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Review

Wetland Removal Mechanisms for Emerging Contaminants

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Abstract: In recent decades, previously unobserved trace compounds have become more widely detected in wastewater treatment effluents and freshwater ecosystems. Emanating from various sources and presenting potential human health and ecological risks at much lesser concentrations than traditional contaminants, detection of "emerging contaminants" has increased with improvements in analytical techniques. The behavior of emerging contaminants in wetlands is a topic of increasing interest, as natural wetlands are known to transform and sequester pollutants and constructed or treatment wetlands are widely utilized to address elevated concentrations of constituents of concern. Both natural and constructed wetlands are complex biogeochemical systems with interrelated abiotic and biotic mechanisms leading to the removal of emerging contaminants. A literature review was performed to assess the current state of knowledge of various wetland mechanisms involved in removing these contaminants from surface waters and effluents. The primary mechanisms discussed in the literature are sorption, photodegradation, microbial biodegradation and phytoremediation. The most influential mechanisms are dependent on the properties of the contaminants and wetland systems studied. Common trends exist for different constructed wetland designs to leverage various mechanisms based on hydrology, substrate and vegetation plantings. Much remains to be understood about the various processes occurring in wetlands as they relate to emerging contaminant removal. Improving the understanding of the potential role of wetland mechanisms can help manage this environmental challenge more effectively.

Keywords: constructed wetlands; treatment wetlands; micropollutants; constituents of emerging concern; contaminants of emerging concern; sorption; photodegradation; phytoremediation; biodegradation

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1. Introduction

Many novel organic compounds have been detected more frequently in freshwater systems in recent years [1,2] and these emerging contaminants (ECs) are becoming an increasing global concern. ECs encompass a wide variety of human made chemicals with a range of uses, including industrial, agricultural and pharmaceutical and are proposed to potentially have detrimental impacts on human health and aquatic ecosystems [2,3]. Limited understanding of the impacts of ECs exists in part because they often occur at very low concentrations, with levels often in the ng/L range [4]. A vast number of these compounds and many transformation products exist. Because of the advanced analytical techniques required to detect occurrence at trace levels, regular testing for ECs can be prohibitively costly [5].

ECs enter aquatic ecosystems through agricultural runoff, industrial effluents [6] and, most notably, municipal wastewater treatment facilities [5]. Traditional wastewater treatment technologies are not designed to remove ECs [5,7] and therefore treatment of ECs within municipal wastewater treatment facilities is often incomplete or inadequate [8]. Advanced treatment technologies such as ozonation, membrane filtration and ultraviolet

radiation [2] have been found to be effective for the removal of some ECs [6], but they are energy intensive and expensive, making them impractical for many communities [9,10]. Therefore, interest in nature-based technologies has developed for more cost-effective and environmentally sustainable technologies for EC removal [2].

Wetlands, both natural and human-made, are able to assimilate and transform nutrients and other constituents through natural processes such as biodegradation, sorption, photodegradation and phytoremediation (Figure 1) [2,3,11]. These naturally occurring mechanisms have been demonstrated to be effective for removal of organic matter, biochemical oxygen demand, chemical oxygen demand, suspended solids, total nitrogen and total phosphorus, as well as organic xenobiotics such as ECs [6,7,12,13].

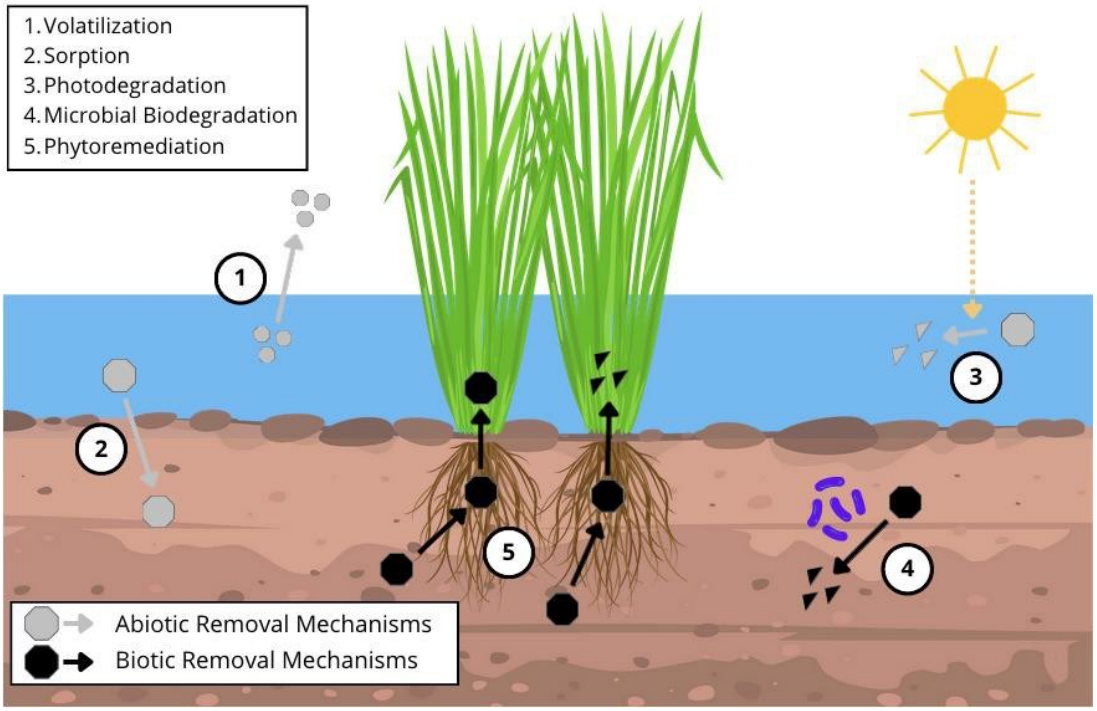

Figure 1. Wetland removal mechanisms of emerging contaminants. Octagons represent parent compounds, triangles represent transformation products. Abiotic removal mechanisms in grey, biotic removal mechanisms in black. Purple rods represent bacteria involved in biodegradation.

Constructed wetlands (CWs) or treatment wetlands are ecologically engineered ecosystems designed to utilize natural mechanisms to improve water quality through the removal of organic and inorganic pollutants from contaminated waters [14,15]. CWs are often classified by hydraulic design (e.g., free-water surface flow (FWS), horizontal subsurface flow (HF), or vertical subsurface flow (VF) [16]) and have been used to treat domestic wastewaters, industrial effluents, agricultural effluents and stormwaters, among others [14]. The degree of contaminant removal in wetland systems is dependent on a combination of conditions, such as age of the system [17,18], hydraulic design [2], vegetation [10] and the physicochemical characteristics of the contaminants [6,7].

Research on ECs in wetlands is often focused on the overall removal efficiency of selected compounds in an entire system [14,19–22] and does not address the complex and interrelated removal mechanisms involved. Understanding the relationships between removal mechanisms provides greater insight into how wetlands treat contaminants and therefore provides a wider and deeper knowledge base for design and maintenance strategies that can optimize EC removal in wetlands.

This review paper presents a summary of some of the current literature regarding wetland removal mechanisms for ECs and discusses some limitations of our current understanding. In this review, "removal" of contaminants is used as a general term for decreasing

the measured concentration of a parent compound from the water phase in the system. Although this term is nonspecific, it will encompass the transformation, degradation, or mineralization of contaminants in a system. The use of the term "removal" does not convey all facets of a compound's interaction within a system as it neglects transformation products and metabolites formed during degradation [1], but it will be used for this discussion as the majority of research found in the systematic review also used this terminology. The current knowledge around sorption, photodegradation, biodegradation and phytoremediation will be explored in depth within this review. It is not designed to be comprehensive, but the findings can provide context and background for the state of research around these topics.

2. Scope of Review

The primary eligibility criteria for studies to be included in this review were: (1) the study considered the fate of at least one EC in wetlands or (2) one or more wetland removal mechanisms for ECs were studied or hypothesized to be active. The primary literature search was conducted using the SCOPUS database. The search string that was used was "TITLE-ABS-KEY ((wetland) AND ("contaminants of emerging concern" OR cec OR "compounds of emerging concern" OR "chemicals of emerging concern" OR "emerging contaminants" OR micropollutants))". The search was further limited to articles available in English. The search, conducted on 10 June 2022, resulted in 312 documents. Sixty-one (61) more research articles were added to our database that were not found in the SCOPUS search but which were found to be relevant to our search criteria. Since this search was limited to English articles only, the final search results might exclude relevant studies that were not available in English. Additionally, the keywords used to limit our search criteria might not represent all relevant papers. Although the search was conducted systematically, the search result may still be biased in some respects.

Once the initial database was developed, a systematic table was created to list all the relevant information for the research. Each study was reviewed by at least one author, who entered relevant information to the database. Articles that did not include the specific criteria that were required were excluded from the search. A total of 180 articles were included [2,3,6,8–15,17–185], of which 125 were primary research. The additional 61 research articles found outside of the initial search were not included in the database of primary research and were not used in calculations or statistics for the final database.

3. Emerging Contaminants

Hundreds of thousands of different types of human-made organic chemicals are found in the environment and, therefore, are difficult to quantify [186]. Many of them pose potential risks to human and ecological health [4–6,23,187,188]. Advances in analytical techniques combined with the increased production and use of anthropogenic compounds have led to the increased environmental detection of many substances in the last few decades, sometimes at extremely low concentrations [2,6,186,189,190]. In addition, the varied nomenclature for these compounds (e.g., emerging contaminant; contaminant, chemical or constituent of emerging concern; micropollutant) can also increase the difficulty of communication between researchers and the public.

The increasingly widespread detection of ECs and their potential toxicity, even at low concentrations, has led to them being targets of growing scrutiny [4,191]. Although much of the research on this subject is focused on pharmaceutical compounds, there are a wide range of compounds considered to be ECs, including, but not limited to, plasticizers, disinfectants, flame retardants, herbicides, insecticides, corrosion inhibitors and synthetic dyes [4,24,25,192]. Traditional wastewater treatment processes such as activated sludge can effectively address many ECs but are less effective for others [26], making wastewater discharges widespread point sources of ECs into freshwater ecosystems [24,25,27]. Stormwater runoff is also a common source of ECs, especially for agricultural chemicals such as pesticides and antibiotics [28,29,193]. These point and nonpoint source releases lead to the transport of ECs into freshwater ecosystems, including wetlands [186,194]. In

addition to the natural transport of ECs to wetlands, engineered wetlands are frequently utilized to treat wastewater effluent and manage stormwater runoff [7,17,30,31,193,195]. Therefore, it is critical to understand wetland EC interactions.

There appears to be no consensus for the nomenclature of these compounds and a number of different terms are used interchangeably to refer to them [32]. In the 180 articles selected from the literature search used for this review, at least 15 terms were used for these types of compounds, six of which occurred five or more times (Table 1). "Micropollutant" (MP) was the most common term, appearing in 70 articles, "emerging contaminant" (EC) was second at 68, followed by "contaminant of emerging concern" (CEC) at 26. MP has also been used to refer to inorganic pollutants such as trace metals, in addition to organic MPs, potentially decreasing the specificity of this term. The use of the acronym CEC can create ambiguity for multiple reasons, including that the first "C" in "CEC" can have different meanings and may refer to chemicals, compounds, or constituents of emerging concern. In addition, a notable number of articles found from using "CEC" as a search term for this review were unrelated to the topic of organic compounds as pollutants, but rather appeared in the search because they focused on "cation exchange capacity", which uses the same acronym and is an important parameter in wetland science. These examples highlight the challenge of choosing a definitive term for labeling these compounds that avoids potential ambiguity. For this review, EC has been chosen to refer to these compounds because this term is frequently used in the literature and may be less ambiguous than other options.

Table 1. Most frequently used terms to describe emerging contaminants from the literature search (n = 180). Articles that referred to multiple terms or used terms interchangeably were recorded as using multiple listed terms.

Term	# of Studies
Micropollutant (MP)	70
Emerging Contaminant (EC)	68
Contaminants of Emerging Concern (CEC)	26
Organic Micropollutant	21
Emerging Pollutant	15
Emerging Organic Contaminant	9

In addition to a lack of standardization around nomenclature, there is also no consensus on how to categorize these compounds [21], though they are often delineated into three main categories based on their human use: Pharmaceuticals and personal care products (PPCPs), pesticides and industrials [4]. ECs are also characterized based on toxicological endpoints, such as being an endocrine disrupting chemical (EDC) or carcinogen [33,196] or by their chemical structure, such as polycyclic aromatic hydrocarbons (PAHs), perfluorinated and polyfluorinated substances (e.g., PFASs, PFOS, PFOA) and polybrominated diphenyl ethers (PBDEs) [4,197].

For the articles selected from the literature search, the most commonly targeted ECs in primary research on wetland removal were carbamazepine, diclofenac, sulfamethoxazole and ibuprofen (Table 2). The six most studied compounds were all PPCPs and of the 22 compounds appearing in 10 or more studies, 19 were PPCPs with two industrial chemicals and one pesticide.

It is important to note that just because certain ECs are more commonly studied does not mean they necessarily pose the greatest toxicological risks or are the most abundant in municipal wastewater or freshwater ecosystems [1,4]. There is selection bias towards studying compounds based on the previous body of research, well established analytical techniques, or lower financial costs for analysis. Yang et al. [186] proposed a "weighted average risk quotient" (WARQ) metric to assess the potential risks of ECs on a global scale by averaging existing risk quotient values from the literature. The relative abundance of ECs studied in this literature review show some similarities with the results of the WARQ, as a majority of the 53 priority compounds and the ten highest ranked compounds were

PPCPs. Only two of the most common compounds from this literature review (metoprolol and clofibric acid) were not on the WARQ priority list and all but one of the top ten compounds on the priority list (ofloxacin) appeared in ten or more articles from the review (Table 2).

Table 2. Most studied emerging contaminants in primary research from literature review.

Compound	CAS	Class	Subclass	# Studies	WARQ Rank from Yang et al. [186]
Carbamazepine	298-46-4	PPCP	Anticonvulsant	58	10
Diclofenac	15307-86-5	PPCP	Anti-inflammatory	57	2
Sulfamethoxazole	144930-01-8	PPCP	Antibiotic	42	1
Ibuprofen	15687-27-1	PPCP	Anti-inflammatory	36	7
Caffeine	58-08-2	PPCP	Stimulant	31	4
Naproxen	22204-53-1	PPCP	Anti-inflammatory	31	26
Benzotriazole	273-02-9	Industrial	Corrosion Inhibitor	27	46
Bisphenol A	80-05-7	Industrial	Plasticizer	23	25
Metoprolol	37350-58-6	PPCP	Beta-blocker	22	-
Trimethoprim	738-70-5	PPCP	Antibiotic	19	18
Atenolol	29122-68-7	PPCP	Beta-blocker	18	52
Ketoprofen	22071-15-4	PPCP	Anti-inflammatory	18	40
Triclosan	3380-34-5	PPCP	Disinfectant	16	6
Propranolol	525-66-6	PPCP	Beta-blocker	15	27
Acetaminophen	103-90-2	PPCP	Anti-inflammatory	12	3
Atrazine	1912-24-9	Pesticide	Herbicide	12	31
Clarithromycin	81103-11-9	PPCP	Antibiotic	12	9
Erythromycin	114-07-8	PPCP	Antibiotic	12	8
Gemfibrozil	25812-30-0	PPCP	Antihyperlipidemic	11	23
Ethynylestradiol	57-63-6	PPCP	Contraceptive	10	20
Clofibric acid	882-0907	PPCP	Blood lipid regulator	10	-
Galaxolide	1222-05-5	PPCP	Synthetic Musk	10	32

The study of ECs in freshwater environments is a complex topic for a variety of reasons. It is unclear if concentrations measured in natural or treated waters actually pose health risks [34,196,198] and this uncertainty is exacerbated by the fact that they generally occur in complex mixtures with other organic and inorganic contaminants [5,188,194]. ECs also have a wide range of physical properties including polarity, molecular weight, solubility and partitioning coefficients, which influence their environmental recalcitrance, making it difficult to generalize about removal mechanisms [5,25,35,36]. Therefore, the following discussion provides a conceptual framework of wetland processes that may be important in mitigating ECs. It includes examples of certain compounds that are susceptible to these mechanisms and relates this susceptibility to the properties of these compounds.

4. Abiotic Removal Mechanisms of ECs in Wetlands

Multiple abiotic removal mechanisms occur within wetlands that contribute to EC removal (Table 3). Sorption and photodegradation are discussed relatively often in published research, while few studies found in the systematic review process mention volatilization or hydrolysis, other than to state that they may play a role in removal. Sorption and photodegradation will be discussed in further detail in their respective sections, but volatilization and hydrolysis will be briefly discussed here.

Table 3. Abiotic emerging contaminant removal mechanisms in wetlands.

Abiotic Mechanism	Mechanism Description	Reference
Sorption	Binding or transfer of compounds from the bulk liquid phase to a physical surface; includes both adsorption and absorption	[199]
Photodegradation	Degradation of compounds via reactions caused directly by light or mediated by light	[2]
Volatilization	Transfer of compounds to the external atmosphere in a gaseous form	[200]
Hydrolysis	Breakdown of an organic compound into two or more new compounds through interaction with water molecules	[201]

Volatilization occurs when an organic compound is transformed from the liquid phase into a gaseous phase [200,202]. The volatilization of a compound is related to the compound's vapor pressure with higher vapor pressures experiencing increased volatilization [6]. Wetland hydraulic design (e.g., surface flow or subsurface flow) and treatment conditions like aeration, agitation and temperature also impact volatilization in wetlands [18,202].

Hydrolysis is the cleavage of an organic compound through interaction with water molecules [201]. The hydrolysis rate constant, K_{hydro}, for a given compound can be calculated to understand the rate of hydrolysis within a system [37]. The impact of hydrolysis is often found to contribute little to the overall removal of ECs in wetlands [37] and can even be negated in certain systems [38,39]. In general, compounds that are known to be stable, such as sulfonamide antibiotics, experience limited removal by hydrolysis or volatilization [19].

4.1. Sorption

Sorption is an abiotic process that naturally occurs due to either electrostatic interactions between compounds and substrate particles, referred to as adsorption, or through hydrophobic interactions between the lipophilic cell membrane of a microorganism or the lipid fraction of a solid, referred to as absorption [15,40]. ECs of various chemical structures and compositions will interact differently within the wetland substrate and may experience sorption at different rates.

Environmental factors within the system further complicate the modeling of sorption, as specific functional groups of an EC interact differently with substrate media and may experience complex pH-dependent speciation [203]. Sorption is frequently modeled through linear, Langmuir and Freundlich isotherm modelling [41]. The models produce sorption coefficients that can be used to understand sorption capacity and efficiency for comparison to other compounds. The Langmuir model uses the Langmuir parameter, K_L and the maximum adsorption capacity for a substrate, Q_M [42]. The Freundlich model produces the Freundlich distribution constant, K_f and the Freundlich exponent, n [203]. These modeling coefficients can be used to compare adsorption rates across multiple experiments and substrates.

As sorption and desorption are used to predict the mobility of contaminants within a wetland, it is necessary to understand what physicochemical interactions impact sorption for modeling long-term water quality [203–206]. Environmental conditions within the wetland, the physicochemical structure of the compound for removal and the physicochemical characteristics of the substrate all impact sorption within a system [8,20,203,206].

4.1.1. Sorption Systematic Review Summary

EC removal via sorption was discussed in 54% (68 of 125 articles) of the primary research articles found in the literature review, making it the second most discussed removal mechanism. Of the 68 primary research articles that discussed sorption, 49 were field experiments and 19 were laboratory or greenhouse experiments. Thirty-three of the primary

research articles studied mesocosm or microcosm systems, 31 studied full-scale CWs and 5 studied natural wetland systems. Research on sorption of ECs in wetlands focused broadly on the physicochemical parameters of ECs that affect sorption, the physicochemical conditions of sediment and soil mixes used in CWs and their effect on sorption, the interaction between organic matter in sediment and EC removal and changes to EC sorption due to seasonal or operational changes in a system. The large majority of sorption-centered research found in the systematic review was conducted through batch sorption experiments of wetland substrates.

4.1.2. Partitioning Coefficients and Hydrophobicity

The degree of sorption within a system is due to multiple factors related to the contaminant, substrate and environmental conditions. Much research has been conducted to understand what parameters are most indicative of EC removal by sorption within a system. Although no single chemical parameter has been determined to fully predict the efficacy of sorption, many researchers utilize various physiochemical properties of a compound, such as molecular weight, ionic strength and partitioning coefficients to predict sorption rates within a given system [6,42].

The sorption potential is often characterized by a compound's partitioning coefficients, which represent interactions with a variety of removal mechanisms within a system. A common partitioning coefficient for sorption is the distribution coefficient, K_d, which approximates the ratio of the contaminant in the sediment phase to the water phase [6] and is a key component to a contaminant's mobility in a system. The dissociation constant, pK_a, of the target compound is commonly used to characterize the potential for adsorption in wetlands [15]. The octanol-water coefficient, K_{OW}, represents the hydrophobicity of a compound and is commonly used to predict and understand the strength of absorption within a system [6,15]. Compounds with a higher K_{OW} are more hydrophobic and correspondingly have a greater degree of removal by sorption. Research conducted by Ren et al. [43] and Ravichandran et al. [44] on vertical flow wetlands found similar results with the greatest removal by adsorption seen for the most hydrophobic compounds. Ravichandran et al. [44] suggested that the hydrophobicity and elevated influent concentrations of the hydrophobic compounds were both influential in the sorption of the ECs within the studied system.

The relationship between sorption and compound hydrophobicity cannot explain all facets of sorption in a system [40,44]. Specific interactions between functional groups of the sorbent (the substrate being sorbed to) and the sorbate (the compound being sorbed) can affect the strength of sorption in a system beyond compound hydrophobicity [12]. He et al. [34] conducted a study on the attenuation of pharmaceutically active compounds in CWs and found no correlation between overall removal of these ECs and pK_a and K_{OW}, suggesting that no single parameter can determine the degree of removal in a system and that multiple removal mechanisms were likely to influence removal.

Other removal mechanisms in a wetland can also complicate the correlation between hydrophobicity and sorption. Brunsch et al. [45] did not find a significant relationship between the hydrophobicity or ionic charge of neutral and positive charged compounds, respectively, and their removals in a vertical flow CW. These results were thought to indicate that other removal mechanisms such as plant uptake and biodegradation were impacting removal efficiency [45]. Ravichandran et al. [44] found that the sorption of ECs to wetland substrates was lower in field conditions than in laboratory adsorption batch experiments. This result was attributed to the influence of biodegradation and phytoremediation in the wetland decreasing the concentration of contaminants in the bulk liquid and therefore decreasing the amount of ECs available in solution to sorb to the substrate [44].

4.1.3. Surface Charge and pH

The surface charge interaction between ECs and substrates is one of two major interactions governing sorption in CWs. Research has found that sorption affinity to soils is greatest for positively charged compounds, followed by neutrally charged compounds and

least for negatively charged polar compounds [203]. ECs that are positively charged tend to experience greater rates of removal by adsorption to negatively charged substrate media such as organic matter or clay, while neutrally charged ECs are sorbed to wetland substrate through weaker Van der Waal's forces of attraction [2]. Research on the impact of ionization on sorption in batch experiments with sediment collected from the Madrid Detrital Tertiary Aquifer in Spain found that cationic ECs, such as atenolol and caffeine, experienced greater rates of removal through sorption to the largely negative surface charges of the sediment than neutral or negatively charged compounds [204]. Over 70% of the total PPCP sorption was due to these electrostatic interactions between the inorganic mineral surface of the sediments, demonstrating the impact of substrate charge to EC sorption [204].

Neutrally charged compounds such as trimethoprim have been shown to experience greater sorption to substrate media than negatively charged compounds such as sulfamethoxazole [41]. However, negatively charged contaminants can still sorb to negatively charged substrate active sites despite electrostatic repulsion, likely due to strong hydrophobic interactions [41]. These hydrophobic interactions allowed for a slow mass transfer of the contaminant, but in turn, required longer contact times for adequate sorption.

The pH of a system impacts EC sorption by altering the electrostatic charge of both the compound and substrate. The electrostatic charge of a compound is based on its pK_a value compared to the overall pH of the system [46]. Compounds with an acidic pK_a would be largely present in their anionic forms at circumneutral pH, inducing electrostatic repulsion between the anionic compound and the negatively charged sorption sites of the substrate [41]. Substrate surface charge is dependent on the pH of the system and the point of zero charge (PZC) of that substrate. The PZC is the pH condition where the anionic and cationic sites of the substrate are equal, resulting in an overall neutral surface charge of the substrate [204]. Wetland pH values over the substrate PZC induce a negative surface charge and pH values under the substrate PZC induce a positive surface charge [204].

Research has found that sorption capacity is often negatively correlated with system pH, as substrates, such as wood fiber and coconut fiber, which are negatively charged in circumneutral pH conditions have been found to have poor sorption affinity for negatively charged compounds such as naproxen (pK_a = 4.15) and ibuprofen (pK_a = 4.91) [41]. It was hypothesized that this is a result of the strong electrostatic repulsion between the substrate and compound overpowering other forces of attraction within the system [41].

Other substrate characteristics such as surface charge density and cation exchange capacity can also influence sorption [203,204,206,207]. Experiments examining the adsorption of sulfamethazine, sulfamethoxazole and sulfa-pyridine to montmorillonite and kaolinite clays showed that the surface charge density of montmorillonite influenced the sorption of sulfamethazine, with the sorption of sulfamethazine being highly sensitive to a change in surface charge density [207].

4.1.4. Chemical Structure

Compound or chemical class specific structures have also been found to impact adsorption. This result has been well documented in perfluoroalkyl and polyfluoroalkyl substances (PFASs) [11,47,205]. Various long-chain PFASs such as PFOA and PFAA experience greater removal by sorption to suspended particles in sedimentation tanks in a hybrid CW system than short-chain PFASs [11,47,205]. Long-chain PFAS are more hydrophobic and therefore experience a greater degree of removal by sorption than the more hydrophilic short-carbon chain compounds [11]. Chain length also increases the retention of PFAS compounds to the air-water interface in unsaturated soils, aiding in removal [47].

The molecular size of an EC is important to sorption [8,42]. Compounds with higher molecular weights have been found to experience greater removal in wetlands than compounds with lower molecular weights and sorption is hypothesized to be involved in this trend [20]. Gao and Pedersen [207] saw a general trend of increasing adsorption coefficients with increasing molecular surface area when examining the adsorption of sulfonamide antimicrobials to clay minerals. However, a lower molecular weight might enable better

mass transfer into pores on a substrate [41]. This contrast between the optimal compound size for sorption could be due to differences between the ideal parameters for adsorption and absorption in a system.

4.1.5. Substrate Size and Structure

Different sediment or substrate properties influence sorption (Table 4). The natural sorption capacity and permeability of a substrate is essential to increasing the total removal of ECs by sorption while also allowing adequate flow of water through the system [6,48]. Lower porosity substrates are thought to have relatively larger surface areas and a greater number of adsorption sites than similar substrates with higher porosity [63].

Table 4. Parameters of wetland substrates that impact the sorption capacity and system flow.

Wetland Substrate	Parameters Impacting Sorption Capacity	Advantages	Disadvantages
Gravel/Sand	Particle size [208]	Low cost [209] Widely available [209,210] Non-polluting [209]	Relatively small surface area [48] Issues with clogging [48]
Natural sediments	Charge of mineral surface (related to system pH and PZC) [49,204] Organic carbon content [41,49] Clay content [203,207] Cation exchange capacity [49,203,204]	Low cost [209] Widely available [209]	Less control of internal structure Location/source dependent
Volcanic rock	Particle size [50]	Relatively large porosity as compared to gravel [50] High hydraulic conductivity [209]	Availability depends on region Poor sorption capacity compared to other substrate classes [48]
Biochar	Thermal treatment conditions [211] Feedstock characteristics [211]	Well-developed pore structure [42,208] Large specific surface area [42]	Requires regeneration or replacement to restore sorption capacity once saturated [208] Energy intensive pyrolysis process [212]
Light Expanded Clay Aggregates (LECA)	Particle size [210] Use of coating or additive to enhance sorption capacity [210] Thermal treatment conditions [210]	High porosity and large specific surface area [210] Large cation exchange capacity [210] Ability to modify LECA using additives to meet needs of the system [210]	Requires surface enhancement as naturally chemically inert [210,213] Energy intensive manufacturing [210]
Activated Carbon	Contact time [214] Particle size [199] Manufacturing conditions like type of activating agent and temperature and duration of activation [214]	Large specific surface area [48,209] Large micropore structure [48]	Must be removed and replaced when sorption capacity exhausted [214] Limited ability to regenerate or reactivate [214]

Traditional CW substrates include gravel, sand and local sediments, but new substrate designs include additions of biochar, activated carbon and Light Expanded Clay Aggregates (LECA) to traditional soil support matrices to improve sorption capacity [48]. Some research has been conducted into more unique or local substrate mixes and properties. For tezontle, a volcanic rock substrate commonly used in CWs in Mexico, particle size of the substrate was the only significant ($p < 0.05$) factor related to the sorption of carbamazepine [50]. The smaller grained substrate was associated with greater removal efficiencies of the compound regardless of temperature and pH of the system, likely due to smaller particle sizes providing greater surface areas for sorbate/sorbent interactions [50].

Lei et al. [48] completed batch experiments with both traditional substrates (sand and gravel) and more experimental substrate mixes (LECA, bark, compost, lava rock, granular activated carbon (GAC) biochar and granulated cork). GAC and biochar are designed for high surface area and a high micropore structure and both substrates achieved nearly 100% removal of the 11 ECs tested at influent concentrations of 5 µg/L. Sand and gravel saw much lower removal efficiencies at this same concentration which could be due to their smaller surface area [48]. Compost and biochar amendments to soil have been associated with increases in the partitioning coefficient K_D and the isotherm modeling coefficients K_L, K_F and Q_M with sulfamethoxazole [42]. This improvement in sorption affinity was hypothesized to be a result of complexing action from organic functional groups on the surface of the compost.

4.1.6. Influence of Organic Matter

The organic matter content of a substrate is highly influential for compound removal by sorption [41]. Organic matter can be represented by measures such as total organic carbon, soil organic matter, or the percentage of organic matter [41,49,206]. Soil organic matter is made of biomolecules and organic debris such as extracellular polysaccharides and cell walls from the decomposition of organisms [206]. Organic matter increases the sorption capacity of a system through interactions with the decomposed and largely hydrophobic organic matter [206].

Sithamparanathan et al. [41] found a positive correlation to EC sorption and the organic matter percentage of the substrate regardless of the compound hydrophobicity in a study of the hydroponic substrate of wood fiber, coconut fiber, mineral wood and pumice. The upper layers of different CW substrates have been observed to have greater organic matter and EC removal than those deeper in the substrate [45,51]. This effect was suggested to be caused by a combination of organic matter and small particle size providing ample sorption capacity and substrate for microbial degradation of compounds. In these settings there is no clear delineation between removal by sorption and removal by biodegradation, demonstrating the integrated nature of wetland removal mechanisms.

Xu et al. [49] found a significantly positive relationship between the sorption of the pesticides atrazine and trifluralin and increasing total organic carbon. The sorption coefficient, K_d, was significantly greater in natural prairie wetland sediments with larger total organic carbon content for these two compounds. Other compounds in this study were less impacted by the organic matter content of the substrate or were influenced by other sediment properties. In addition, 2,4-D sorption was found to be positively linked to increasing total organic carbon and the total cation exchange capacity within the substrate, although there was no statistically significant relationship found between total organic carbon and the sorption of glyphosate glycine [49].

4.1.7. Adsorption Saturation

The adsorption capacity of substrates can become exhausted from long periods of exposure or from elevated concentrations of compounds for removal [6]. Sorption batch experiments have shown decreasing removal efficiency with increasing influent EC concentrations, as elevated concentrations likely caused adsorption saturation while lesser concentrations did not [48]. The PFAAs' adsorption capacity of sediment was also shown to be concentration-dependent [52].

Time of operation can impact the sorption capacity of a system. Research on the changes in the removal efficiencies of ECs from a VFCW after 7 and 10 years of operation found that there was a decrease in the removal efficiency of compounds resistant to biodegradation such as carbamazepine, sulfamethoxazole, 1-h-benzotriazole, EDTA and TCPP regardless of inflow concentration [17]. The removal decrease was hypothesized to be due to an exhausted adsorption capacity of substrates in the system indicating that after an extended duration in operation, system maintenance like exchanging CW filter media is needed to maintain adequate adsorption capacity in the system [17]. Ruppelt et al. [18]

found a similar relationship of decreasing removal efficiency of diclofenac and metoprolol in a pilot-scale CW over time, suggesting that the sorption capacity of the media was exhausted during the study [18]. However, this correlation did not consider environmental conditions such as rainfall during the study that would cause corresponding changes in the concentrations of contaminants by other means. As many CWs are designed for long design lives of over 20 years, more research into substrate sorption capacity over time is necessary to provide guidance for best management practices of CW in operation.

4.1.8. Desorption

Desorption occurs when a previously sorbed compound is released back into solution. It is impacted by changing environmental conditions, the strength of the initial adsorption, the saturation of adsorption sites on a substrate and competition from inorganic compounds for sorption sites [42,204,205]. CWs are ideally designed to retain and degrade contaminants, but this situation may not always be realized.

The equilibrium relationship between sorption and desorption can sometimes cause negative removal efficiencies through the release of previously held contaminants into solution due to changes in system pH or oxidation reduction potential (ORP) [12]. The addition of fresh electrolyte solution into sediment with specific PFAS compounds increased desorption of these contaminants from the solid phase to the liquid phase due to competition between the stronger solvent (the fresh electrolyte) and the initial solution [205]. This result implies that a change in water chemistry in a system, such as the introduction of reclaimed wastewater, could induce a shift in the sorption/desorption equilibrium through competition with inorganic ions and other compounds in the influent [204]. Additionally, a drastic change in the pH of a system could shift the sorption/desorption equilibrium of a system by altering the surface charge of a contaminant and substrate.

Some research has suggested that cationic compounds that are easily sorbed in a CW are more likely to be desorbed with competition from inorganic compounds or changing environmental conditions. The cationic compounds, atenolol and caffeine, demonstrated the greatest rates of sorption and desorption in a set of batch experiments [204]. Venditti et al. [15] found a similar relationship with the elevated sorption rate for the pesticide glyphosate in a pilot CW system, and its associated rapid desorption after competition with inorganic phosphates, from previous research studies.

This relationship between a strong sorption capacity and greater desorption does not hold true across all ECs and all studies. Long-chain PFAS, which experience strong sorption to suspended solids and sediments, have been found to have a higher degree of irreversibility in sorption when compared to short-chain PFAS compounds with a lower sorption capacity and a higher degree of desorption [205]. Research into the sorption/desorption equilibrium of contaminants needs to be conducted on a compound specific basis as changing environmental conditions and the physicochemical characteristics of a compound alter its degree of sorption/desorption within a system.

Where and how a compound was initially sorbed also impacts desorption. Sulfamethoxazole has been shown to experience greater desorption from compost-amended substrates than biochar substrates, likely due to sulfamethoxazole being largely adsorbed onto the surface of the compost-amended substrate but absorbed into the internal pores of the biochar system [42].

4.1.9. Sorption Summary

The sorption of ECs in wetlands is dependent on a variety of parameters that govern the hydrophobic and electrostatic interactions of sorption (Figure 2) [8,49,206]. Most notably, compound hydrophobicity, surface charge and environmental pH, chemical structure, substrate adsorption capacity, substrate organic matter content and time of operation of a system impact sorption [11,17,41,43,49,50,205]. Although it is important to understand how each of these individual parameters function in sorption, perhaps more important is the understanding of how these parameters compete and interact with each other in a complex

and dynamic system. More research is needed to further the knowledge of sorption, and correspondingly desorption, in the long-term operation of wetland systems and to further characterize sorption interactions with other removal mechanisms in wetlands. Research into the sorption/desorption equilibrium of contaminants needs to be conducted on a compound specific basis as changing environmental conditions and the physicochemical characteristics of a compound alter its degree of sorption/desorption within a system.

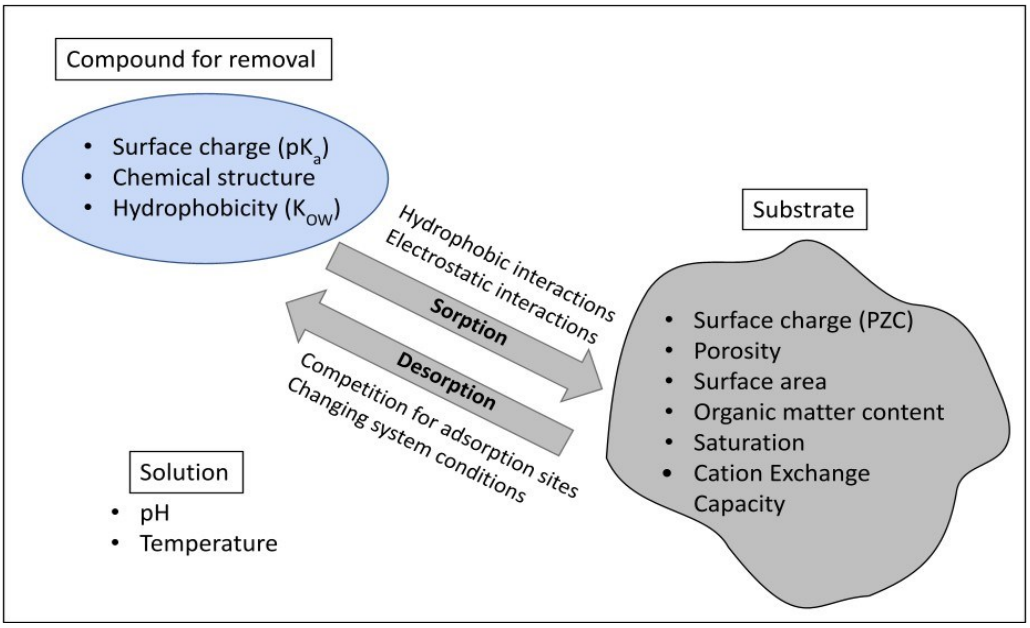

Figure 2. Influencing parameters in the sorption of emerging contaminants to substrate in constructed wetlands. These parameters are not listed in order of relevance or influence in a system.

4.2. *Photodegradation*

Photodegradation refers to the degradation of contaminants by solar radiation and can occur via direct or indirect photolysis (Figure 3). Direct photolysis happens through the absorption of ultraviolet rays by the contaminant, breaking them down into potentially less harmful forms. In indirect photolysis, energy from light is absorbed by photosensitizers such as nitrate, nitrite and dissolved organic matter (DOM), generating reactive species that then degrade contaminants [30]. The efficiency of photodegradation as a removal mechanism in of ECs in wetlands depends on the structure of the molecule, environmental conditions and the physicochemical composition of the water for both direct and indirect photodegradation [30].

4.2.1. Photodegradation Systematic Review Summary

Photodegradation was discussed in 25% of the research articles from the literature review (31 out of 125 articles), making it the fourth most discussed removal mechanism. Of the 31 research articles that addressed photodegradation, 13 were performed in laboratory or greenhouse settings and 18 were performed in outdoor field settings. Additionally, 16 of these studies were performed in microcosms or mesocosms and 15 discussed operational CW systems. The reviewed studies on photodegradation often focused on it coupled with other mechanisms and examined kinetic constants, half-lives and different light sources, among other variables related to EC removal.

4.2.2. Photodegradation Studied on Its Own

Of the 31 studies of photodegradation, only four focused solely on this removal mechanism [30,53–55]. Lee et al. [53] investigated the photodegradation of PPCPs and their metabolites within different natural organic matter (NOM) enriched solutions in aquatic environments, comparing photodegradation efficiency between indirect and direct

photolysis. Mathon et al. [30] studied the photodegradation of 23 organic contaminants in CWs with in situ photoreactors at different depths of water, investigating the contribution of both direct and indirect photodegradation in the removal of ECs. Mathon et al. [54] tested direct photodegradation on 36 ECs under simulated solar radiation evaluating their half-lives and suggesting chemical characteristics of contaminants that are sensitive to photodegradation. Felis et al. [55] performed laboratory-scale experiments with artificial sunlight and found removal efficiencies of 42% for benzotriazole and 58% for benzothiazole.

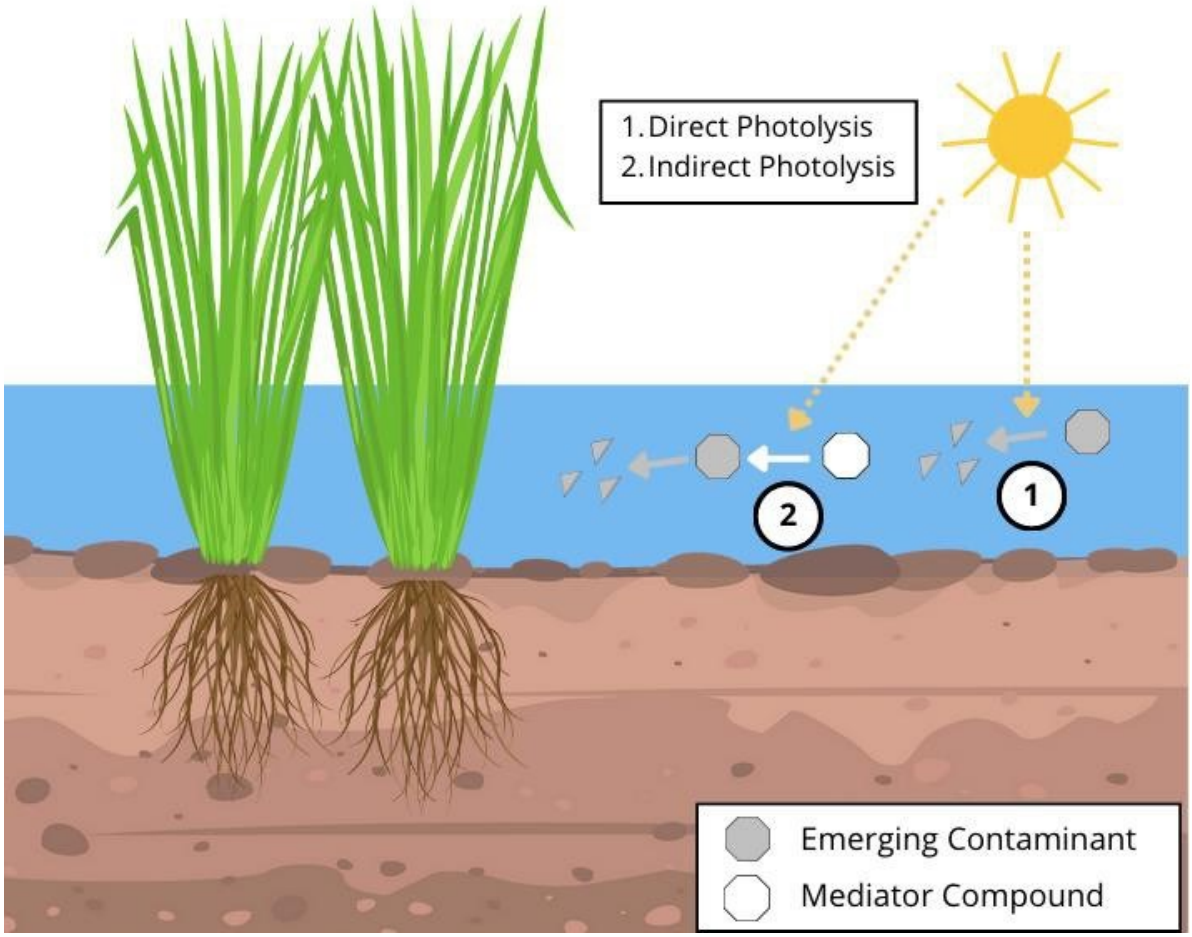

Figure 3. Photodegradation of emerging contaminants in wetlands. Photodegradation occurs through direct photolysis of the compound, or is mediated by another compound, shown in white, through indirect photolysis. Octagons represent parent compounds; triangles represent transformation products.

4.2.3. Photodegradation Studied with Other Mechanisms

A larger number of studies looked at photodegradation in tandem with other removal mechanisms. A study on the removal of atenolol, carbamazepine and diclofenac by *Canna indica* and *Chrysopogon zizanioides* under hydroponic conditions focused on plant uptake, but also explored the effects of photodegradation by utilizing unplanted control mesocosms where photodegradation was assumed to be the primary removal mechanism in the surface water [56]. It was found that 11–12% of atenolol, 8–9% of carbamazepine and 22–23% of diclofenac mass underwent photodegradation [56]. Several studies used methods like mass balance models to assess the relative contributions of multiple removal mechanisms like hydrolysis, plant uptake, sorption and biodegradation alongside photodegradation and often found it to not be a major removal mechanism [37,57]. Another study on EC removal efficiency in a full-scale CW assessing biodegradation, sorption, volatilization, hydrolysis and photodegradation found greater than 80% removal for all compounds photodegradation playing an important role for certain compounds [215].

4.2.4. Half Lives and Kinetic Constants

Several studies focused on photodegradation kinetic constants and the resulting half-lives of ECs [37,54,57]. Batch experiments found kinetic constants ranging from 0.001 to 0.045 for benzotriazoles [37] and 0.005 to 0.055 for a variety of ECs, including cefadroxil, metronidazole, trimethoprim and sulfamethoxazole [57]. Mathon et al. [54] looked at the influence of chemical structure on the photodegradation half-lives for 36 ECs with a large range of physicochemical properties and classified them into seven classes based on their results. This study showed that the micropollutant functional groups OH–C=O, C=N–O, =N–OH, –CH=N–, –O–P=O and –C=C– were more sensitive to photodegradation, while –O– and –Cl had a lower sensitivity [54].

4.2.5. Photodegradation Light Source

The light source used for photodegradation can play a role in its efficacy [53–55,215]. While most laboratory photodegradation studies use artificial light exposure that differs significantly from solar radiation, artificial light has also been designed to mimic natural sunlight and better approximate outdoor wetland conditions [54]. Experiments using light sources more similar to sunlight intensity and wavelength ranges, circumneutral pH and no organic solvents in solution, may better represent natural photodegradation [54]. A study with artificial sunlight looked at titanium dioxide as a photosensitizer and found the kinetic constants were two orders of magnitude higher with indirect photolysis utilizing titanium dioxide than from the direct photolysis of the contaminants caused by artificial sunlight [55]. Direct and indirect photolysis of ECs has also been compared with UV radiation, testing natural organic matter as a photosensitizer [53]. In operational CWs photodegradation through direct sunlight exposure has been shown to play a large role in contaminant removal [215]. The emission spectrum of light sources, natural and artificial, impacts the efficiency of photodegradation and is an important component of studies on this mechanism.

4.2.6. Other Influences on Photodegradation

Lee et al. [53] explored the effects of using NOM enriched solutions and DOM interaction with ECs in wetlands when photodegradation is involved. Allochthonous NOM inhibited photolysis of some PPCPs, wetland NOM increased the photodegradation of some recalcitrant PPCPs and photolysis increased degradation of some contaminants through interaction with DOM in wetlands. Additionally, benzotriazole and benzothiazole have been shown to not be susceptible to photodegradation in the absence of titanium dioxide, but were removed by 42% and 58%, respectively, with titanium dioxide present [55]. Different organic and inorganic materials in the wetland water column may therefore play a role in EC photodegradation, potentially as photosensitizers facilitating indirect photolysis. Another study on the influence of water depth and season on photodegradation of ECs in CWs showed that photodegradation was most effective in the top 10 cm of the water column and no significant difference was apparent between seasons [30].

4.2.7. Photodegradation Summary

Photodegradation is potentially an important removal mechanism of ECs in wetlands, especially in settings with more open water and less vegetation, allowing more sunlight penetration in the water column. Most of the research on photodegradation in this review looked at it in conjunction with other mechanisms, with much less research focusing on it as an isolated mechanism. Studies on EC photodegradation in wetlands commonly addressed relationships with other removal mechanisms, half-lives and kinetic constants, photosensitizers for indirect photolysis and light sources More research on photodegradation on a field-scale natural wetlands and CWs with natural solar radiation will be beneficial to the better understanding the importance and applications of the mechanism.

5. Biotic Removal Mechanisms of ECs in Wetlands

A variety of biotic EC removal mechanisms occur in wetlands (Table 5), which are often interrelated with each other as well as with abiotic mechanisms. Biodegradation, mostly performed by microorganisms, is often the greatest contributor to EC removal [58]. However, various plant-based (phytoremediation) processes like phytoextraction and phytodegradation also play prominent roles and the presence of plants and their influences on the rhizosphere shape the microbial community present [59]. Other organisms such as algae and filter feeding animals can also influence EC fate in wetlands, although research on their role in CWs is limited.

Table 5. Biotic emerging contaminant removal mechanisms in wetlands.

Biotic Mechanism	Mechanism Description	Reference
Biodegradation	Degradation of compounds by microorganisms through direct metabolism or co-metabolism	[2]
Phytoremediation	Removal of compounds either directly by plants or due to a plant's physical presence or associated organisms	[78]
Phycoremediation	Removal of compounds by micro or macro algaes by sorption or degradation	[94]
Filter Feeding	Sorption or digestion of compounds by filter feeding animals like clams and mussels	[95]

5.1. Microbial Biodegradation

Microorganisms are the primary decomposers of organic matter in wetlands [216], which also makes them instrumental in the degradation of organic xenobiotic compounds like ECs [2]. Direct metabolic consumption, co-metabolic processes and secondary metabolite production in bacteria and fungi can break down complex and recalcitrant compounds in environmental matrices [217]. Although the term "biodegradation" can technically refer to contaminant breakdown by any living organism, it usually refers to microbial processes in the research literature on CWs.

The microbes performing biodegradation in wetlands primarily live in the following settings: (1) rhizosphere soil very near to plant roots, (2) inside of plant roots as endophytes, (3) free living in surface or porewater, or (4) in biofilms growing on plant surfaces, organic matter and wetland substrates [60–62]. Larger amounts of total biomass in CWs have been shown to increase biodegradation rates of ECs [9] as more organic substrate promotes the growth of organisms responsible for contaminant degradation. A diversity of organic substrates may be important to fuel biodegradation as the removal of certain ECs has been correlated with the use of specific carbon sources [63].

Long term sampling of CWs suggests that one potential advantage of biodegradation as an EC removal mechanism is that it is a sustainable process that can occur in perpetuity, compared to sorption, which can become impaired as the substrate becomes saturated [17]. Biodegradation is considered to be an important, if not the most important, mechanism for EC removal in wetlands [58,61], but this varies on a compound-by-compound basis [64].

5.1.1. Biodegradation Systematic Review Summary

Biodegradation as an EC removal mechanism in wetlands was discussed in 60% of the primary research articles found in the literature review (75 out of 125 articles), making it the most discussed removal mechanism. Of the research articles addressing this mechanism, 24 were performed in laboratory or greenhouse settings, while 51 were in outdoor field settings. Additionally, 43 were performed in microcosms or mesocosms, 31 were studies of operational CW systems and two were in natural wetlands. It was far more common for biodegradation to be discussed in the context of indirect measurements and EC physicochemical properties, than through direct measurements of microbial abundance and activity.

5.1.2. Parameters Influencing Biodegradation

Various parameters have been shown to be influential on the type and efficacy of biodegradation in wetlands. Some of the most prominently discussed parameters in the literature are temperature, hydraulic retention time (HRT), dissolved oxygen (DO) and ORP.

Seasonal changes in efficacy are often observed in CWs treating ECs, with lower removal rates in winter months [65]. These differences are generally attributed to decreases in temperature slowing down microbial growth and metabolism, thus lowering biodegradation rates [39,45,218]. The presence of aquatic plants may mitigate this seasonal shift in temperature, as microbial activity in planted mesocosms have been shown to maintain EC removal in cold weather when unplanted mesocosms showed declines in removal rates [36].

HRT often shows a positive correlation with EC removal, likely related to an extended period of time for microbes to interact with contaminants in the wetland [35]. However, the effect can be minimal for some compounds [21].

DO is often associated with higher rates of biodegradation, which is likely due to its influence on ORP [39]. Microbially mediated reduction-oxidation reactions are important to xenobiotic degradation [219] and many organic compounds have favorable ORP ranges for biodegradation to occur [64]. The transformation products formed from ECs during biodegradation can indicate whether aerobic or anaerobic biodegradation has occurred [55].

ECs have a wide range of biodegradability in wetlands [198] and the level of recalcitrance of certain compounds can be traced to a lack of sufficiently reduced environments for optimal degradation. This variability in the biodegradation potential of ECs between aerobic and anaerobic settings highlights the important difference between these two types of environments and the microbes that inhabit them when it comes to contaminant degradation. Differences between aerobic and anaerobic biodegradation of ECs in wetlands will be discussed in the following two sections.

5.1.3. Aerobic Biodegradation

Aerobic biodegradation is performed by microorganisms capable of performing aerobic respiration, using oxygen as an electron acceptor, which is far more energetically favorable than the various forms of anaerobic respiration or fermentation. Most ECs that are considered easily biodegradable are more susceptible to aerobic biodegradation and their removal can be correlated to higher DO and ORP levels [39,66].

A side-by-side comparison of pilot HFCWs receiving continuous aeration, intermittent aeration and no aeration showed removal efficiencies for all 13 ECs measured increased with more aeration [12], likely from enhanced aerobic biodegradation. VFCWs tend to show better EC removal results than HFCWs [39] and this is thought to be related to VF systems promoting more aerobic conditions for biodegradation than HF systems [198,220].

The removal of ECs like tebuconazole and ibuprofen has been positively correlated with nutrient removal and DO, indicating that aerobic biodegradation plays an important role, as well as there being potential co-metabolism between organisms degrading ECs and transforming nitrogenous compounds, such as ammonia oxidizing bacteria [59,60]. For phosphate, which is more susceptible to sorption than biological transformation, the correlation could be due to increased biofilm abundance as a result of microbial growth and metabolism [63]. In these studies, contaminant removal was greater in planted mesocosms, but the concentrations measured in the substrates and plant tissues were low enough to suggest that aerobic biodegradation was the dominant mechanism.

Many ECs have been shown to be effectively biodegraded in aerobic but not anaerobic conditions. Radiolabeled carbon experiments have demonstrated this to be the case for caffeine, nicotine, cotinine, estrone, 17b-estradiol, 17a-ethinylestradiol, BPA and 4-nonylphenol using sediment microcosms. [67,221–223]. However, some ECs are recalcitrant to aerobic biodegradation. One of the most prominent examples is carbamazepine, which has had negative removal rates measured in CWs, possibly because its metabolites are present in the inflow and are transformed back into the parent compound in aerobic conditions [17].

5.1.4. Anaerobic Biodegradation

Anaerobic biodegradation is performed by microorganisms that are capable of fermentation or anaerobic respiration, utilizing electron acceptors other than oxygen, such as sulfate or nitrate. They can be obligate or facultative anaerobes. Wetland systems have anaerobic compartments, the extent of which is related to various environmental and, in the case of CWs, design factors. HFCWs and other designs with greater saturated substrate depths tend to be more anaerobic than VFCWs [39]. Emergent vegetation, while contributing to rhizosphere oxygenation through radial oxygen loss, can also cast shade over the water surface, which can potentially prevent the growth of microalgae and subsequent water column oxygenation [224].

Degradation reactions mediated by anaerobic microbes in wetlands include the transformation of amide and urea functional groups from compounds such as carbamazepine and atenolol and reductive dehalogenation of compounds such as diclofenac and triclosan [68]. Removal rates of around 50% for diclofenac and carbamazepine have been demonstrated in a HFCW with reduced conditions, suggesting that anaerobic biodegradation was a key process [22]. However, removal rates of ECs via anaerobic degradation can vary greatly within and between studies. One anaerobic HFCW pilot system reached a removal rate near 50% on some measurements while also frequently displaying negative removal rates for various ECs [12].

A comparison between different CW designs treating a range of ECs showed that HFCWs were least effective for removing all the measured ECs, with the exception of carbamazepine, for which it was most effective [39]. Carbamazepine is known to be recalcitrant to aerobic biodegradation, so the HFCW may have been the only system that achieved sufficiently reduced conditions to foster anaerobic biodegradation.

Sometimes it can be difficult to generalize about the susceptibility of a compound to specifically anaerobic or aerobic degradation. For example, ibuprofen has been observed to be resistant to anaerobic degradation [64,195], but Li et al. [69] demonstrated increased ibuprofen degradation in HFCW mesocosms along horizontal flow path that was correlated with decreased DO. The production of enzymes and cofactors from anaerobic microbial metabolic processes such as glucose fermentation and sulfate reduction were also observed, indicating anaerobic biodegradation mechanisms in this study [69].

5.1.5. Microbial Communities in EC Affected Wetlands

A number of microbial surveys have been performed in the sediments and rhizospheres of wetlands treating ECs, giving some examples of taxa that may be involved in the biodegradation process [61]. The populations of bacteria or fungi in wetlands affected by ECs can be influenced by the selective forces favoring organisms that are able to utilize the xenobiotics as a carbon source as well as tolerate potential toxicity from the ECs to which they are exposed [69]. This second point is especially noteworthy for many PPCPs that are used as antibiotics, designed to kill bacteria [225]. This result has been shown initially to decrease taxonomic diversity but can eventually lead to a toxin tolerant population better adapted to perform biodegradation [69,70].

Table 6 lists some examples of bacterial taxa identified in wetlands that are thought to be involved in the biodegradation of ECs. Sometimes the microbes in greater abundance in EC treatment wetlands are of unknown taxa [63], highlighting the fact that many environmentally important microorganisms are yet to be characterized. Understanding the communities and metabolic relations of these organisms is important because the biodegradation of ECs can be a result of direct metabolism or co-metabolism involving microbes that do not directly involve the EC of interest [69,217].

Table 6. Examples of microbial taxa associated with the degradation of emerging contaminants in wetlands.

Taxonomic Rank	Name	Associated ECs	Reference
Phylum	Proteobacteria	1-H-benzotriazole, diclenofac, ibuprofen	[63,69]
	Actinobacteria	1-H-benzotriazole, diclofenac, BPA	[63]
	Firmicutes	1-H-benzotriazole, ibuprofen	[63,69,226]
Family	Burkholderiaceae	Toluene	[227]
	Flavobacteriaceae	Ibuprofen	[69]
	Mehylcoccaceae	Ibuprofen	[69]
Genus	Tetrasphaera	1-H-benzotriazole, diclenofac	[63]
	Acinetobacter	1-H-benzotriazole	[63]
	Oceanicella	1-H-benzotriazole	[63]
	Flavobacterium	Diclofenac	[63,71]
	Burkholderia	Toluene	[227]
	Ralstonia	Toluene	[227]
	Pseudomonas	Carbamazepine	[71]
	Ignavibacterium	Ibuprofen	[69]
	Propionibacterium	Ibuprofen	[226]

5.1.6. Methods for Assessing Biodegradation

The microscopic nature of organisms involved in biodegradation makes measuring their contributions to EC removal in wetlands complicated, often relying on indirect estimates or advanced experimental techniques. Methods for determining the presence of microbial activity and hence biodegradation potential in wetlands, can include direct measurements such as environmental DNA surveys [60,69], stable isotope probing (SIP) [72], carbon radiolabeling of compounds [67] and enrichment culturing of field samples [73], or indirect measurements such as water physico-chemistry, enzyme activity, carbon content, HRT and temperature [39,45,74,218].

EC removal is often attributed to biodegradation if decreases in effluent concentrations cannot be accounted for by sequestration in the wetland's various compartments (water, plant tissue, or substrate) using mass balance calculations [39,75,228]. This approach provides a useful approximation but only indirect evidence of biodegradation, as it does not necessarily rule out abiotic mechanisms such as hydrolysis, photolysis and volatilization. These factors can, however, be potentially ruled out through experimental designs such as decreasing light penetration into the water column [36] and using subsurface flow systems [60]. Biodegradation can also be predicted as a significant mechanism for EC removal based on the physicochemical properties of a given compound, such as molecular weight, hydrophobicity, polarity and partitioning coefficients [39,60].

Correlations between EC removal and Community Level Physiological Profiling (CLPP) can be used to assess relationships between microbial metabolism and biodegradation rates [63,70]. The fluorescence diacetate hydrolysis approach (FDA) has been employed to measure overall microbial activity when studying EC affected wetlands [74], but EC removal has been shown to not be correlated with this measurement [63].

Genomic techniques can be used to assess the microbial communities in wetlands treating for or affected by ECs. These methods include denaturing gradient gel electrophoresis and 16S rRNA amplicon surveys [60,63,71]. The microbial taxa observed in genetic surveys can be examined for organisms with known traits related to EC removal [69,71,226]. Studies of this kind have indicated that microbial communities can adapt over temporal or spatial gradients to have a greater abundance of organisms capable of direct EC metabolism, as well as increased tolerance for EC toxicity [69,226]. Therefore, acclimation of the microbial community to the wastewater or xenobiotics to be treated is an important consideration.

More direct measurements of biodegradation can be performed using contaminant compounds with radiolabeled carbon [67,221–223]. Protein-SIP can also be used as a direct measurement to better understand which microorganisms are performing metabolic

biodegradation by measuring labeled ^{13}C-peptides, which allows the peptide sequences to be matched to microorganisms at a species level [72]. This technique was used in mesocosms planted with *Juncus effusus* to determine that toluene degradation was primarily performed by bacteria from the family Burkholderiaceae and mostly from the genera *Burkholderia* and *Ralstonia* [227].

5.1.7. Biodegradation Summary

The biodegradation rates of ECs are closely related to a range of environmental and design parameters such as temperature, HRT and ORP. Most ECs appear to be susceptible to biodegradation in the proper conditions, but some groups of compounds like PFASs appear to be resistant to biodegradation in natural settings and will likely have to be addressed by other mechanisms [52].

Environmental and design parameters such as ORP and HRT along with physicochemical properties of ECs remain important tools for predicting microbial biodegradation processes in CWs. More direct studies on the topic have been enabled by advances in molecular biology techniques and environmental genomics that are starting to create a picture of which taxa of microbes, metabolic interactions and degradation pathways might be important the biodegradation of ECs in wetlands [63,69,72]. However, there remains much to be discovered before there can be a comprehensive understanding of the relationships of microbial ecology and biogeochemistry with EC degradation in wetlands. Rather than favoring certain methodologies for all studies, tailoring the techniques used to the goals of a given study can be a beneficial approach moving forward [61].

5.2. Phytoremediation

Phytoremediation refers to the use of plants to mitigate environmental pollutants. The efficacy of phytoremediation is influenced by physicochemical properties of the contaminants. For example, hydrophobic chemicals are most likely to be available to the root and vascular system of the plants for removal. Organic chemicals can be transported and translocated through the plant body by means of different types of bonding with water molecules. ECs are also metabolized in plant tissues to other chemicals of lesser or greater toxicity [76]. Some plants have shown the capacity to remove and withstand elevated concentrations of emerging contaminants with little to no toxic effects while others show signs of stress due to accumulation of the hazardous contaminants [77,78].

Phytoremediation is largely influenced by seasonal variability [229,230] and the presence of microbial communities, which inhabit the rhizosphere and can increase degradation potential. There are various phytoremediation mechanisms, with phytoextraction, phytodegradation, phytovolatilization and rhizo-degradation potentially playing important roles in the fate of ECs in wetlands (Figure 4). Phyto-stabilization is another common mechanism where contaminants are immobilized in the rhizosphere, but it is difficult to disentangle phyto-stabilization from sorption and was generally not discussed in the literature reviewed here.

Phytoremediation mechanisms work in conjunction with other biotic and abiotic processes in wetlands, so many factors affect its role in EC removal. An examination of ECs sequestered in the leaves collected from five different plant species over a 24-month period showed that biodegradation, plant uptake, microbial degradation, seasonal variation, solar radiation and air temperature can all simultaneously influence removal rates [8].

5.2.1. Phytoremediation Systematic Review Summary

Phytoremediation was the third most researched removal mechanism from the literature review and was discussed in 52% of the primary research articles (65 out of 125 articles). Of these, 42 studies were conducted in controlled mesocosms, while 24 studies examined full-scale wetlands. Thirteen (13) studies were conducted under laboratory conditions, 16 in greenhouse settings and 36 in outdoor field settings. These experimental setups had

different types of plantings that included free floating plants, submerged plants, floating leaved plants and emergent plants along with varied flow regimes.

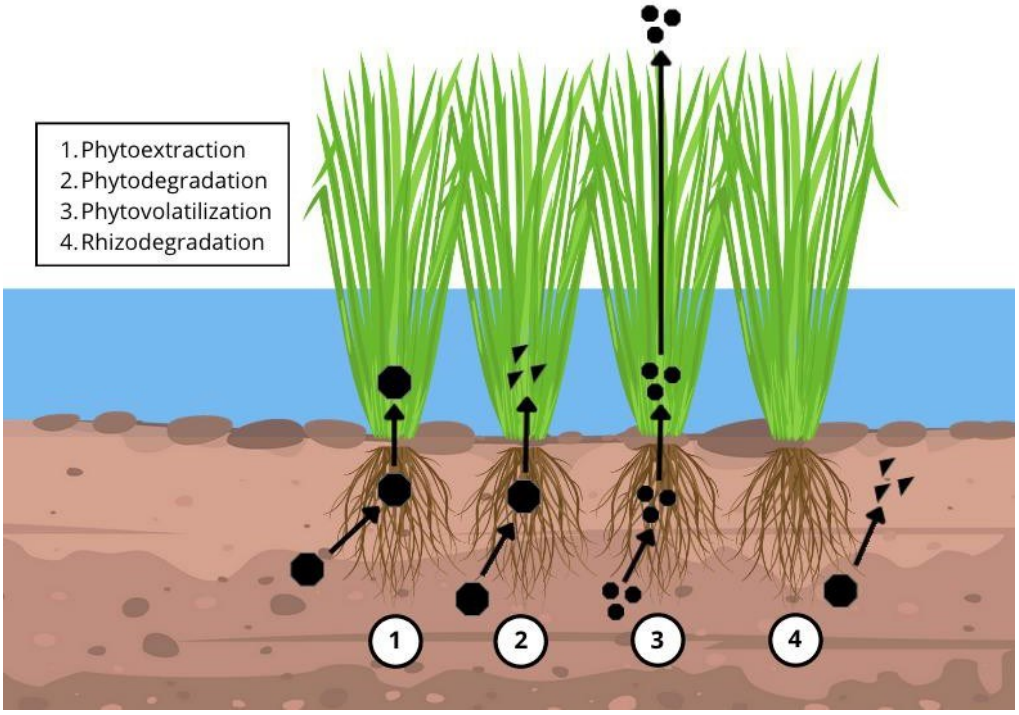

Figure 4. Types of phytoremediation for emerging contaminants in wetlands. Octagons represent parent compounds, triangles represent transformation products.

5.2.2. Phytoextraction

Phytoextraction involves removing chemicals from an aqueous or solid medium through the root system. It requires ECs to be in a water-soluble form in order for the root system to extract them and is aided by chelation inside the plant, which increases contaminant mobility and the potential to safely sequester them [231]. Phytoextraction is generally synonymous with the phrase "plant uptake", which is frequently used in the literature.

Different parts of the plant body can store ECs following extraction. When the parent compound is sequestered in plant tissue, it is sometimes called phytoaccumulation. For example, *Typha latifolia* has been shown to have an even distribution of ibuprofen in the lamina and sheath tissues in a HFCW system, demonstrating the translocation of the contaminant from the roots to the leaves [79]. EC accumulation via phytoextraction can be assessed through the bioconcentration factor and translocation factor of plants. The root and shoot system of *Scirpus validus* has been shown to be responsible for 6–13% and 22–49% removal of clofibric acid, respectively [80]. The translocation factor indicated that more clofibric acid was stored in the shoots than the roots, suggesting phytoextraction was important [80]. Similarly, Lyu et al. [59] attributed 2.5–12% of tebuconazole removal to phytoextraction. Phytoextraction can occur with transformation products as well as parent compounds. For example, ibuprofen from a sand-based medium was degraded by microorganisms alongside simultaneous decreases transformation product concentrations, suggesting uptake by the plants [81].

The efficacy of phytoextraction is influenced by various properties of the contaminants, plants and treatment system design. Water content of the root tissue and low K_{OW} values of contaminants could be key for effective phytoextraction [79]. Increased aeration has also been shown to enhance phytoextraction and translocation and result in elevated concentrations of ECs in the shoot system of *Scirpus grossus* in a VFCW [81].

Various species of aquatic plants have been studied for EC phytoextraction capabilities. *Glyceria. maxima*. was found to be effective for the removal of ibuprofen [76].

Phragmites australis is one of the most studied plants in CWs and phytoextraction is well documented for this species [58,82–84]. *Typha*, *Juncus*, *Lemna* and *Canna* species are also widely studied, with demonstrated plant uptake of various ECs [36,37,47,85–87].

Phytoextraction may have potential for treating PFAS, an especially problematic group of compounds. The tolerance and accumulation of PFAS in a greenhouse experiment using a native Australian wetland plant *Juncus sarophorus* indicated it is an excellent candidate for PFAS removal, as it has tolerance to PFAS compounds and potential to extract perfluorohexane-sulfonic acid and PFOA from soil [47]. However, it is less efficient in the uptake of long chained PFOS compounds [47]. Yin et al. [11] also found also evidence of plant uptake of PFAS in a reed bed CW system with multiple aquatic plant species.

5.2.3. Phytodegradation

Phytodegradation refers to the ability of plants to internally modify the molecular structure of contaminants into potentially non-toxic metabolites [88]. It usually follows the uptake of contaminants from the external environment via phytoextraction and results from intracellular enzymes and plant metabolism, with additional influences from microbial activities [89]. However, it has been found that some transformation products can be more toxic than the parent compounds [90].

The potential for phytodegradation of ECs is supported by observations of the accumulation of ibuprofen metabolites in the shoots and the leaves of *Typha angustifolia* combined with limited root uptake of these transformation products, suggesting they are not directly uptaken via phytoextraction [79]. In this study, transformation products such as carboxylated ibuprofen, 2-hydroxy ibuprofen and 1-hydroxy ibuprofen were measured in the leaf tissues, demonstrating the incomplete degradation of ibuprofen inside the plant body. The transformation of the ibuprofen to carboxylated and hydroxylated metabolites has been attributed to the enzymatic activity of carboxylesterases and monooxygenases [232]. *P. australis* can form similar ibuprofen metabolites through phytodegradation [89].

5.2.4. Phytovolatilization

Phytovolatilization is the process of plants absorbing contaminants and releasing them from their aboveground tissue by transpiration [78]. When phytoextracted ECs are transported to the shoots and leaves of the plant body, compounds with lower molecular weight can be removed by phytovolatilization [233]. The rate and efficiency of the mechanism are dependent on the properties and concentrations of the EC and the type of plant species [91]. Phytovolatilization is less commonly addressed in primary research than other phytoremediation mechanisms because it is more difficult to measure the release of volatile compounds from plant tissue in a wetland setting than plant tissue concentrations of compounds and metabolites, which can be indicative of phytoextraction and phytodegradation.

5.2.5. Rhizodegradation

Rhizodegradation refers to the degradation of organic compounds, including ECs, in the plant rhizosphere, a diverse and complex microhabitat created by plant roots (Figure 5). In wetlands, plant roots are especially influential in shaping their surrounding environment where, in addition to releasing acids and carbon-based exudates like terrestrial plants, they also release oxygen into the surrounding sediments, creating aerobic microhabitats in what would likely otherwise be an anaerobic environment [234].

A variety of different organisms can be involved in rhizodegradation (Table 7). Plants are generally not directly responsible for contaminant degradation in this setting, but instead facilitate the biodegradation of organic compounds by microbes that rely on the plant root architecture and radial oxygen loss for habitat and root exudates for food [219]. With respect to EC degradation, this creates a very complex and dynamic system to assess as exposure to ECs has been shown to alter the composition and patterns of aquatic plant root exudation [92], likely causing downstream effects on rhizosphere ecology. Improved

EC removal is often observed in planted CWs compared to unplanted systems where phytoextraction and other phytoremediation processes do not appear to be dominant, supporting the concept of rhizosphere microbial communities being instrumental [59]. Ignavibacterium and Rhodocyclaceae were found to be correlated with ibuprofen removal but were only present, or present at far greater abundance, respectively, when *T. angustifolia* was present [69].

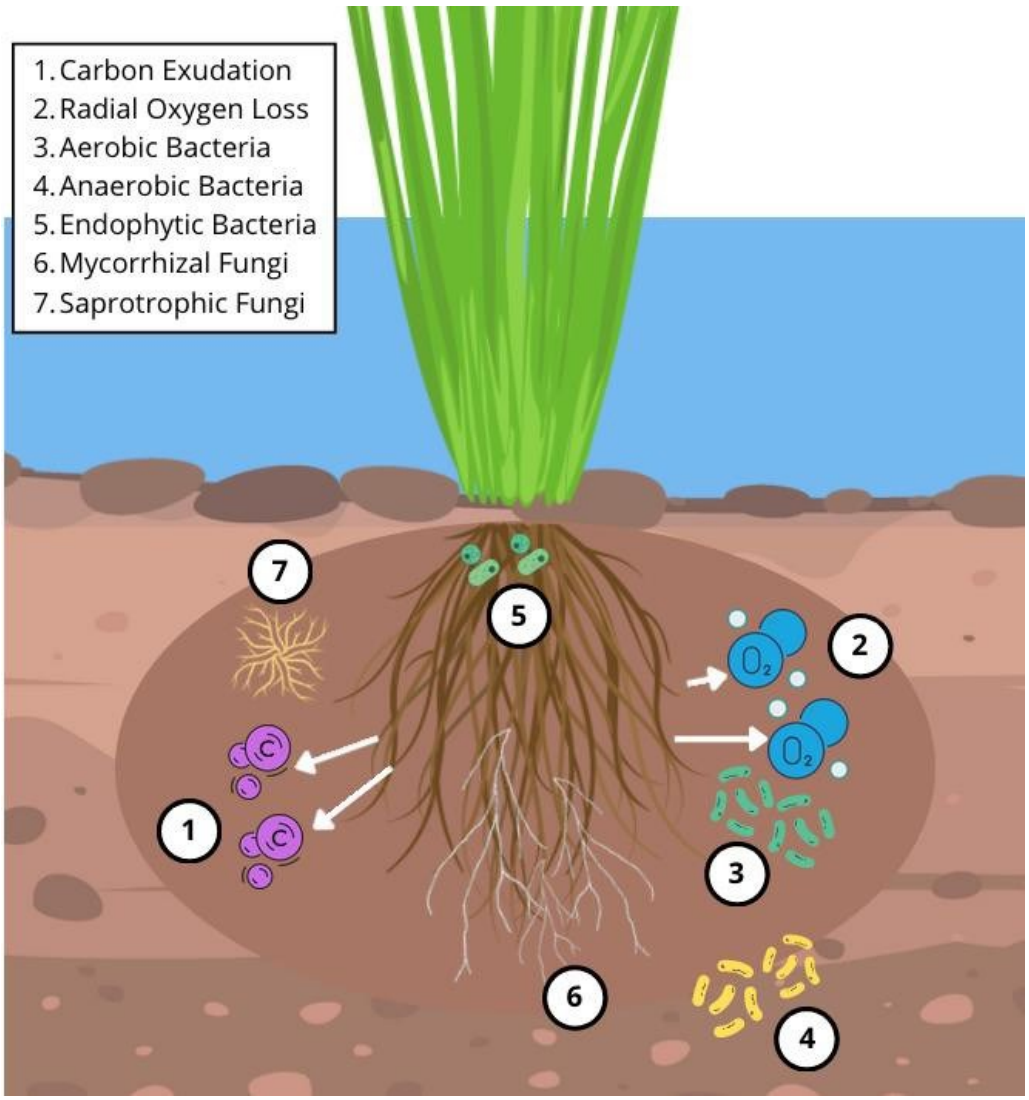

Figure 5. Rhizosphere processes and organisms related to rhizodegradation. The roots of emergent macrophytes perform the listed functions or facilitate the presence of listed organisms.

Table 7. Types of organisms and functional roles for emerging contaminant removal in the rhizosphere.

Organism	Role
Plants	Radial oxygen loss and carbon exudates from roots, root uptake of compounds, root surface adsorption and plaques
Aerobic Bacteria	Aerobic biodegradation in oxygenated zones
Anaerobic Bacteria	Anaerobic biodegradation in non-oxygenated zones
Endophytic Bacteria	Assist in compound degradation and reducing phytotoxicity
Mycorrhizal Fungi	Improve plant growth and rhizosphere adsorption, reduce phytotoxicity
Saprotophic Fungi	Biodegradation

Endophytic bacteria living inside the roots of wetland plants also have the potential to improve rhizodegradation capacity [62]. Inoculation of *Juncus acutus* with cultured root endophytes has been shown to increase plant growth promotion as well as improve the decoloration of synthetic dyes [73] and the degradation of BPA, ciprofloxacin and sulfamethoxazole [235]. In the case of Riva et al. [73], endophytes were cultured from *P. australis* in a wastewater treatment plant in Morocco and used to inoculate *J. acutus* in Greece, suggesting potential for intercontinental and interspecies application of this technique. Inoculations with a consortium of endophytic isolates have been more effective than using a single strain [73,235].

The influence of fungi in aquatic rhizospheres is far less commonly studied and likely less influential than in terrestrial systems but may also be important to EC rhizodegradation in wetlands. Symbiotic mycorrhizal fungi, which are generally aerobic and must grow from a plant host, were once thought to be rare in wetland plants [236]. However, they have become increasingly observed in the roots of emergent macrophytes [71] and have been shown to influence rhizosphere conditions in CWs, though their ability to improve EC removal is somewhat ambiguous and appears to be highly substrate dependent [76,237].

Although even less well studied than mycorrhizal fungi, saprotrophic fungi have been cultured from wetland sediments and strains cultured from these settings have demonstrated the ability to degrade the PAHs anthracene and fluoranthene [238], suggesting potential contributions to biodegradation of ECs in the rhizosphere by these organisms as well.

5.2.6. Phytoremediation Summary

Most of the studies discussing phytoremediation from the literature review focused on overall contaminant removal performance based on mass balance calculations. However, phytoremediation processes are complex and depend on plant metabolism, chemical properties of the contaminant, microbial communities associated with the plant and other physical factors. Studies that focused on these factors in more depth were less common [36,37,79,89]. There are many unknowns to be explored for phytoremediation of ECs in wetlands. For example, genetic modification involving the insertion of foreign genes associated with degradation (e.g., GS, laccase, γ-ECS, CYP450) into plants is becoming more common [77]. In addition, potential toxic effect on plants from the ECs they are remediating could be further explored. Although the capability of aquatic plants to remove ECs from water has been demonstrated, the underlying mechanisms like internal translocation of compounds, transformation into metabolites and the roles that microorganisms play in these processes needs more investigation. Each EC is unique, their characteristics often vary and the subsequent fate via phytoremediation also varies.

5.3. Other Biotic Mechanisms

Although research into biotic mechanisms driving EC removal in wetlands is largely dominated by microbial biodegradation and phytoremediation, wetlands can be complex ecosystems with a wide range of biological organisms, trophic levels and niches that may directly or indirectly influence EC removal. For example, oligachaete worms are known to enhance bioturbation in wetland sediments, a process in which they create burrows and change the physical structure of the substrate while transporting oxygen and contaminants into anaerobic sediments and the plant rhizosphere [239,240]. These actions would likely influence some of the above-mentioned mechanisms such as sorption, biodegradation and phytoremediation. Phyco-remediation and removal by filter feeders are two other biotic mechanisms that have been found to improve the removal of ECs in wetlands. These mechanisms naturally occur in wetland systems, but their impacts in CWs are often overlooked.

5.3.1. Phyco-Remediation

Phyco-remediation, or the use of algae and microalgae for biosorption, bioaccumulation and biodegradation of contaminants, is an emerging field of study in the context of wetlands [93,94]. Research has shown phyco-remediation to be a viable removal mech-

anism for ECs [35,93,94]. Favorable compound structure, such as high hydrophobicity and solid-water distribution coefficients (K_d) above 3.2, increase the biosorption of ECs to algae [94]. Contaminants with cationic groups can be easily biosorbed through electrostatic interactions to the negatively charged algae cell wall [93]. Bioaccumulation through passive diffusion, passive-facilitated diffusion and active uptake of a contaminant into the cell can facilitate biodegradation and transformation of a contaminant [202]. Degradation of ECs by algae is influenced by the concentration of the contaminant in the water source, the algal species, enzymatic pathways, treatment conditions and the type and structure of the contaminants [202].

The presence of algae in a system can also work in tandem with other mechanisms for removal. Algae have been found to induce indirect photodegradation by providing excess chlorophyll, extracellular organic matter and enzymes that generate free radicals to act as photosensitizers [94]. Microalgae systems can also indirectly increase the volatilization rates contaminants because of their standard system conditions with high aeration rates, sunlight exposure and known operating temperatures that encourage volatilization [202].

Phyco-remediation can be used in conjunction with CWs for EC removal. Microalgae tanks can be used as a pre-treatment unit to a CW [93]. An algae pretreatment unit would provide oxygen through photosynthesis to the system, benefiting aerobic contaminant removal processes in the CW [93]. A CW could also be inoculated with algae species to enhance phyco-remediation in the system [94]. The efficacy of these algae tank-CW combined systems are dependent on treatment parameters such as wetland hydraulic design, pH, temperature and seasonality and ORP and further research and evaluation is needed into how these parameters impact the efficacy of EC removal in these systems [94].

5.3.2. Filter Feeders

Filter feeders occur in wetlands and may be another influential biotic removal mechanism. The use of mollusks for monitoring pharmaceutical loads and impacts is a developing field of study. Bayen et al. [95] used the Asian green mussel (*Perna viridis*) and the lokan clam (*Polymesoda expansa*) to understand the bioavailability of pharmaceuticals in a mangrove ecosystem. Concentrations of bisphenol-A, caffeine, carbamazepine, sulfamethoxazole and lincomycin were detected in the bivalve tissues of a portion of the mussels and clams used in the experiment [95]. From this, Bayen et al. [95] hypothesized that some pharmaceuticals are bioavailable in mangrove waters for uptake by mussels and at the mangrove sediment-water interface for uptake by clams. Although this research provides more information about how ECs are removed by other biotic systems in an ecosystem, these studies were not designed to determine the efficacy of mollusks for EC removal. More research is needed to expand the current knowledge base of EC removal by filter feeders.

6. Discussion

The application of wetlands for mitigating ECs in aquatic systems is a growing topic of research and knowledge of the removal mechanisms at play in CWs and natural wetlands are key to an improved understanding of this environmental issue. This review sought to provide an overview of the current research on the various EC removal mechanisms in wetlands. The mechanisms receiving the most attention in primary research reviewed were microbial biodegradation (60%), sorption (54%), phytoremediation (52%) and photodegradation (25%), which were discussed in the primary research articles utilized in the review. Other abiotic mechanisms such as volatilization and other biotic mechanisms such as phyco-remediation were also discussed in the literature, but at much lower rates of 7% and 4%, respectively.

The feasibility of creating concise and comprehensive reviews on topics related to ECs is inherently difficult because of the thousands of chemical compounds that fall into this category that have a wide range of physical properties and environmental behavior [1]. Therefore, it is assumed that this review is not comprehensive. For example, the search terms used focused on “emerging contaminants”, “contaminants of emerging concern”

and "micropollutants" but did not include the individual classes of ECs (e.g., PPCPs, pesticides, industrials), so there is likely to be a large body of related research which may not be covered in this review. However, useful trends related to CW design and operation and wetland removal mechanisms of ECs were discussed, as well as methodologies and techniques used to determine EC removal mechanisms in wetlands.

Some common trends were observed for removal mechanisms based on CW designs. Photodegradation is dependent on sunlight penetrating the water column and therefore plays a larger role in FWSCWs than subsurface systems [2,6]. VFCWs tend to have higher rates of biodegradation because they maintain more aerobic conditions in the substrate than HFCWs or FWSCWs, promoting the aerobic microbial activity which usually leads to the most effective biodegradation [2,6]. Although phytoremediation is obviously more influential when plants are present, biodegradation also tends to be higher in planted systems because of the stimulation of the microbial community in the plant rhizosphere [2]. The selection of wetland substrate with high organic matter content can improve the rate of sorption and biodegradation within a system [45]. Increased HRT can increase the efficacy of most removal mechanisms as it provides more time to act on contaminants present [45,96].

It can be challenging to delineate the removal mechanisms from each other, as wetlands are complex biogeochemical systems where the various processes involved are often highly intertwined. As previously mentioned, the presence of emergent vegetation influences the microbial community in wetland substrates, so it is questionable whether phytoremediation and rhizodegradation, specifically, can be disentangled from microbial biodegradation. Additionally, sorption is influenced by substrate structure, which is in turn influenced by plant roots and microbial biofilms which affect HRT. Although there are methods to interrogate and estimate the individual contributions of different mechanisms, their assessment should consider their interconnected natures and the components of a wetland should be viewed holistically when addressing EC removal.

There were general research gaps identified across all mechanisms examined in this review. Although many studies investigated the removal efficiency of the ECs by CWs, there is a lack of uniformity in reporting. While some studies defined removal efficiency as the difference between the influent and effluent concentration of ECs, other studies defined removal efficiency as the mass balance removal rate. Additionally, removal mechanisms were studied indirectly more often than not. Many studies hypothesized and alluded to specific removal mechanisms being responsible for the attenuation of certain compounds based on prior literature. There is a need for more research that utilizes competent experimental designs to isolate the effects of specific mechanisms. Timeframe is also important for confirming the long-term influence of mechanisms for EC removals in a given setting. Wetlands are dynamic systems with changing environmental conditions, hydrology and vegetation, so sampling a system in a limited temporal range does not necessarily imply that the observed mechanisms and efficacy will persist over time. More studies like Tondera et al. [17] would be helpful for the latter, where the same CW was sampled in intervals of multiple years after the system began operation and seemed to indicate that sorption efficacy declined over time while biodegradation remained stable.

There are a variety of important research gaps for sorption specifically, including a lack of uniformity across substrates in research. Much of the research regarding sorption of ECs to substrate is conducted on local sediment collected in the proximity of the initial study, which makes it difficult to replicate these conditions across multiple studies. More uniform substrate materials exist for sorption experiments, but these are not as commonly used in traditional CWs and therefore provide necessary, but less utilized, information. The lack of uniformity of wetland design and environment conditions also can limit direct comparison between the sorption capacities of contaminants, as it would be difficult to replicate the exact conditions and influent water characteristics between full-scale systems in different locations.

Uncertainty also exists with regard to system pH, which is known to influence the electrostatic interactions between contaminants and substrates that cause sorption. Most of the sorption research found in the systematic review was performed at a circumneutral pH [41,47–49,203,205], which does not provide adequate information about the sorption mechanisms of CWs that might routinely operate outside of this pH range. Future studies could examine sorption mechanisms in more extreme environmental conditions, providing a more thorough understanding of these mechanisms in different environments.

There is also a lack of research into the desorption of contaminants over time [204]. Sorption and desorption are difficult to measure directly in full-scale systems, as other removal mechanisms, such as biodegradation, influence removal simultaneously. The majority of sorption research is conducted through batch experiments that look at sorption specifically or indirectly through hypothesized connections between overall CW removal and sorption. Neither of these methods can appropriately estimate the impact of contaminant desorption over the design life of a CW. More long-term studies are needed to continue to develop a literature base for this occurrence.

A great deal of photodegradation research is conducted in a laboratory setting with light exposure that differs from solar radiation. Although there is ample research on kinetic constants and half-life of ECs with photodegradation, there is a less thorough understanding of solar-driven photodegradation efficiency in real secondary treated wastewaters with full-scale conditions and the role of photosensitizers promoting indirect photolysis in these settings. There is also a lack of stand-alone experiments focused solely on photodegradation and its performance as a removal mechanism in CWs. Photodegradation is frequently mentioned in studies, but it is often not the main focus. More studies on photodegradation in field scale wetlands with measured solar radiation could help maximize its role in CWs, especially FWS systems.

In recent years there have been many advances in environmental microbiology that are leading to a greater ability to understand microbial biodegradation of ECs in wetlands. Advances in genomics technologies have allowed more robust and sophisticated analyses and the lowered costs of environmental DNA analyses makes it more affordable to run numerous samples for assessing microbial communities. SIP techniques also enable more specific tracking of the fates of contaminants, as they are designed for determining specific microorganisms involved in EC degradation [72].

Despite these advances, there are still many hurdles to understanding the nuances of biodegradation in wetland EC removal. In general, the majority of studies addressing biodegradation in this literature review primarily utilized correlations between commonly measured water quality parameters and physicochemical properties of target contaminants with their removal rates but did not specifically measure microbial communities or behaviors.

The techniques used and information generated in the more specific studies point towards future steps that could greatly advance the understanding of microbial biodegradation in wetlands. More studies that not only identify microbial taxa in EC affected wetlands, but also identify the genes and metabolic mechanisms responsible for EC degradation, are needed. CLPP can be a valuable tool for the functional analysis of microbial metabolism and carbon source utilization as it relates to EC degradation [241]. Enzyme assays for estimating microbial activity, such as FDA and beta glucosidase, can potentially be useful [74], but more studies are needed to determine the efficacy of these assays as it is currently unclear how accurate of a predictor they are for EC degradation [63], or if certain assays have differential utility for different types of contaminants. More SIP studies on a wider range of representative ECs also have the potential to improve the understanding of which microbial taxa, metabolic processes and biochemical reactions are responsible for contaminant degradation in wetlands. Fortunately, EC affected wetlands are one of the largest areas of focus for microbial community analysis in treatment wetlands, so the understanding of this topic is ripe for advancement, but deliberately planned, multiphasic

studies will be required to effectively link microbial taxa and communities with microbial activity and biodegradation [61].

Much remains to be understood with regards to phytoremediation of ECs in wetlands. Many studies only look at one species of plant, especially with field scale CWs, a majority of which utilize *P. australis*. While there are a number of mesocosm scale studies that compare different aquatic plant species, mixtures of different plant species are rarely studied, which could be an important factor when treating wastewater effluent with complex mixtures of contaminants.

There is a general gap in the research with regards to the mechanisms of phyto-volatilization and phyto-stabilization for ECs in wetlands. These pathways are likely contribute to the removal of the contaminants, but they are usually not adequately interrogated in studies on EC removal in wetlands. It is challenging to experimentally disentangle phyto-stabilization from abiotic sorption processes and, while evapotranspiration combined with decreased plant tissue concentrations could be an indicator of phytovolatilization, it is difficult to distinguish this from phytodegradation.

7. Conclusions

Despite various gaps in the understanding of wetland removal mechanisms of ECs, a solid foundation exists for which mechanisms are at play in different natural and constructed wetlands, as well as direct and indirect methods for assessing their relative importance. The primary mechanisms discussed in the literature are microbial biodegradation, sorption, phytoremediation and photodegradation. In general, CWs appear to be an effective tool for treating a wide range of ECs from wastewater effluent and stormwater runoff. Future efforts to expand the knowledge and understanding of this topic include:

- Utilizing innovative experimental designs and analytical techniques
- Creating more uniformity in data collection and analysis
- More frequent and long-term monitoring of CWs receiving ECs
- Adopting more standardized terminology for ECs
- Focusing research on pertinent compounds that have received less attention

Creating a more thorough understanding of the various EC removal mechanisms at play in wetlands and how they are related to various environmental and design parameters will improve the ability to mitigate this novel environmental challenge and improve the efficacy of already effective nature-based solutions such as CWs.

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