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Review

Insights into the Use of Phytoremediation Processes for the Removal of Organic Micropollutants from Water and Wastewater; A Review

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Abstract: Greater awareness of micropollutants present in water and wastewater motivates the search for effective methods of their neutralization. Although their concentration in waters is measured in micro- and nanograms per liter, even at those levels, they may cause serious health consequences for different organisms, including harmful effects on the functioning of the endocrine system of vertebrates. Traditional methods of wastewater treatment, especially biological methods used in municipal wastewater treatment plants, are not sufficiently effective in removing these compounds, which results in their presence in natural waters. The growing interest in phytoremediation using constructed wetlands as a method of wastewater treatment or polishing indicates a need for the evaluation of this process in the context of micropollutant removal. Therefore, the present work presents a systematic review of the effectiveness in the removal of micropollutants from polluted waters by processes based on plant used. The article also analyzes issues related to the impact of micropollutants on the physiological processes of plants as well as changes in general indicators of pollution caused by contact of wastewater with plants. Additionally, it is also the first review of the literature that focuses strictly on the removal of micropollutants through the use of constructed wetlands.

Keywords: emerging contaminants; endocrine disrupting compounds; phytoremediation; constructed wetlands; removal efficiency

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1. Introduction

Water bodies nowadays demonstrate a strong chemical presence that is caused by human activities including farming, runoff from urban areas, industrial production, and transportation. This results in the appearance of new organic contaminants or a significant increase in the concentration of some compounds. Recent environmental research has also revealed new organic contaminants that were not detected before or whose presence has not been considered as significant. These have been dubbed Emerging Organic Contaminants (EOCs) and are created by hundreds of organic compounds belonging to different chemical classes. They have been detected in clean surface waters at levels of a few ng dm^{-3} and in polluted waters, where their range may span from a few to hundreds of μg dm^{-3} [1–3]. EOCs include substances of natural or anthropogenic origin whose presence has been detected for years as well as new ones that have become discernible due to the development of current detection techniques (GC-MS/MS, LC-MS/MS) and the introduction of new extraction procedures [4–7]. The presence of most of these within environmental compartments is not regulated by law, since they do not exert acute toxicity. However, their influence on aqueous organisms as well as on water consumers is more subtle and may manifest itself, for example, through endocrine disruption. Compounds that exhibit such activity or are suspected of it are named Endocrine Disrupting Compounds, or EDCs. According to a definition for EDCs provided by The Endocrine Society, they are "an exogenous chemical, or a mixture of chemicals, that interferes with any aspect of hormone action" [8]. They are products of a very diverse group of substances belonging to different chemical classes and causing varying biological effects. Most numerous among this group are pesticides, industrial chemicals, including phthalates or bisphenol A, surfactants, and steroids. Another genre of EOCs encompasses pharmaceutical and personal care products (PPCP). Due to the advancement of civilization, including the greater availability of medicines for humans and animal medicines as well as the increasing need for personal hygiene products, the annual production of PPCP is still growing. This group of compounds includes non-steroidal anti-inflammatory drugs (NSAID), synthetic hormones, antibiotics, lipid regulators, beta-blockers, retardants, preservatives, disinfectants, anti-inflammatory, and repellent agents [1–3]. The overconsumption of pharmaceuticals, free access to personal hygiene products, as well as human agricultural activity provide a huge amount of organic micropollutants that enter treatment plants along with wastewater [9,10]. Traditional activated sludge treatment plants are not very good at dealing with this type of contamination, making municipal wastewater plants the main source of EDCs in surface and ground waters. Persistant and nonadsorbed EDCs together with treated wastewater end up in surface waters. Worldwide studies have confirmed the presence of EDCs everywhere [1], including the surface waters of the northern Antarctic Peninsula [11]. Environmental concentrations of pharmaceuticals vary from a few ng dm^{-3} up to several hundreds of µg dm^{-3}. Their concentrations in drinking water are several orders of magnitude smaller than those found in raw wastewaters [1,12]. Table 1 summarizes concentrations of selected endocrine active compounds within the environment. Levels of pharmaceuticals in wastewater depend on the technology used to treat it and a given place's density of human population [13]. Non-steroidal anti-inflammatory drugs enter the environment mainly from sewage from hospitals and households as well as in human and animal excreta [9]. For example, hormonal therapies used for infertility treatment and hormonal contraception become sources of artificial hormones that wind up in the aqueous environment [14,15]. Another PPC group consists of sunscreen products used in cosmetics. Creams with filters that are applied to the skin to protect against the harmful effects of the sun easily wash off while playing on the beach. UV filters penetrate the skin, entering the body where they undergo metabolic transformation, resulting in the formation of new compounds, some of which are more persistent than their parent compounds [16]. People are also a source of mosquito and other insect repellants, which cause oxidative stress in those animals [17,18]. One representative of this group, N, N-diethyl-3-methylbenzamide (DEET), was found in and groundwater within the vicinities of landfills. Yet another group of pollutants includes industrial chemicals that have a wide range of applications and origins. Sources of these substances are the synthesis of plastics, production of surfactants, photography, metallurgy, vulcanization, or the production of drugs [11,19]. This group includes various types of surfactants, plasticizers, and benzotriazoles with the last category creating a problem for the environment on account of their resistance to UV radiation [20]. The prevalence of EDCs within the aqueous environment is unfavorable for organisms that live there. The harmful effects of these substances depend on their concentration and the animals' time of exposure to them. Since they are present in such minute quantities, they do not cause acute symptoms or toxic aftereffects. Nevertheless, continuous contact with small but constant amounts of EDCs has a negative impact on gender balance, reproductive success, functioning of the endocrine system, as well as on the health of new generations of aquatic organisms (with teratosis and low survival outcomes being quite frequent) [21]. It has been found that these compounds undergo bioaccumulation in mollusks, fish, as well as in aqueous plants [22–24], thus being incorporated into subsequent links of the food chain. It is believed that human exposure to EDCs is mainly caused by diet [25]. Although adverse health effects of long-term human exposure to EDCs are difficult to predict, some researchers have linked them to the obesity epidemic, the increasing number of ADHD cases, and insulin resistance to prenatal and early childhood exposure to

EDCs [26,27]. For these reasons, the search for better, more effective methods of wastewater treatment preventing the infiltration of these substances into the environment is still an important issue.

Table 1. Concentrations of selected endocrine active compounds within the environment.

Compounds	Water Sample	Concentration	References
Benzotriazoles			
4-methyl-1H-benzotriazole	Surface water	1.44–4345 ng·dm^{-3}	
5-methyl-1H-benzotriazole	Surface water	2–7181.4 ng·dm^{-3}	[11]
1H-benzotriazole	Surface water	<0.2–8529.8 ng·dm^{-3}	
UV filters			
benzophenone	Landfill leachate	0.15–16 µg·dm^{-3}	[12]
	Groundwater from landfill	0.028–0.492 µg·dm^{-3}	
benzophenone 3	Landfill leachate	<0.01–0.646 µg·dm^{-3}	[28]
Plasticizers			
di-n-butylphthalate	Freshwater	0.22–3.86 µg·dm^{-3}	[29]
dimethyl phthalate	Freshwater	0.02–0.25 µg·dm^{-3}	[29]
bis(2-ethylhexyl) phthalate	Freshwater	5.16–20.80 µg·dm^{-3}	[29]
Industrial chemicals			
bisphenol A	Surface water	up to 330 ng·dm^{-3}	[30]
		2.2–1030 ng·dm^{-3}	
4-tert-octylphenol	Surface water	1.0–2470 ng·dm^{-3}	[31]
4-nonylphenol	Surface water	28.1–8890 ng·dm^{-3}	
	Wastewater	0.19–102.54 µg·dm^{-3}	[32]
Insect repellents			
N,N-diethyl-3-methylbenzamide	Landfill leachate	73.7 ng·cm^{-3}	[33]
	Groundwater from landfill	0.019–16.901 µg·dm^{-3}	[34]
Pesticides			
atrazine	Surface water	1.8–18 mg·dm^{-3}	[14]
dimethomorph	Groundwater	0.002–0.03 µg·dm^{-3}	[10]
Antibacterial and antibiotics			
sulfamethoxazole	Drinking water	440 µg·dm^{-3}	[14]
ofloxacin	Drinking water	11.1–1330 ng·dm^{-3}	[35]
Non-steroidal anti-inflammatory drugs			
diclofenac	Surface water	8.8–127 mg·dm^{-3}	[14]
	Landfill leachates	0.081–0.61 µg·dm^{-3}	[36]
naproxen	Wastewater	0.78–551.96 µg·dm^{-3}	[32]
ibuprofen	Drinking water	50 µg·dm^{-3}	[14]
ketoprofen	Wastewater	0.95–233.63 µg·dm^{-3}	[32]
Hormones			
estrone	Wastewater	0.2–10.1 ng·dm^{-3}	[14]
17-β-estradiol			
17-α-ethinylestradiol	Ground water	0.5–230 ng·dm^{-3}	[14]
estrone	Surface water	0.7–75 ng·dm^{-3}	[31]
17-β-estradiol	Surface water	0.7–7.5 ng·dm^{-3}	[31]

Technological wastewater treatment in biological wastewater treatment plants consists of primary (suspension removal), secondary (scattering, dilution, baffle, biodegradation, and abiotic transformation), and optional tertiary (advanced physicochemical processes applied for the production of high-quality water, which is rarely used due to their high costs) processes. The main mechanisms of wastewater purification in classical wastewater treatment plants are biodegradation and sorption [37]. However, traditional water treatment plants relying on processes of coagulation and filtration are not designed to remove organic micropollutants. The modification of existing installations by adding supplementary modules such as membrane separators, membrane bioreactors, equipment utilizing ozonation and advanced oxidation methods, as well as through the use of adsorption onto unconventional solids, biosorption, or nanofiltration, allows a significant reduction in the content of micropollutants [38–40]. Although chemical and physicochemical processes are characterized by high purification efficiency, their main drawback is high extra costs incurred through the building of additional installations, application of chemical reagents, and utilization of more energy. What is more, the use of photochemical processes creates problems connected with the generation of reactive photo-intermediates and low levels of mineralization [41] as well as causes the generation of new contaminants, through, for example, the formation of halogenated aliphatic compounds or products of indirect photochemical reactions [42–44].

However, there is an alternative to traditional wastewater treatment and chemical cleaning methods. It involves the use of appropriate plants (aquatic, terrestrial, marsh) in special installations called constructed wetlands (CW). The removal of pollutants using the metabolic processes of plants is called phytoremediation [44] and meets all the requirements of a "green" purification process. The plants create favorable living conditions for microorganisms—bacteria and fungi which, in turn, play a very important role in processes that take place in CWs. Wetlands inhabited by plants employed for phytoremediation purification are also called natural, marsh, or root wetlands. The advantages of using phytoremediation modules, in comparison to other types of treatment plants, are tolerance to a lack of wastewater, simple operation and construction, as well as low maintenance costs. Studies analyzing the results of wastewater purification using plant systems show that phytoremediation is both environmentally friendly and characterized by high efficiency in removing pollutants. It has been found that nearly 100% efficiency in removing organic compounds from wastewater may be achieved in such systems [45]. The efficiency of phytoremediation depends on the types of utilized plants, their number, process time, as well as the type of soil and constructed wetland [46].

Plants used for phytoremediation should be characterized by fast biomass growth, low sensitivity to the variability in the composition of treated waters or soils, as well as high content of salts, phosphates, and nitrogen [47]. These requirements are fulfilled by some higher terrestrial plants such as the energetic willow *Salix vinimalis* L. [48], poplar *Populus deltoides*, hydrophytic plants (common reed *Phragmites australis*, common bulrush *Typha latifolia*, lakeshore bulrush *Schoenoplectes lacustris*, burreed *Sparganium ramusom*, great manna grass *Glyceria aquatica*, yellow flag *Iris pseudoacorus*, grassy plant species *Carex* from the family of *Cyperaceae* commonly known as sedges, some species of *Myriophyllum spicatum*, for example, the *Eurasian watermilfoil*, flowering plants such as fool's-watercress, *Apium nodiflorum* (L.) [49], as well as aquatic plants of the genus *Lemnaceae* such as duckweed *Lemna minor* [50,51] and watermeal *Wolffia arrhiza* [52].

Currently, phytoremediation is a rapidly developing method for water purification that is especially recommended for small, household sewage treatment plants. Originally, plants were used to remove heavy metals from contaminated mediums [49] and improve their general parameters such as pH, electric conductivity (EC), total suspension (TS), chemical (COD) and biological oxygen demand (BOD_5), total organic carbon (TOC) and its dissolved form (DOC), total nitrogen (TN), dissolved nitrogen (DN), total phosphorus (TP), its orthophosphorus form (OP), and others [49]. Today, the interests of researchers are focused on the effectiveness of plant organisms in removing organic micropollutants, including compounds from the EDCs group, and possibilities for the use of selected plants to achieve this purpose. The proper selection of a plant or plants requires detailed studies that provide information regarding their rate of toxicant biodegradation and EDCs removal. Although some great review articles on water or soil phytoremediation [44,53–56] have been published recently, none of them deals with details concerning organic micropollutant removal based on plant metabolism.

Taking the above into account, the authors of the present article have decided to collect information on the removal of organic micropollutants using phytoremediation processes.

2. Main Phytoremediation Mechanisms

It has been found that the removal of pollutants in plant-based treatment installations occurs through two types of processes: (a) abiotic, based on hydrolysis, photodegradation, and adsorption on the plant's surface, and (b) biological, through bioaccumulation and metabolism in plant tissues [45]. Detailed information on the individual contributions of each of the processes allows us to explain the mechanisms of removing micropollutants supported by plant metabolism. It is possible to distinguish the following biological processes:

- Phytostabilization—the plant immobilizes inorganic impurities in its roots and does not eliminate them with binding occurring mainly through the mechanism of complexation;
- Phytoacumulation or phytoextraction—pollutants accumulate in different parts of the plant. This process, which uses the mechanism of hyperaccumulation, is very significant in respect to heavy metal removal;
- Phytodegradation or phytotransformation—plants metabolize, using the mechanism of degradation within the plant, organic pollutants taken from water or soil into, usually, less toxic substances;
- Phytovolatilization—plants absorb organic and inorganic pollutants and then eliminate them through the processes of transpiration;
- Rhizofiltration—the roots of the plant create conditions favorable to soil-dwelling microorganisms, which then biologically degrade organic and inorganic pollutants present within that root system [44,48,56–58].

The contribution of biotic and abiotic processes to the overall efficiency of xenobiotic removal depends on the type of compound that is to be removed, its concentration, the composition of its matrix, as well as the type of plant, and access to light [44,59]. It is obvious that different parts of a given plant process contaminants in varying ways. Additionally, the absorption of organic micropollutants by a given plant is highly dependent not only on the physicochemical properties of that particular chemical compound but also on its concentration [59]. If the pollutants are not photodegradable, the plant may further metabolize or accumulate them. For example, it has been found that photodegradation processes are responsible for 67–77% degradation of diclofenac, while only 2–3% of this compound is removed as a result of phytoremediation [60]. It has also been observed that the accumulation level of diclofenac in plant roots increases along with exposure time. Root absorption was highest during the first 72 h of exposure, but there is also evidence that translocation of the drug from the roots to the plant's shoots may occur. The mass balance is higher for roots than for shoots, and it is higher at lower concentrations of this particular micropollutant (0.5 mg dm^{-3}). The plants' uptake of diclofenac through the roots at concentrations of 0.5 mg dm^{-3}, 1.0 mg dm^{-3}, and 2.0 mg dm^{-3} is 1.02–1.79%, 0.17–0.62%, and 0.7–0.72%, respectively [60], while the drug's uptake through the shoots at the same concentrations is 0.28–0.75%, 0.13–0.34%, 0.11–0.24% [60]. However, some substances, such as hydrophobic compounds, bind strongly to the roots and are not transferred to other parts of the plant [60].

Another factor influencing the effectiveness of EDC removal by plants is the presence of endophytic microorganisms (fungi and bacteria) that support the plants' metabolism or protect them against the harmful effects of xenobiotics [61,62].

3. Characteristic of Phytoremediation Modules Used in Sewage Treatment Plants

Phytoremediation modules used in wastewater treatment installations are specially designed wetlands containing aquatic or marsh plants. The constructed wetland consists of a reservoir, a suitable filter material (substrate), and planted vegetation. Contaminated water is introduced into a tank filled with ground and plants and, depending on the type of flow, flows freely or under the surface. At the end of the process, it is expelled [63,64]. CWs employing floating aquatic vegetation are usually built as a system with rooted aquatic vegetation or as a chain of several ponds with a primary stage of mechanical wastewater treatment [65]. One type of constructed wetland has originated in China and is especially suitable to its prevailing climate [66].

The effectiveness of constructed wetlands, as well as the phytoremediation process itself, can be influenced by many variables including:

- Type of flow—within this category, it is possible to distinguish the following systems:

(a) free water surface systems (FWS), (b) subsurface flow systems (SSF) that can be further divided into (b1) horizontal subsurface flow constructed wetlands (HSSF CWs) and (b2) vertical subsurface flow constructed wetlands (VSSF CWs), and (c) hybrid systems that combine different types of flows to create multistage processes. Depending on the type of flow used within a given treatment plant, parameters such as insolation, access to oxygen, redox potential, and adsorption surface impact the efficiency of pollutant removal. For example, HSSF CWs are very effective in the removal of organic pollutants [63,64]. Flow types have been presented in Figure 1:

- Type of plants used—this variable is very important, since some plants can accumulate pharmaceuticals while others may accumulate heavy metals;
- Type of substrate—this variable affects, among others, the physical and chemical processes occurring within a treatment plant with biodegradation processes being more effective under aerobic conditions than under anaerobic conditions [32,56,57,67–69].

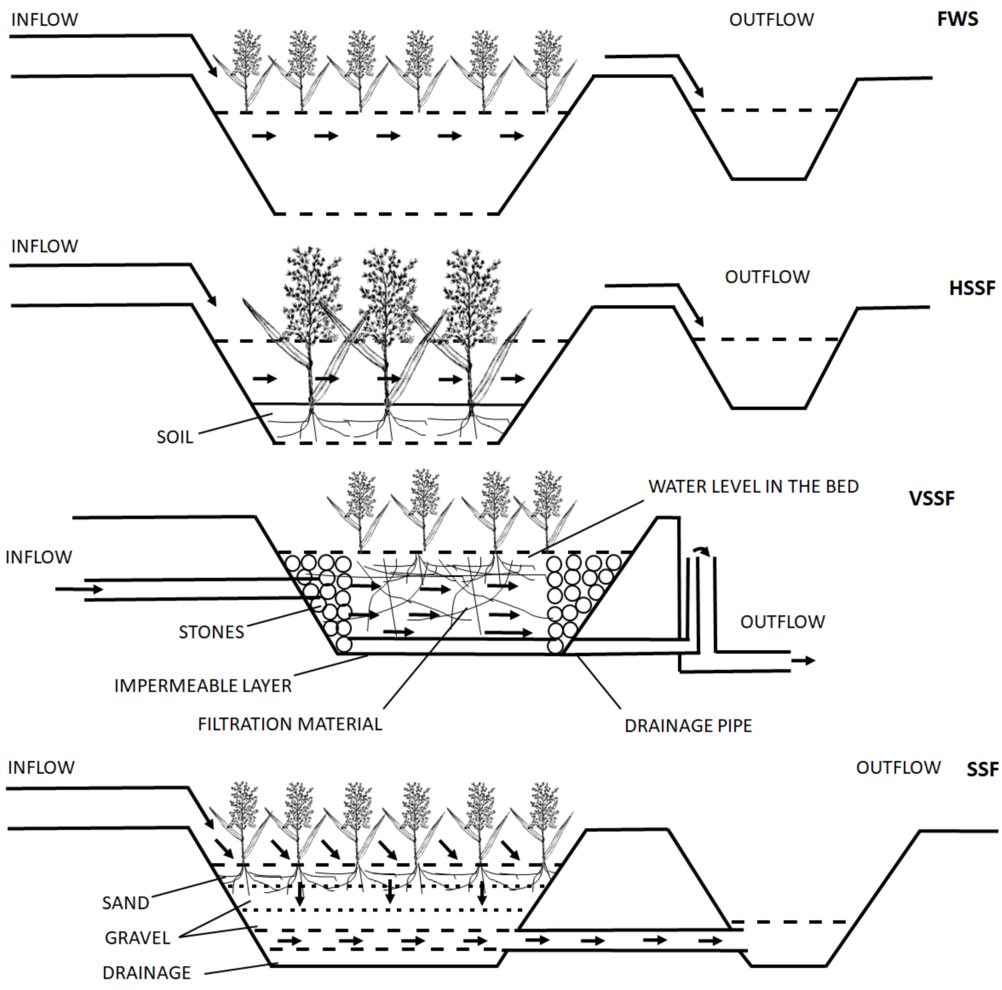

Figure 1. Types of flows in constructed wetlands.

Constructed wetlands are environmentally friendly, use natural processes, and create ideal conditions for insects and amphibians. What is more, they can be used in protected areas, and are cheap as well as easy to build, operate, and maintain. Furthermore, plants used in phytoremediation can later be used for the production of biofuel. One limitation of these types of systems is that they require significant area to be constructed [63].

4. Plants Applied in the Remediation of Wastewater

Ensuring an adequate level of contamination removal through the use of wetlands requires the selection of appropriate plant species. The construction of CWs requires the use of aquatic plants that can transport oxygen to their roots, creating an oxygen zone favorable to microorganisms. Subsequently alternating aerobic and anaerobic zones form, ensuring a higher efficiency of pollutant removal. The selection of plants depends on the climatic conditions in which they will live as well as on the type of pollution that they will be required to neutralize. It is essential that they are able to grow in wetlands with low oxygen content and are resistant to temporary shortages of sewage and introduced pollutants. The two types of plants usually utilized in CWs are called macrophytes or hydrophytes. These can further be divided into the following groups:

1. Surface macrophytes (helophytes)—grow on wetlands, develop above ground in boggy or fully submerged soil. Examples include *Acorus calamus* L., *Carex rostrata* Stokes, *Phragmites australis*, *Schoenoplectus lacustris* (L.) Palla, and *Typha latifolia* L.;
2. Floating leaf macrophytes (rhizophytes)—their leaves float on or break the water's, surface but they are rooted in soil. Examples include *Nymphaea alba* L. and *Nuphar lutea*;
3. Submerged macrophytes—underwater plants that float in water but are rooted in soil. Examples include *Myriophyllum spicatum*, *Ceratophyllum demersum* L.;
4. Freely floating macrophytes (pleustophytes)—plants that float on the water's surface without being rooted in soil. Examples include *Lemna minor* L., *Spirodela polyrhiza* L., and *Eichhornia crassipes* [69].

Plants from the *Lemnaceae* family are most commonly used in systems with floating aquatic vegetation. This family of plants consists of two subfamilies: *Lemnoideae* and *Wolffioideae*. *Lemnoideae* consist of genera *Lemna* (seven species: *L. aequinoctialis*, *L. gibba*, *L. minor*, *L. minuta*, *L. trisulca*, *L. turionifera*, and *L. valdivana*) and *Wolffia* (five species: *W. arrhiza*, *W. borealis*, *W. brasiliensis*, *W. columbiana*, and *W. globosa*), while the *Wolffioideae* is made up of genera *Spirodela* (two species: *S. polyrhiza*, *S. punctata*) and *Wolffiella* (two species: *W. lingulata*, *W. oblonga*) [70]. The plants of the *Lemnaceae* family occur in freshwater reservoirs at all latitudes except for the polar regions. Due to their ease of cultivation and resistance to contamination, they are often employed in processes utilizing phytoremediation but are also used as ornamental plants in ponds and aquariums, in herbal medicine as well as food for humans and animals [71]. Duckweeds are also currently used for the production of biofuels [72–74].

5. Effectiveness of Micropollutant Removal by Plants

As had been mentioned earlier, the effectiveness of phytoremediation removal of organic compounds depends on the type of chemical compound, its concentration, the plant used, as well as the type of wetland. Available data on plant organisms' pollutant removal efficiency have been gathered in Table 2.

Table 2. Removal efficiency of micropollutants in plants-based systems.

Plants	Compounds	Analytes Concentrations	Time of Exposure	Type of Installation, Type of Medium	Removal	Remarks	Ref.
	Benzotriazoles						
Lemna minor	1H-benzotriazole (BTR)	150 $\mu g \cdot dm^{-3}$	constant illumination	laboratory experiment, SIS medium	>99.9%	36 days experiment	[45]
	4-methyl-1H-benzotriazole (4TTR)				48.2%		
	5-methyl-1H-benzotriazole (5TTR)				>99.9%		
	xylytriazole				>99.9%		
	5-chlorobenzotriazole (CBTR)				>99.9%		
Cyperus alternifolius	oxybenzone	$5 \cdot 10^{-6}$ $mol \cdot dm^{-3}$	12/12h light/darkness	laboratory experiment, Hoagland medium	86.5%	120h experiment	[75]
		$2{,}5 \cdot 10^{-5}$ $mol \cdot dm^{-3}$			81.4%		
Wolffia arrhiza	dibutyl phthalate	100 $\mu g \cdot dm^{-3}$	16/8h light/darkness	laboratory experiment, non-treated wastewaters from WWTP and Hutner's medium	87.2%		[76]
	bis(2-ethylhexyl) phthalate				97.7%		
Lemna minor L.		1 $\mu g \cdot dm^{-3}$	12/12h light/darkness	tap water	62.5%		[77]
Lemna sp.		100 $\mu g \cdot dm^{-3}$	12/12h light/darkness	secondary-treated wastewaters from WWTP	96%	external experiment, natural conditions, Spain	[78]
Ceratophyllum demersum	bisphenol A	1–100 $\mu g \cdot dm^{-3}$	16/8h light/darkness	laboratory experiment, Hoagland medium	99.2%		[79]
Riccia fluitans					99.5%		
Spirodela polyrhiza					98.5%		
Limnobium leavigatum					89.9%		
Cyperus isoclaudus					85.9%	in 4 days	
Eichhornia crassipes		20 $\mu g \cdot dm^{-3}$		SSF-CW, synthetic wastewater	87.7%		[80]
Cyperus isoclaudus					95.5%	gravel and bamboo charcoal as support medium	
Phragmites australis		~0.6 $\mu g \cdot dm^{-3}$		HSF-CW, effluent of an anaerobic pond as primary treatment	70.2%		[81]
Helicionia pistacorum					73.3%		
Lemna minor L.	bisphenol S	1 $\mu g \cdot dm^{-3}$	12/12h light/darkness	tap water	72.2%		[77]
	bisphenol B				85.7%		
Helicionia pistacorum	nonylphenol			HSF-CW, effluent of an anaerobic pond as primary treatment	62.8%		[81]
Phragmites australis					52.1%		
Azolla filiculoides	hydrazine		16/8h light/darkness	Hoagland medium	93%	24h experiment	[82]
					>99.9%	96h experiment	
Azolla filiculoides	sodium dodecyl benzene sulfonate	10–40 $mg \cdot dm^{-3}$	16/8h light/darkness	nitrogen-free medium	77.79%		[83]
Typha domingensis	phenol	500 $mg \cdot dm^{-3}$		tap water	75%	15 days experiment	[84]
Typha angustifolia L.	1,2-dichloroethane	390 $mg \cdot dm^{-3}$		wastewater from a petrochemical industry; HSSF CWs	>99.9%	external experiment, natural conditions, Malaysia	[85]
Spirodela polyrhiza	4-tert-butylphenol	$3.3 \cdot 10^{-5}$ $mol \cdot dm^{-3}$	16/8h light/darkness	water samples from aquatic systems	63%	external experiment, natural conditions, Japan	[86]
Eichornia crassipes	pentachlorophenol	$2 \cdot 10^{-6}$ $mol \cdot dm^{-3}$	12/12h light/darkness	distilled water	47%	12h experiment	[87]
Spirodela polyrhiza	DEET	25 $mg \cdot dm^{-3}$	14/10h light/darkness	laboratory experiment with synthetic wastewaters	32.6%		[88]
Lemna minor	dimethomorph	25 $\mu g \cdot dm^{-3}$		laboratory experiment, nutrient medium	36%	96h experiment	[89]
Spirodela polyrhiza					32%		
Lemna minor		50 $\mu g \cdot dm^{-3}$			28%		
Spirodela polyrhiza					26%		
Lemna minor		600 $\mu g \cdot dm^{-3}$			19%		
Spirodela polyrhiza					14%		
Lemna minor		600 $\mu g \cdot dm^{-3}$			15%	plant density 0.05 g/flask	
					19%	plant density 0.10 g/flask	
					20%	plant density 0.15 g/flask	
					22%	plant density 0.20 g/flask	
Spirodela polyrhiza		600 $\mu g \cdot dm^{-3}$			8%	plant density 0.05 g/flask	
					14%	plant density 0.10 g/flask	
					17%	plant density 0.15 g/flask	
					18%	plant density 0.20 g/flask	
Lemna minor	diquat	44.4 $\mu g \cdot dm^{-3}$	14/10h light/darkness	dechlorinated tap water filtered through activated-charcoal cartridges	>99.9%	16 days experiment	[90]
		222.2 $\mu g \cdot dm^{-3}$			>99.9%		
	fomesafen	44.4 $\mu g \cdot dm^{-3}$			54.7%		
		222.2 $\mu g \cdot dm^{-3}$			74.4%		

Table 2. *Cont.*

Plants	Compounds	Analytes Concentrations	Time of Exposure	Type of Installation, Type of Medium	Removal	Remarks	Ref.
	Benzotriazoles						
Typha latifolia	terbuthylazine	0.4 mg·dm^{-3}		HSF-CW	58.4%		[91]
Phragmites australis					73.7%		
Canna indica	chlorpyrifos	100 µg·dm^{-3} 200 µg·dm^{-3} 300 µg·dm^{-3}		SSF-CW	88.07% 93.76% 96.55%	24h experiment	[92]
Phragmites australis/ Typha oruentalis/ Vetiveria zizanioides/ Canna indica	sulfamethoxazole (SMX)	2.3 µg·dm^{-3}		HSSF-CW, VSSF-CW	70.1–76.3%		[93]
Lemna minor		10 µg·dm^{-3}	16/8h light/darkness	secondary biologically treated wastewater	73%	14 days experiment	[94]
Cyperus alternifolius					<80%		
Cyperus alternifolius	ofloxacin (OFX)	10–500 µg·dm^{-3}		water simulating synthetic secondary sewage from WWTP	90%		[95]
Cyperus alternifolius	roxithromycin (ROX)				85%		
Lemna minor	cefadroxil (CFD)				>99.9%		
Lemna minor	metronidazole (METRO)	10 µg·dm^{-3}	16/8h light/darkness	secondary biologically treated wastewater	96%	14 days experiment	[94]
Lemna minor	trimethoprim (TRI)				59%		
Chrysopogon zizanioides (L.) Roberty	ciprofloxacin (CIP) tetracycline (TTC)	10 mg·dm^{-3}		wastewaters from WWTP	60–94% 89->99.9%	external experiment, natural conditions, USA	[96]
Spirodela polyrhiza		25 mg·dm^{-3}	14/10h light/darkness	laboratory experiment with synthetic wastewaters	97.7%		[88]
Salix alba+ Iris pseudacorus+ Juncus effusus+ Callitriche palustris+ Carex carophyllea		10 mg·dm^{-3}		SVF-CW, wastewaters from hospital's WWTP	0->99.9%	external experiment, natural conditions, Thailand	
Phragmites australis	paracetamol				20–69%	external experiment, natural conditions, Thailand	[97]
Phragmites australis + Phalaris arundinacea				HSF-CW, wastewaters from hospital's WWTP	86.2-99.6%.		
Phragmites australis		35 µg·dm^{-3} 30 µg·dm^{-3}			95% 45%		
Phragmites australis + Typha latifolia		750 ng·dm^{-3}			51.7–99%		
Typha angustifolia+ Chrysopogon zizanioides+ Cyperus papyrus					58.1%		
Phragmites australis		3–71 µg·dm^{-3}		HSSF-CW in the water- shed of a drinking water reservoir	89–99.6%	external experiment, natural conditions, Czech Republic	[98]
Phalaris arundinacea		0.35–180 µg·dm^{-3}			86.2–88.7%		
Lemna gibba L.		0.02–1 mg·dm^{-3}	14/10h light/darkness	laboratory experiment, Murashige and Skoog medium	89–92.5%		[99]
Juncus effusus/ Typha latifolia/ Berula erecta/ Phragmites australis/ Iris pseudacorus	ibuprofen	10 µg·dm^{-3}		water unsaturated CW, water saturated CW, aerated water saturated CW, tap water	29–99%		[100]
Lemna sp.		100 µg·dm^{-3}	12/12h light/darkness	secondary-treated wastewaters from WWTP	93%	external experiment, natural conditions, Spain	[78]
Spirogyra sp.					92%		
Phragmites australis		1.9–64 µg·dm^{-3}		HSSF-CW in the water- shed of a drinking water reservoir	51.6–75%	external experiment, natural conditions, Czech Republic	[98]
Phragmites australis + Phalaris arundinacea		6.6–36 µg·dm^{-3}			45.6–49.4%		
Phragmites australis		140–5400 ng·dm^{-3}			11.5–67.1%		
Phragmites australis + Phalaris arundinacea		10–12000 ng·dm^{-3}			29.3–58%		
Phalaris arundinacea	diclofenac	0.5 mg·dm^{-3}	14/10h light/darkness	laboratory experiment, vertical-flow CW, synthetic wastewaters	47.3%	the loading frequency 1 pulse per day	[101]
Phalaris arundinacea					74.2%	the loading frequency 4 pulses per day	

Table 2. *Cont.*

Plants	Compounds	Analytes Concentrations	Time of Exposure	Type of Installation, Type of Medium	Removal	Remarks	Ref.
	Benzotriazoles						
Spirogyra sp. *Lemna sp.*		100 μg·dm^{-3}	12/12h light/darkness	secondary-treated wastewaters from WWTP	54% 48%	external experiment, natural conditions, Spain	[78]
Scripus validus		0.5-2 mg·dm^{-3}	12/12h light/darkness	laboratory experiment, Hoagland medium	85–98%		[60]
Phragmites australis	ketoprofen	<10–6500 ng·dm^{-3}		HSSF-CW in the water- shed of a drinking water reservoir	46.9–91.2%	external experiment, natural conditions, Czech Republic	[98]
Phragmites australis + Phalaris arundinacea		19–2600 ng·dm^{-3}			18.1%		
Scripus validus	naproxen	0.5–2 mg·dm^{-3}	12/12h light/darkness	Hoagland medium	90%	external experiment, natural conditions, Spain	[60]
Lemna sp. *Spirogyra sp.*		100 μg·dm^{-3}	12/12h light/darkness	secondary- treated wastewaters from WWTP	94% 94%	external experiment, natural conditions, Spain	[78]
Ceratophyllum demersum, Riccia fluitans, Limnobium laevigatum, Spirodela polyrhiza	17-α-ethinylestradiol	1–100 μg·dm^{-3}	16/8h light/darkness	laboratory experiment, Hoagland medium	>99.9%	addition of crude enzymes	[79]
Lemna species		10 mg·dm^{-3}	12/12h light/darkness	laboratory experiment, synthetic wastewater	>95%	6 days experiment	[102]
Cyperus isoclaudus *Eichhornia crassipes* *Cyperus isoclaudus*		20 μg·dm^{-3}		SSF-CW, synthetic wastewater	81.4% 67.8% >99.9%	gravel and bamboo charcoal as support medium	[80]
Phragmites australis		0.94–3.62 ng·dm^{-3}		vertical-flow CW, effluent from WWTP	75.3%	external experiment, natural conditions, Japan	[102]
Phragmites australis + Phalaris arundinacea		5.9 ng·dm^{-3}		HSSF-CW, municipal wastewater	85%	external experiment, natural conditions, Czech Republic	[103]
Ceratophyllum demersum, Riccia fluitans *Limnobium laevigatum* *Ceratophyllum demersum*	estrone	1–100 μg·dm^{-3}	16/8h light/darkness	laboratory experiment, Hoagland medium	>99.9% 89.4% 87.4%	addition of crude enzymes	[79]
Lemna species		10 mg·dm^{-3}	12/12h light/darkness	laboratory experiment, synthetic wastewater	>95%	6 days experiment	[102]
Phragmites australis		1.17–6.18 ng·dm^{-3}		vertical-flow CW, effluent from WWTP in Japan	67.8%		[104]
Ceratophyllum demersum, Riccia fluitans *Limnobium laevigatum* *Ceratophyllum demersum*	17-β-estradiol	1–100 μg·dm^{-3}	16/8h light/darkness	laboratory experiment, Hoagland medium	>99.9% 95.5% 95.7%	addition of crude enzymes	[79]
Lemna species		10 mg·dm^{-3}	12/12h light/darkness	laboratory experiment, synthetic wastewater	>95%	6 days experiment	[102]
Phragmites australis		2.94–4.65 ng·dm^{-3}		vertical-flow CW, effluent from WWTP in Japan	84.0%		[104]
Cyperus isoclaudus *Eichhornia crassipes* *Cyperus isoclaudus*	levonorgestrel	100 μg·dm^{-3}		SSF-CW, synthetic wastewater	>99.9% >99.9% 99.8%	gravel and bamboo charcoal as support medium	[80]

CW—constructed wetland; HSSF—horizontal subsurface flow; SVF—subsurface vertical flow.

The data collected in Table 2 show that the removal efficiency of micropollutants by plants spans a wide range varying from 8% to nearly 100% and is dependent on both the type of compound and plant species. For example, the biodegradation of dimethomorph by *Spirodela polyrhiza* was achieved at only 8%, while that of benzotriazole (without 4TTR), hydrazine, 1,2-dichloroethane, diquat, cefadroxil, tetracycline, paracetamol, 17-α-ethinylestradiol, estrone, β-estrelestradiol, and levonorgestradiol approached 100% (>99.9%). Species of *Lemna minor*, *Azolla filiculoides*, *Typha angustifolia* L., *Chrysopogon zizanioides* (L.) Roberty, *Eichhornia crassipes*, and *Cyperus isoclaudus* have demonstrated the best (as high as >99.9%) efficiency in the removal of examined micropollutants. *Canna indica* removed concentrated chlorpyrifos displaying similar effectiveness as *Lemna minor* with respect to fomesafen. On the other hand, oxybenzone, at lower concentrations is removed well by *Cyperus alternifolius*, while DEET is efficiently eliminated by *Spirodela polyrhiza* or *Lemna minor*. It was also observed that tetracycline removal efficiency was higher with the application of two plant species (*Phragmites australis* and *Phalaris arundinacea*) rather than just one (*Phragmites australis*). Additionally, it was found that dimethomorph removal efficiency using *Spirodela polyrhiza* or *Lemna minor* was higher at greater plant density. Yet another factor that impacts phytoremediation efficiency is the length of time during which the plant remains in contact with contaminated water and, for example, the removal of hydrazine by *Azolla filiculoides* increases constantly over time. The data presented above suggest that higher removal rates can be expected at longer process times as well as with the presence of more plants. The lack of correlations between the removal efficiency and concentration of micropollutants is related to the individual properties of studied pollutants and the plants' resistance to a given type of compound. The efficiency of benzotriazole removal (BTR, 4TTR, 5TTR, CBTR) was also tested under different conditions: in the dark, with the presence of light, as well as with the combination of light and plants (*Lemna minor*). Concentrations of benzotriazoles decreased slightly after some time in the dark, while in the presence of light removal efficiency, they rose to about 36% over 36 days. In the presence of *Lemna minor*, however, BTR, 5TTR, or CBTR removal efficiency approached 100%. Therefore, it can be concluded that the removal efficiency of these substances through plant uptake is much greater than through hydrolysis or photodegradation.

6. Toxic Effects of Micropollutants on Plants

Numerous plants can survive in toxic conditions and are capable of reducing some pollutants' harmful effects or eliminate them [57]. However, most plants are not completely immune to harmful substances and may, depending on the nature of specific micropollutants, undergo various changes including the acceleration or inhibition of growth, an increase in the number of antioxidant enzymes, inhibition of the process of photosynthesis, or, simply, plant poisoning and death. Various factors can cause abiotic stress leading to the overproduction of reactive oxygen species that can destroy the plant's lipids and proteins. However, there are some plants that try to cope with these circumstances through, for example, the synthesis of antioxidants or the enhanced activity of antioxidant enzymes. The observation of plants cultivated in wetland tanks revealed that plants that can cope with stressful conditions do not decrease in size or exhibit inhibited development. They also do not change color, indicating normal photosynthesis as well as the efficiency of their antioxidant enzyme systems. After some time, the plants adapt to conditions where pollution is present and display normal growth. The data in Table 3 show the response of tested plants to various concentrations of xenobiotics. This information can become the basis for the design and construction of purification installations.

Table 3. The impact of micropollutants on aquatic plants.

Micropollutants	Plants	Symptoms of Abiotic Stress	Comments	Ref.
dimethomorph, C = 600 $\mu g \cdot dm^{-3}$	*Lemna minor*	inhibition of growth by 21%, inhibition of photosynthesis	the increase in plant density caused a lower growth rate, reaching 26% at 0.20 g/E-flask	[89]
	Spirodela polyrhiza	inhibition of growth by 19%, inhibition of photosynthesis	the increase in plant density caused a lower growth rate, reaching 24% at 0.20 g/E-flask	
oxybenzone	*Cyperus alternifolius*	increased activity of antioxidant enzymes	12/12 h light/darkness	[75]
hydrazine, C = 0–1000 $\mu g \cdot dm^{-3}$	*Azolla filiculoides*	plant growth	hydrazine was a nitrogen source for the plant; 16/8 h light/darkness	[82]
sodium dodecyl benzene sulfonate, C = 0–40 $mg \cdot dm^{-3}$	*Azolla filiculoides*	stunted growth, effect index increases with increasing concentration, the activity of antioxidant enzymes was stimulated at each concentration and increased with longer exposure to the surfactant, content of anthocyanins, H_2O_2, antioxidant activity, and electrolyte leakage were higher with the duration of the test, the total content of chlorophylls tested after 7 days decreased with increasing concentration	after 7 days, the total carotenoid content was reduced by surfactant at a concentration of 30 $mg \cdot dm^{-3}$ and 40 $mg \cdot dm^{-3}$; 16/8 h light/darkness	[83]
diquat, C = 44.4 $\mu g \cdot dm^{-3}$	*Lemna minor*	80% fading leaves, lower relative leaf number (FRG) and relative leaf area (RFA)	14/10 h light/darkness	[90]
diquat, C = 222.2 $\mu g \cdot dm^{-3}$		100% leaf fading, much lower FRG and RFA		
fomesafen, C = 44.4 $\mu g \cdot dm^{-3}$		relative leaf number increase, RFA lower		
fomesafen, C = 222.2 $\mu g \cdot dm^{-3}$		inhibition RFN, much smaller RFA		
diclofenac, C = 25 $mg \cdot dm^{-3}$	*Lemna minor*	reduction in the amount of chlorophylls by 64%	diclofenac, nicotine, and 3,4-dichlorophenol inhibited chlorophyll content more than leaf number; 16/8 h light/darkness	[105]
bisphenol A		the total content of chlorophylls decreased with increasing concentration		
3,4-dichlorofenolu				
ketokonazol benzyldimethyldodecylammonium chloride climbazole fluconazole	*Lemna minor*	the size and number of leaves decreased and the color of the leaves changed	the plant had a hard time returning to previous state; 16/8 h light/darkness	[106]
mixture of micropollutants	*Lemna minor*	100% inhibition of the growth rate	samples of wastewater contained bisphenol A, valsartan and 2-OH-benzothiazole	[107]
mixture of micropollutants	*Lemna minor*	plant's growth inhibited by about 25% in the summer, no growth in the winter, in winter is more antioxidants	samples of sewage contained surfactants (heavy metals, hydrocarbons, nitrogen and phosphorus compounds)	[108]
perfluorooctanoic acid	*Lemna minor*	no change in the number of leaves, growth rate, chlorophyll content, and photosynthesis efficiency	any harmful effect on the plant; 16/8 h light/darkness	[109]

The data provided in Table 3 show that in most cases, contact between plants and organic micropollutants causes the deterioration of the plants' condition manifested by a reduction in their number or their leaf surface as well as in the lower concentration of chlorophylls reflected through changes in leaf color. Plant degeneration depends on the type of added compound and its concentration. However, it is clear that the plants attempt to fight the toxicity of their environment evident in an increase in the activity of antioxidant enzymes. The enzymatic and non-enzymatic response of the considered plants to abiotic stress allowing them to survive in unfavorable conditions is a key condition for the plant to be successfully used for the phytoremediation of organic micropollutants.

7. Improving Overall Pollution Indicators

Aquatic plants cultivated in wastewater change the physicochemical parameters of purified water such as its pH, conductivity, and temperature, as well as its chemical parameters including chemical oxygen demand (COD), biological oxygen demand (BOD), dissolved organic carbon (DOC), total organic carbon (TOC), nitrogen NH_4^+-N, NO_3^--N, total nitrogen (TN), total phosphorus (TP), and total suspended solids (TSS). Some indicators concerning pollution were observed to decrease, such as the content of nitrogen or phosphorus, while others were seen to increase. Table 4 summarizes the impact of aquatic plants used for purification purposes on the parameters of pollutants in tested water and wastewater samples.

Table 4. The influence of plants growth on the general pollution indicators.

Plants	Pollution Indicators Changes	Ref.
Lemna minuta	DO ↓65%, NO_3^--N ↓85%	[110]
Lemna minor	DO ↓30%, NO_3^--N ↓60%, PO_4^--P ↓70%	
Lemna minor	BOD ↓90%, COD ↓88%	[76]
Wolffia arrhiza	BOD ↓95%, COD ↓90%, TN ↓65–90%, TP ↓15–83%	
Cyperus alternifolius	COD ↓70–85%, NH_4^+-N ↓ 30–55%, TN ↓60–75%	[95]
Cyperus papyrus	TSS ↓75%, BOD ↓61%, COD ↓44%, NH_4^+-N ↓26%, NO_3^--N ↓22%, TP ↓57%	[111]
Bacopa monnieri (L.) Pennell	TSS ↓90%, COD ↓76%, BOD ↓80%, P ↓55%, NO_3^--N ↓65%	[112]
Eichhornia crassipes	NO_3^--N ↓75%, PO_4^--P ↓79%, TSS ↓, EC ↓, S ↓, pH ↑	[113]
Eichhornia crassipes	NO_3^--N ↓55%, PO_4^--P ↓86%, TSS ↓, EC ↓, S ↓, pH ↓	
	COD ↓79%, BOD ↓86%, TN ↓76.61%, TP ↓44.84%, TSS ↓73.02%, PO_4^-–P ↓38.69%, NH_4^+-N ↓72.48%	[114]
Salvinia molesta	TP ↓83%, NH_4^+-N ↓73%, COD ↓74.1%	[52]
Pistia stratiotes	TN ↓64.2%	
Typha domingenis	TOC ↓, BOD↓, COD↓, pH ↑	[84]
Typha angustifolia	BOD ↓76%, NH_4^+-N ↓86%, NO_3^--N ↓41%, PO_4^-–P ↓100%	[115]
Canna iridiflora	BOD ↓85%, NH_4^+-N ↓82%, NO_3^--N ↓50%, PO_4^-–P ↓100%	
Phalaris arundinacea	DOC ↓95%, TKN ↓90–95%, NH_4^+-N ↓59–96%, PO_4^--P ↓25–31%	[101]
Phragmites australis	TSS ↓55%, BOD ↓98%, COD ↓ 98%, NH_4^+-N ↓86%, NO_3^--N ↓50%, TP ↓87%	[116]
Scirpus validus	TSS ↓80%, NH_4^+-N ↓95%, TKN ↓65.3%, COD ↓88.1%, TN ↓22.5%, TP ↓67.1%	[97]
Spirodela polyrhiza	COD ↓79.3–89.3%, TOC ↓85.3–91.3%	[88]
Iris pseudacorus	NH_4^+-N ↓71.9%, NO_3^--N ↓96.1%	[117]
Eichhornia crassipes + *Salvinia natans*	BOD ↓84.5%, COD ↓83.2%, TKN ↓53%, NO_3^--N ↓26.6%, PO_4^--P ↓56.6%	[118]
several species of plants	DO ↓60–70%, NO_3^--N ↓80%, pH ↓	[119]
several species of plants	DO ↓, pH ↓	[120]

↓—decrease, ↑—increase, DO—dissolved oxygen, COD—chemical oxygen demand, BOD—biological oxygen demand, DOC—dissolved organic carbon, TOC—total organic carbon, TN- total nitrogen, TP—total phosphorus, TKN—total Kjeldahl nitrogen, NH_4^+-N—ammonium nitrogen, NO_3^--N—nitrate nitrogen, PO_4^--P—phosphate phosphorus, TSS—total suspended solids, EC—electrical conductivity, S—salinity.

Conducted experiments usually lasted from 14 to 96 days. One parameter that was always seen to increases was temperature, while others including total suspended solids, electrical conductivity, salinity, chemical oxygen demand, biological oxygen demand, dissolved organic carbon, total organic carbon, as well as compounds containing nitrogen and phosphorus decreased. The maximum reduction efficiencies were 65% for dissolved oxygen by *Lemna minuta*, 95% for DOC by *Phalaris arundinacea*, 98% for BOD and COD by *Phragmites australis*, 91.3% for TOC by *Spirodela polyrhiza*, 90% TSS by *Bacopa monnieri* (L.) Pennell, and 95% for total Kjeldahl nitrogen by *Phalaris arundinacea*.

It has been found that the use of plants in wastewater treatment technologies improves the overall parameters related to pollution such as chemical oxygen demand or biochemical oxygen demand.

8. Conclusions

The presence of organic micropollutants originating from various fields of human activity in natural waters is one of the most important contemporary environmental problems. The main sources of these compounds are municipal and industrial wastewater. This work reviews research concerning the removal of organic micropollutants from wastewater through the use of phytoremediation. A number of plants, both rooted and floating, have been used to remove compounds belonging to various groups including pesticides, industrial contaminants, drugs, and personal care products. The research described in analyzed articles was carried out both in laboratories and under natural conditions with the use of matrices in the form of model solutions and wastewater. The effectiveness of removing micropollutants by plants ranges from a few to almost 100% depending on the species and structure of the compound, its concentration, and plant density. Studies show that removal efficiency increases along with contact time between the solution and the plants as well as with the weight of plants per unit volume. It is difficult to unequivocally assess the effect of compound concentration on the effectiveness of its removal since the results described in literature are, at times, contradictory. In all likelihood, many different aspects affect the experiments' final results. Although the contact of plants with most micropollutants causes their slower growth and other unfavorable symptoms, biochemical studies indicate that the activation of intense stress response allows plants to adapt to unfavorable conditions. The removal of micropollutants by plants is accompanied by the elimination of other contaminants present in the treated medium. Pollution indicators such as BOD_5, COD, nitrogen and phosphorus content, suspension content, and conductivity are significantly reduced. In the presence of plants, the pH of solutions increases or decreases toward values that are closer to neutral (pH = 7). On the basis of a review of the literature, it can be concluded that phytoremediation can be an effective method for the removal of micropollutants from polluted waters; however, the selection of appropriate process conditions is very important. On account of its specific needs including high levels of light and temperature, this method is particularly suited for countries located within relatively warm climates. The simplicity and the low cost of necessary installations make phytoremediation a particularly attractive solution for developing countries.

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