to change the structure of dried macroalgae through appropriate treatment. Biomass can be converted into carbon-rich biochar through slow pyrolysis, resulting in a product with properties similar to activated carbon [29]. Additionally, biomass and biochar can be pre-treated with an iron solution to enhance adsorption [30]. Iron coating on dried biomass or biochar provides a positive charge, promoting complex formation and natural interactions for sorption [31,32].

Natural sorbents, such as common minerals like zeolites, saponites, brucites, shungites, and oceanic iron-manganese shales, as well as clays, have been studied for wastewater treatment. Research on the ability of saponites to remove metal compounds from wastewater has shown low capacity for these components. Similarly, low adsorption capacity was found for phosphate ions, consistent with the behavior of zeolites in similar systems.

More promising results were obtained for the removal of copper and cadmium using saponites. In adsorption, saponite derivatives are highly effective materials for water and wastewater treatment, mainly used for removing heavy metals, surfactants, hydrocarbons, toxic metabolites, and dyes. Thus, saponite has promising properties for the efficient removal of organic and inorganic pollutants [33].

Experience using the natural mineral brucite (chemical formula—$Mg(OH)_2$) as an effective sorbent for removing heavy metals, organic compounds, and other contaminants from aqueous environments has led to the assumption of its potential for arsenic removal.

When powdered activated carbon has limited contact with water, the adsorbed substance may not penetrate deep into the carbon particles, making its adsorption capacity dependent on the degree of grinding. Granulated activated carbon is used for continuous water sorption treatment as it can be regenerated, reducing water purification costs.

The characteristics and types of contaminants in water, as well as the intended use of the treated water, influence the choice of purification techniques. Activated sludge and anaerobic digestion mechanisms are century-old methods that continue to be widely used [7].

There are many methods of wastewater treatment—physical, chemical, biological and combined technologies. The most common are filtration, centrifugation, sedimentation, coagulation and flotation. These methods are referred to as preliminary or primary treatment. Biological wastewater treatment, which, depending on the type of microorganisms used, is carried out under aerobic or anaerobic conditions, provides oxidation of organic compounds and is the most common method of secondary treatment. Methods such as precipitation, reverse osmosis, electrolysis and electrodialysis are examples of additional stages of water purification. Modern treatment methods include ion exchange, ultra- and nanofiltration, adsorption/biosorption and advanced biological treatment, which combines algae, bacteria and fungi [7].

Relevance. Water conservation is one of the most pressing environmental issues today. Modern industry, agriculture, forestry, and municipal enterprises use a huge amount of chemicals that are released into the environment and eventually end up in water bodies. They have a detrimental effect on water quality, disrupt the biological balance in water bodies and their self-purification processes. However, if industrial discharges make up the main portion of wastewater, the effect of organic elements in treated water used for non-potable activities requires further investigation [34]. In addition, modern anaerobic wastewater treatment technologies can help reduce natural gas consumption by replacing it with biogas and increase reliability.

Biological wastewater treatment is an environmentally friendly process as it protects ecosystems by releasing fewer pollutants, utilizing sustainable resources, enabling the recycling of unused products, and managing waste residues in a more biologically acceptable manner.

Objective. The presence of pollutants in wastewater and the increasing volume of wastewater entering water bodies require immediate research in this area to ensure safe and clean water, as well as to secure fresh water supplies. Therefore, the objective of this work is to study modern and emerging technologies for the treatment and reuse of wastewater from biotechnological production. This article is part of a research work.

2. Materials and Methods

The methodological foundation of this work is based on the analysis of existing scientific publications, as well as on materials from our own research. To experimentally determine pollutants in wastewater, the photocolorimetric method was used [35]. The activation of natural sorbents was carried out using thermal and chemical methods. The theoretical analysis of the sorption process and the analytical processing of data were conducted using a PC.

Model wastewater solutions were used to study the sorption processes of plant-based sorbents. Glass flasks were filled with 100 cm^3 of wastewater and a specific biosorbent was added. The flasks were hermetically sealed and left for 48 hours at a temperature of +20 °C with periodic stirring. The sorbent was separated from the solution, and the resulting solution was analyzed using a KFK-3 photoelectric colorimeter based on optical density [35].

The adsorption efficiency was determined as a percentage by comparing the optical density of the solution before and after sorption. The sorption capacity of biosorbents was determined using chromatography [36].

The grain stillage was analyzed using well-known methods for determining wastewater pollution indicators [37]. We used the dichromate method to determine chemical oxygen demand (COD) [38]. Five-day biochemical oxygen demand (BOD_5) was determined by measuring oxygen consumption by bacteria during the decomposition of organic matter or by bioassay [39]. Total nitrogen in the post-alcohol stillage was determined by the Kjeldahl method [40], and the ammonium molybdate spectrometric method was used to determine by phosphorus [41].

Cl^-, SO_4^{2-}, K^+, Na^+, Ca^{2+} content was determined by the Quantitative Chemical Analysis method [42]. Determination of volatile fatty acids was carried out for the distillation method [43].

Statistical analysis

The experimental data were obtained in triplicate and processed by the methods of mathematical statistics. The results are presented as means and standard deviation. Data were subjected to ANOVA and significant differences between means were revealed by post – hoc Duncan's multiple range test ($p < 0.05$), using IBM SPSS Statistics 20.0 software.

3. Results and Discussion

A significant amount of waste from the woodworking industry is underutilized and disposed of, leading to environmental issues. Therefore, recent research in the field of environmental biotechnology has focused on using wood bio-waste in various industries, primarily as adsorbents for different pollutants.

To obtain biosorbents for pollutant removal from wastewater, *Populus tremula* wood was first crushed and then soaked for several hours in distilled water. It was then filtered and dried at a temperature of 60 °C. The adsorption capacity of this biosorbent was found to be 2.7 g/kg (**Figure 1**), with a measurement error of 0.03%.

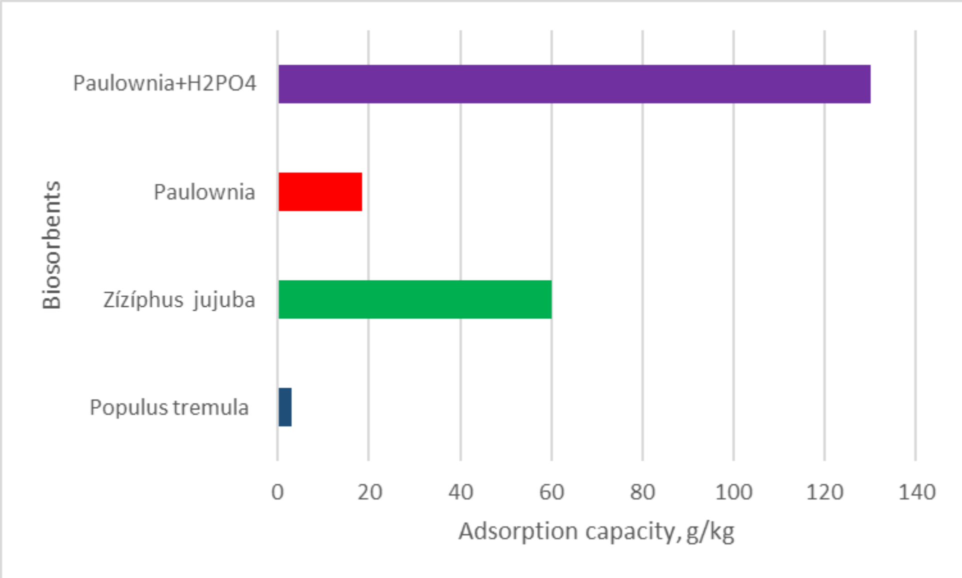

Figure 1. Adsorption capacity of wood-based biosorbents.

For the preparation of biosorbents based on *Carya* wood, a single-stage grinding process was used. Biosorbents were treated with sulfuric acid or sodium hydroxide and then ground. To remove acid or alkali residues, distilled water was passed through the biosorbent several times, then it was dried at a temperature of 80° C for 12 hours and crushed in an electric mill. Such acid or alkaline treatment of *Carya* wood contributed to an increase in the number of oxygen-containing functional groups in the cellular structure of the biosorbent, which subsequently interacted with wastewater pollutants, forming surface complexes.

The resulting acid biosorbent is able to remove up to 88% of pollutants, and the alkaline biosorbent—up to 77% (**Figure 2**), with a measurement error of 0.03%.

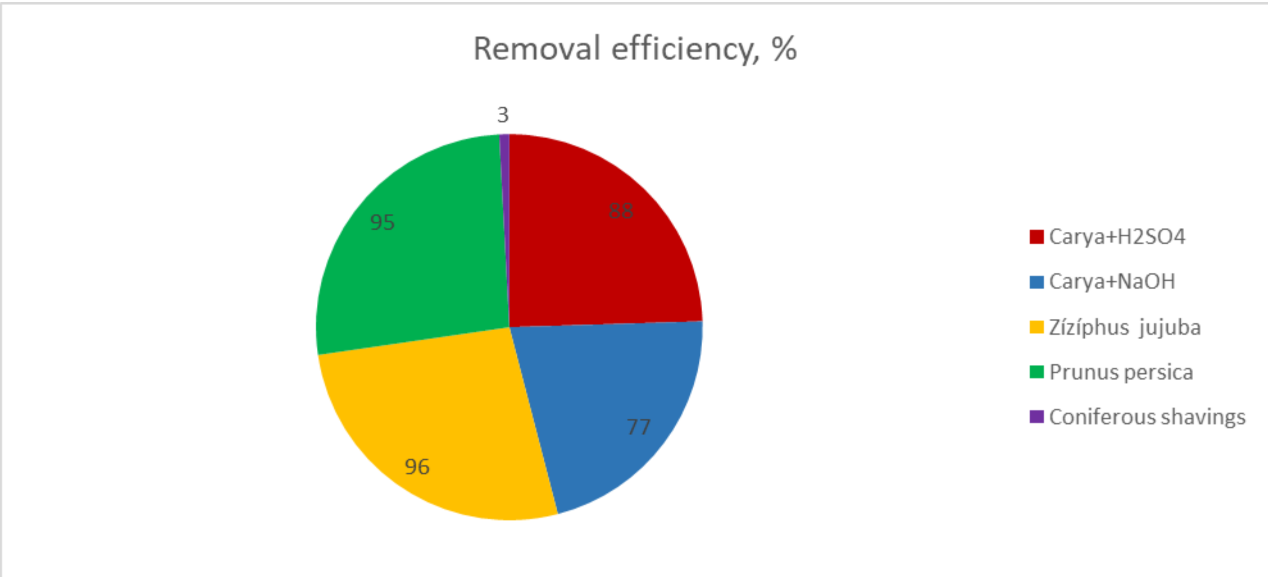

Figure 2. Pollutant removal efficiency of wood-based biosorbents.

The study on water purification from pollutants using a biosorbent obtained from *Zíziphus jujuba* bark consisted of several stages. Initially, the bark was ground and mixed with a sodium hydroxide solution, stirred, filtered, and washed with distilled water. It was then dried at the temperature of 80°C and sieved using a set of sieves.

The pollutant removal efficiency of the *Ziziphus jujuba* biosorbent reached 96% (**Figure 2**). The adsorption capacity of this biosorbent was 59.8 g/kg (**Figure 1**), with a measurement error of 0.03%. This increased adsorption capacity is explained by the formation of carboxyl functional groups on the surface of the powder and irregular cavities, which facilitates the adsorption of molecules. The sorption mechanism consists of complexation between $COOH^-$ groups and the organic pollutant and precipitation of the nonorganic pollutant in the pores of the biosorbent.

The preparation of a biosorbent from *Prunus persica* waste for pollutant removal involved crushing, mixing with ethanol, stirring, centrifugation, and freeze-drying at –60 °C. The resulting biosorbent had a porous structure with minimal impurities, providing adsorption efficiency of up to 95% (**Figure 2**), with a measurement error of 0.01%.

Cellulose, lignin, and hemicellulose remaining in the biosorbent played a crucial role in adsorption through electrostatic, ion-dipole interaction, and π-electron transfer from the biosorbent to the pollutant.

An environmentally and economically efficient method of obtaining a biosorbent for wastewater pollutant removal is the etherification of *Paulownia* wood using environmentally friendly chemicals, such as phosphoric acid and urea, which are commonly used in food production and fertilizers [8]. *Paulownia* wood was first crushed to a particle size of 0,5 mm, mixed with 30% sodium hydroxide solution and heated to 100 °C for one hour, then washed with hot distilled water to neutral pH to obtain a conventional biosorbent. The biosorbent was modified by suspending it with water and further treatment in H_3PO_4 and urea. The mixture was then dried for 12 hours at 80 °C, washed with hot water and dried in the open air.

The chemical modification process contributed to an increase in the adsorption capacity of the biosorbent by almost seven times (from 18.5 g/kg to 130.2 g/kg) (**Figure 1**), with a measurement error of 0.01%.

This is explained by a change in the crystal structural features of the sorbent, an increase in its ion exchange properties due to a change in the composition of exchangeable cations. The high adsorption capacity is also explained by the protection and stabilization of the phosphorylated biosorbent, which is achieved by urea treatment.

Unlike tree-based biosorbents, natural coniferous wood shavings had a low pollutant removal efficiency of only about 3%. This was attributed to the low release of cations from the wood biomass into the solution, resulting in a very low cation exchange rate.

Thus, among the studied sorbents, *Paulownia* waste modified with phosphoric acid and urea demonstrated the highest adsorption capacity, while *Ziziphus jujuba* exhibited the highest pollutant removal efficiency from wastewater.

Many scientific studies present research findings on the efficiency of biosorption using various plant-based raw materials. We studied such plants as *Phytolacca americana, Phragmites australis, Acacia auriculiformis and Oscillatoria princeps.* To prepare the biosorbent, the plant material was treated with a solution of sodium hydroxide, kept at a temperature of 100 ° C for one hour and washed with water. The modification of the biosorbent consisted of soaking the plant material in a dilute solution of nitric acid with shaking for six hours at room temperature. After that, the biosorbent was washed with distilled water to pH 6–7 and dried at a temperature of 60° C. Modification with nitric acid contributed to an increase in the efficiency of adsorption (**Figure 3**) by increasing the volume of pores and microcracks on the surface of the sorbent. The adsorption mechanism is due to electrostatic forces of attraction of particles of the adsorbed substance to particles of the adsorbent. Since the molecules on the surface of a solid are not balanced, due to which ion exchange occurs between the adsorbent and the adsorbate through the corresponding functional groups.

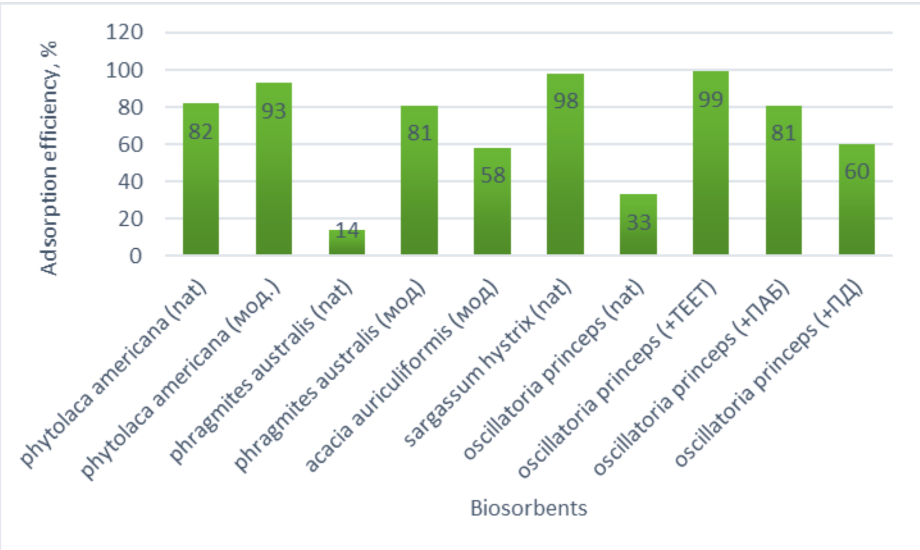

Figure 3. Adsorption efficiency of grass-based sorbents.

Phragmites australis or common reed is a perennial grass belonging to flowering plants. It is a plant that reduces the concentration of dissolved oxygen in water bodies, which in turn threatens aquatic organisms and disrupts the ecosystem. Reed is a tall plant with hollow stems and hard leaves with a high content of cellulose and lignin. The reed biosorbent was prepared by the method described above. To increase its adsorption capacity, modification was carried out by treatment with sodium hydroxide solution, washing with water to neutral pH and drying at a temperature of 90°C to a moisture content of 14%.

According to **Figure 3**, the adsorption efficiency of the *Phragmites australis* biosorbent was 14%, but after its modification it increased by 5.8 times and amounted to 81%, with a measurement error of 0.01%. This is explained by the increase in the pore volume on the surface of the modified sorbent and the change in ion exchange properties.

Another promising plant is *Acacia auriculiformis*, a flowering plant from the legume family. It is a sprawling branched tree characterized by a high growth rate. To prepare the modified biosorbent, the leaf raw material was placed in a 30% NaOH solution for 1 hour, washed with water, then kept in dilute sulfuric acid and dried at 400 °C. The resulting powder was treated with Na_2CO_3 and water to a neutral pH, mixed with ethylenediaminetetraacetic acid (EDTA) and kept at 20–24 °C for two days. EDTA operates as a chelating agent, forming chelates with metal ions. The biosorbent from *Acacia auriculiformis* after this treatment had an adsorption capacity of 58%, with a measurement error of 0.001%.

In some studies, *Sargassum hystrix*, a type of brown algae, was also used as a raw material for biosorbents. The algae biomass was prepared in the usual way by washing it with water, sun-drying, drying it in a thermostat and grinding it into powder. The study showed that the obtained removal efficiency was 98% (**Figure 3**), with a measurement error of 0.03%.

This can be explained by the presence of alginic acid in brown algae in the form of calcium, magnesium, sodium, and potassium salts, which makes up 30% of the dry weight of algae and operates as adsorption centers.

Several subsequent studies have confirmed the effectiveness of biosorbent modification. For example, the biomass of the cyanobacteria *Oscillatoria princeps* was modified by exposure to substances used in affinity chromatography to modify the surface of sorbents—tetraethylenetetramine, para-aminobenzamidine and polydopamine. These substances are characterized by strong adhesion, biocompatibility and high efficiency of photothermal conversion, due to which they have promising applications in the fields of adsorption and surface modification. The ligand with which the sorbent is modified significantly affects the retention and capacity of the adsorbent. The higher the density of this modification, the stronger the retention.

The conducted studies showed that the sorption capacity of the unmodified biosorbent from the biomass of cyanobacteria was approximately 33%, but after modification it increased significantly—for tetraethylenetetramine it was 99%, for para-aminobenzamidine—81% and for polydopamine—60%, with a measurement error of 0.007%. Different values of the sorption capacity of the biosorbent are explained by the different lengths of the hydrocarbon chain of the substance used for modification, as well as the presence of amino groups in it, which form bonds on the surface of the sorbent. For example, tetraethylenetetramine, which is longer, retains polluting components more strongly and has more adsorption centers. Thus, as can be seen from **Figure 3**, different biosorbents are effective in removing pollutants, and the biomass of cyanobacteria *Oscillatoria princeps* demonstrates the highest adsorption efficiency.

The results of determining the sorption capacity of biosorbents depending on their concentration in a model solution with pollutants are shown in **Figure 4**.

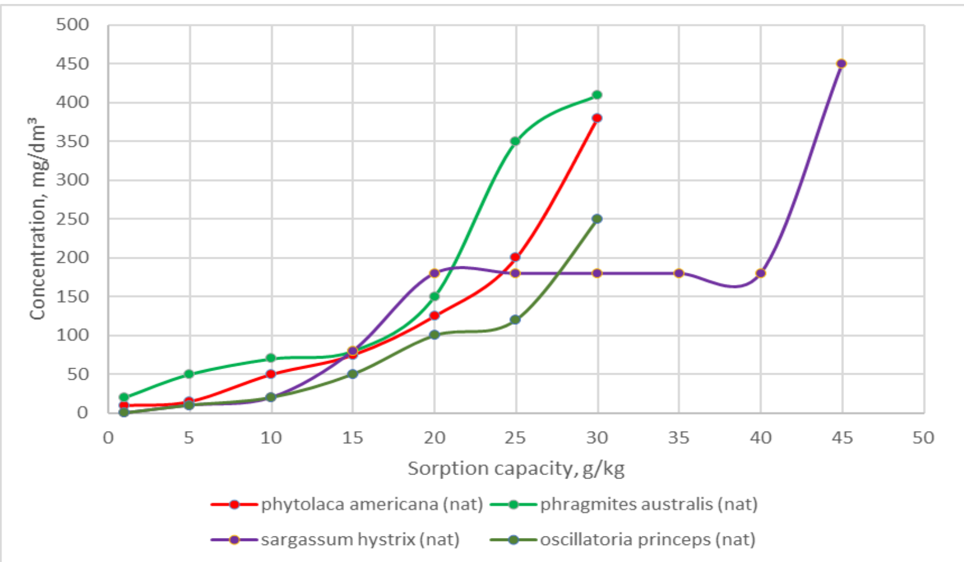

Figure 4. Dependence of sorption capacity on sorbent concentration in solution.

Clearly, for all studied plant materials, pollutant uptake increases with increasing initial concentration. The results show that the ability of plant sorbents to absorb substances depends on the pore volume of the adsorbent. Based on sorption capacity, the studied materials can be ranked in the following order: *Sargassum hystrix* (43 g/kg, Measurement error 0.03%) > *Phragmites australis* (34 g/kg, Measurement error 0.01%) > *Phytolacca americana* (30 g/kg, Measurement error 0.001%) > *Oscillatoria princeps* (27 g/kg, Measurement error 0.007%).

The efficiency of *Sargassum hystrix* biosorbents for treatment from pollutants was checked on the example of alcohol wastewater—grain stillage and methane mashes. This waste was selected because of the different content of pollutants in them. Grain stillage has high pollution. The COD is 15800 mg/l, BOD_5—9800 mg/l, the content of total phosphorus and nitrogen is 250 and 2300 mg/l, respectively. After anaerobic fermentation of the grain stillage, a methane mash is formed, which has an indicator of the COD 1360 mg/l, the BOD_5—250 mg/l, and it also reduces the content of total phosphorus, nitrogen, calcium, potassium. The increase in the sodium content in methane mash is explained by the addition of NaOH to the post-alcohol stillage to neutralize it. The results of the research are shown in **Table 1**.

Table 1. Physical and chemical content of grain stillage and methane mashes before and after biosorption treatment.

№	Parameters	Grain Stillage	After Biosorption Treatment	Methane Mashes	After Biosorption Treatment
1	pH	3.35 ± 0.01	4.5 ± 0.01	7.95 ± 0.01	7.5 ± 0.01
2	Dry residue, mg/l	24340 ± 2	2520 ± 1	2980 ± 1	160 ± 1
3	Content of volatile fatty acids, mg/l	200 ± 0.5	5 ± 0.1	720 ± 1	35 ± 0.1
4	Colloids, g/l	9,64 ± 0.01	1,1 ± 0.01	1,02 ± 0.01	0,04 ± 0.01
5	NO_2^{2-}, mg/l	0,284 ± 0.001	0,07 ± 0.001	0,009 ± 0.001	0,001 ± 0.001
6	NO_3^{3-}, mg/l	0,56 ± 0.005	0,12 ± 0.005	0,9 ± 0.005	0,18 ± 0.005
7	Total nitrogen for Kjeldahl, mg/l	2300 ± 0.01	480 ± 0.01	560 ± 0.01	112 ± 0.01
8	Total P^{3-}, mg/l	250 ± 0.01	40 ± 0.01	65 ± 0.01	11 ± 0.01
9	Cl^-, mg/l	230 ± 0.01	27 ± 0.01	190 ± 0.01	24 ± 0.01
10	SO_4^{2-}, mg/l	109 ± 0.01	16.1 ± 0.01	42 ± 0.01	5 ± 0.01
11	K^+, mg/l	760 ± 0.01	81 ± 0.01	275 ± 0.01	39 ± 0.01
12	Na^+, mg/l	35 ± 0.01	2,4 ± 0.01	681 ± 0.01	79 ± 0.01
13	Ca^{2+}, mg/l	631 ± 0.01	92 ± 0.01	220 ± 0.01	14 ± 0.01
14	COD, mg/l	15800 ± 2	3850 ± 2	1360 ± 1	115 ± 1
15	Efficiency of treatment for COD, %	-	75.6 ± 0.1	-	91.5 ± 0.1
16	BOD_5, mg/l	9800 ± 2	2450 ± 2	250 ± 1	18.3 ± 1
17	Efficiency of treatment for BOD_5, %	-	75.0 ± 0.1	-	92.8 ± 0.1

Data are mean value ± S.D., N=3, significant differences at $p < 0.05$ level.

The study of efficiency on the *Sargassum hystrix* biosorbent treatment of grain stillage showed a decrease in the cod to 3850 mg/l, BOD_5 to 2450 mg/l, which ensures 75% efficiency of treatment, methane mashes with a lower content of COD and BOD_5. The total content of nitrogen and phosphorus after biosorbents treatment decreases by 5 and 6 times, respectively, and significantly reduces the content of other indicators.

Thus, according to the main indicators of COD and BOD_5, as well as the content of other substances, treatment on the *Sargassum hystrix* biosorbent allows reducing the contamination of grain stillage by 75%, and methane mashes by 92%. The proposed biosorption method allows purifying wastewater with any concentration of pollutants, but depending on the initial concentration of COD and BOD indicators, the purification efficiency will be different.

It was found that the pH of the environment has a significant effect on the sorption of pollutants and the highest cleaning efficiency (92%) is achieved at a pH of 7.5. At the same time, at an acidic pH value, the cleaning efficiency was 75%. This is due to the electrokinetic properties of the surface of lignocellulosic biosorbent materials—the ability to change the charge depending on the pH (positive in acidic environment, negative in neutral and alkaline), which is due to the presence of different functional groups.

Industrial wastewater contains a large amount of organic and inorganic pollutants, and depending on their nature, adsorption occurs in different ways. Adsorption can occur using several processes, such as electron exchange, covalent interactions, ion exchange, and surface interaction. Different adsorbents can use different mechanisms, which depends on the type of pollutant (organic or inorganic), reaction conditions, adsorbent characteristics, and the interaction between the adsorbent and the adsorbate. Therefore, in further studies, it is planned to investigate various options for modifying plant materials to obtain highly efficient biosorbents for multifunctional use to solve environmental problems. For example, the combination of biochar modified with iron and manganese precipitate with a biosorbent made from oxidized rice straw makes it possible to use different adsorption mechanisms, such as ion exchange with electrostatic attraction and surface complexation. This opens up wide opportunities for wastewater treatment from pollutants of various nature. Therefore, in further studies, it is planned to investigate various options for modifying plant materials to obtain highly efficient biosorbents for multifunctional use to solve environmental problems. For example, the combination of biochar modified with iron and manganese precipitate with a biosorbent made from oxidized rice straw makes it possible to use different adsorption mechanisms, such as ion exchange with electrostatic attraction and surface complexation. This opens up wide opportunities for wastewater treatment from pollutants of various nature.

After use, plant biosorbents can be separated from the wastewater by filtration or precipitation and regenerated. This significantly reduces the cost of manufacturing biosorbents. However, the adsorption capacity of the regenerated sorbent will already be lower than that of the newly modified one [44]. Several methods are used to regenerate biosorbents: chemical, thermal, microwave, and supercritical fluid desorption (CO_2, water). Thermal regeneration is used to remove heavy metal ions from the adsorbent [45]. Chemical regeneration of biosorbents is the most widely used desorption method. It can be acidic using hydrochloric, phosphoric, nitric acids and alkaline using sodium hydroxide solutions. In an acidic environment, pollutant ions are desorbed from the surface of the adsorbent, and the biosorbent itself must be resistant to the chemical solvent [46]. Desorption by supercritical fluid is carried out at elevated pressure, which increases the cost of the method and limits its industrial application. The same limitations arise with microwave regeneration.

The application of biosorbent regeneration on an industrial scale depends on many aspects, such as the type of adsorbent, the pollutant, the stability of the adsorbent, the cost of the regeneration method and the amount of energy used. In many cases, it will be cheaper to recycle the spent adsorbents. Results have shown that nutrient-enriched biochar is an organic fertilizer that can replace synthetic fertilizers [47]. So far, relatively little attention has been paid to the conversion of spent adsorbents into useful products using various methods, but research is ongoing. Used adsorbents can be recycled for energy transfer, used as capacitors and catalysts. Metal-impregnated biosorbents from green biomass can replace carbon nanotubes and be used as supercapacitors [48].

4. Conclusions

Plant biosorbents after chemical modification increased their sorption capacity. This is explained by a change in the physicochemical properties of the material and an increase in the volume of pores and microcracks, as well as the formation of various functional groups on the surface of the sorbent.

Additionally, the composition and sorption properties of plant-based biosorbents were studied and analyzed, including *Paulownia*, aspen, hickory wood, *Ziziphus* bark, peach tree shavings, *Bagrina* grass, reed, algae, and cyanobacteria in their natural and modified forms. It was found that among the woody materials, biochar from *Paulownia* activated with phosphoric acid and urea had the highest adsorption capacity, while *Ziziphus* bark was highly effective in removing pollutants from wastewater. Among grasses, the best adsorption properties were observed in algae and in the biomass of the cyanobacterium *Oscillatoria princeps*, with the highest sorption capacity recorded in *Sargassum hystrix* (43 g/kg) at a solution concentration of 450 mg/dm³ and *Phragmites australis* (34 g/kg) at a solution concentration of 410 mg/dm³.

The study of the efficiency on the *Sargassum hystrix* biosorbent treatment of graine stillage and methane mashes with different indicators of COD and BOD_5 makes it possible to reduce the pollution of grain stillage by 75% and methane mashes by 92%. The proposed method of biosorption makes it possible to treatment wastewater with any concentration of pollutants, but depending on the initial concentration of indicators, the efficiency of treatment will be different.

Regeneration of biosorbents in the case of alcohol wastewater treatment is considered economically inexpedient, as its cost is equal to the cost of a new adsorbent. Used biosorbents can be utilized as organic fertilizer.

The obtained experimental results can serve as the basis for developing integrated water purification technologies, which will have a positive impact on both the economy and the environment. In further studies, it is planned to investigate various options for modifying plant materials to obtain highly efficient biosorbents for multifunctional use to solve environmental problems.

Author Contributions

Conceptualization, A.P. and K.D.; methodology, L.R. and L.B.-P.; software, H.T. and I.U.; validation, A.P., K.D., and L.B.-P.; formal analysis, H.T. and I.K.; investigation, I.K. and R.R.; resources, I.U. and R.R.; data curation, M.R.; writing—original draft preparation, A.P and B.L.; writing—review & editing, K.D and L.B.; visualization, H.T. and I.K.; supervision, K.D.; project administration, L.B.-P. and K.D.; funding acquisition, K.D. All authors have read and agreed to the published version of the manuscript.

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All data supporting the reported results are provided within the manuscript. Additional data can be made available upon reasonable request.

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Conflicts of Interest

The authors declare no conflict of interest.

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