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Review

Diffuse Water Pollution from Agriculture: A Review of Nature-Based Solutions for Nitrogen Removal and Recovery

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Abstract: The implementation of nature-based solutions (NBSs) can be a suitable and sustainable approach to coping with environmental issues related to diffuse water pollution from agriculture. NBSs exploit natural mitigation processes that can promote the removal of different contaminants from agricultural wastewater, and they can also enable the recovery of otherwise lost resources (i.e., nutrients). Among these, nitrogen impacts different ecosystems, resulting in serious environmental and human health issues. Recent research activities have investigated the capability of NBS to remove nitrogen from polluted water. However, the regulating mechanisms for nitrogen removal can be complex, since a wide range of decontamination pathways, such as plant uptake, microbial degradation, substrate adsorption and filtration, precipitation, sedimentation, and volatilization, can be involved. Investigating these processes is beneficial for the enhancement of the performance of NBSs. The present study provides a comprehensive review of factors that can influence nitrogen removal in different types of NBSs, and the possible strategies for nitrogen recovery that have been reported in the literature.

Keywords: nitrogen; constructed wetlands; buffer strips; vegetated channels; water sediment control basins; water pollution

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1. Introduction

Fertilizers have the potential to provide essential plant macro- and micro-nutrients, ensuring crop growth and yield [1]. However, their inappropriate management and excessive application in agriculture have had detrimental environmental impacts, such as the loss of nutrients to surface waters [2] and groundwater [3], decreasing water quality [4], the promotion of eutrophication [5], soil and water acidification [6,7], and alterations in biodiversity [8]. In 2019, the Food and Agriculture Organization (FAO) of the United Nations released the FAOSTAT fertilizer statistics [9], reporting that 192 $\times$ 10^6 tons of chemical and mineral fertilizers were administered to agricultural soils in 2017, specifically, 109 $\times$ 10^6 tons of nitrogen (N), 45 $\times$ 10^6 tons of phosphorus (P), and 38 $\times$ 10^6 tons of potassium (expressed as K_2O). N is therefore the most used fertilizer, and thus the main cause of surface and groundwater contamination [10]. N is conveyed from agricultural soil to freshwater ecosystems during precipitation events, which control the water flow and regulate the N content in run-off and tile drainage waters. Figure 1 shows the global trend of N fertilizers used in agriculture from 2002 to 2017 [11]. From the graph, it can be observed that the use of N has increased by about 30% in the last two decades and therefore it is of extreme importance to address the problems related to N pollution from agriculture. N, if present in soil and water above a certain threshold, mainly in the form of nitrate (NO_3^-) and nitrite (NO_2^-), can be the cause of pollution of different environmental matrices. In particular, the progressive increase of NO_3^- in soils, and its runoff and leaching into freshwaters, has been caused mainly by agricultural activities.

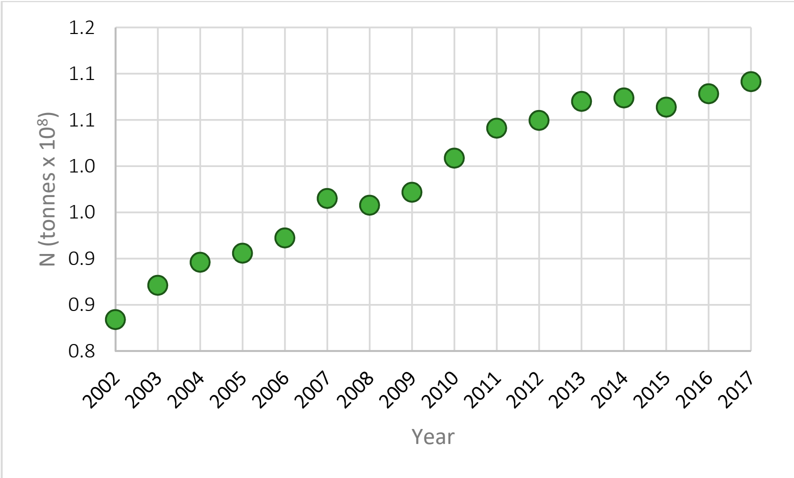


Figure 1. Global trend of nitrogen used as fertilizer—average 2002–2017 [11].

An additional problem is related to the fact that agricultural producers usually apply more N than crops actually need for their growth [12]. The remaining N surplus [12] is not assimilated by crops but released as surplus into freshwater systems. Major international efforts are being made to quantify the amount of N lost since it is useful for the definition of the agricultural pressure on ecosystems. At the European level, for instance, the European Union has defined an agri-environmental indicator know as gross N balance [13], which provides an indication of the N surplus on agricultural land (kg N ha^{-1} $year^{-1}$). Basically, it was observed that agricultural practices commonly exhibit low N-use-efficiency, especially under increasing N inputs [13].

The relevant national institutions have therefore tried to define more restrictive legislation in order to cope with this issue worldwide. For example, the European community recognized this problem in 1991 and it introduced a specific piece of legislation (91/676/EEC) [14], also known as the "Nitrates Directive", which aims to protect the quality of surface and groundwater, preventing pollution caused by NO_3^- of agricultural origin and promoting the use of sustainable agricultural practices.

Unfortunately, management practices, generally supported by agricultural policies and actions, have not been able to mitigate the negative impact of agriculture and N losses have not been reduced to desired levels [15]. Furthermore, the expansion and development of agriculture has resulted in the conversion of natural landscapes into agricultural fields, thereby causing the disruption of the hydrological regime and altering the capacity of ecosystems to reduce N loads from agricultural fields affecting freshwater systems.

For this reason, the construction or restoration of natural systems, such as nature-based solutions (NBSs), have been recently endorsed, aiming to re-establish ecosystem services that would prevent significant losses of N of agricultural origin. NBSs are defined by the International Union for the Conservation of Nature (IUCN) as "actions to protect, sustainably manage, and restore natural or modified ecosystems, which address societal challenges effectively and adaptively, simultaneously providing human well-being and biodiversity benefits" [16]. NBSs can help with wastewater treatment and management, water storage, drinking water provision, groundwater recharge, coastal protection, and agricultural practice, ensuring the provision of ecosystem services and a wide range of other environmental benefits (Figure 2) [17], since they simulate natural processes without further inputs of energy or chemicals. Moreover, different NBSs, namely, constructed wetlands (CWs), buffer strips, vegetated channels, and water sediment control basins, have been recently implemented as edge-of-field strategies to intercept agricultural drainage water and to reduce N loads in receiving water bodies.

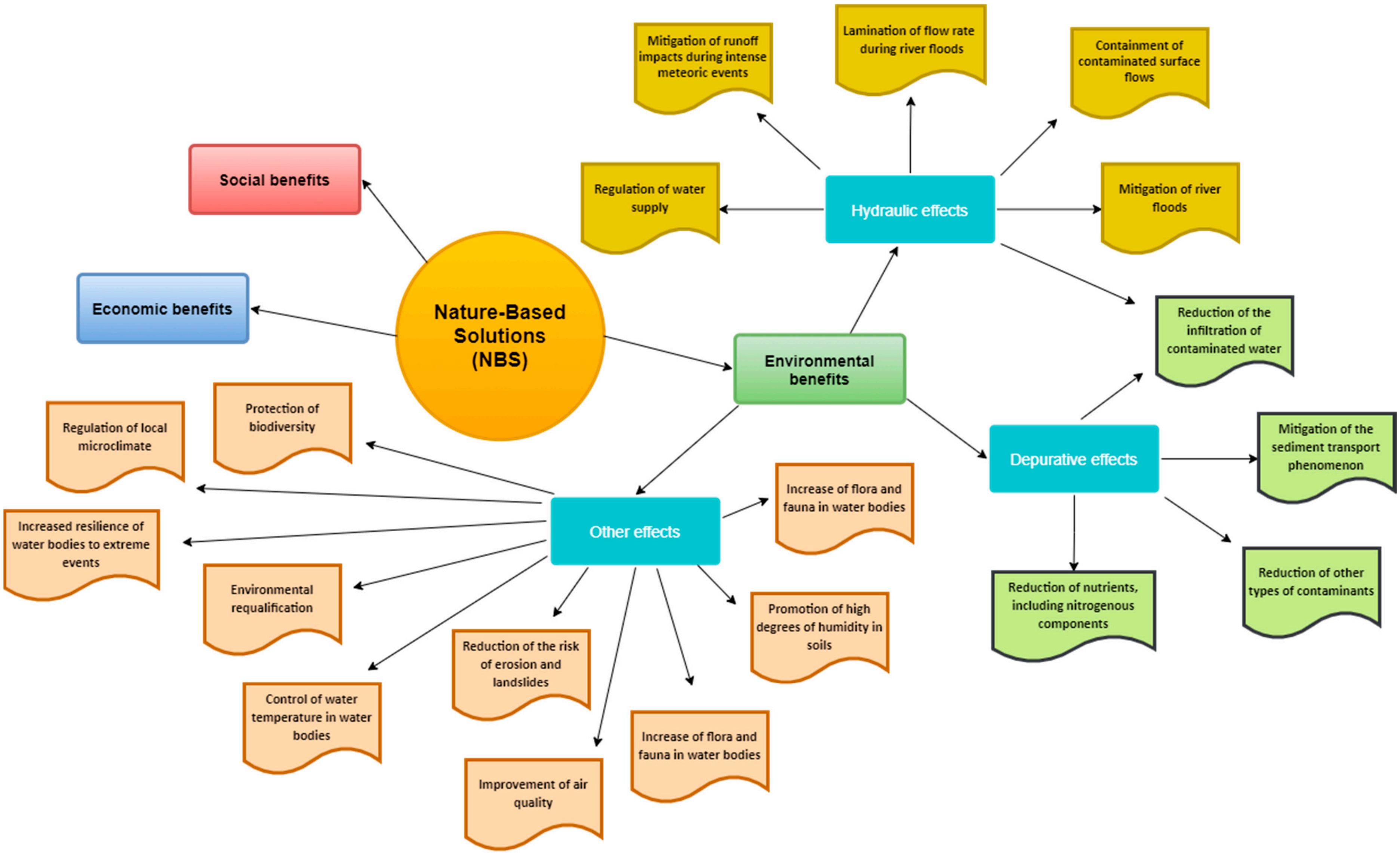


Figure 2. Main effects attributable to the implementation of NBSs.

Several authors have explored the ability of NBSs to mitigate diffuse water pollution. For example, Vymazal (2017) has analyzed the nitrogen removal efficiency of CWs when treating agricultural drainage water [18], whereas Dollinger et al. and Hickey and Doran have assessed the potential of ditches and buffer strips, respectively, to manage pollution from agricultural sources [19,20]. However, the studies available in the scientific literature have often focused on one specific type of NBS and the various contaminants that it can treat. On the other hand, since N has been recognized as the main cause of the contamination of water bodies, the aim of this review was to assess different types of NBS that can be used in agricultural settings and to provide a basis for their wider implementation. CWs, buffer strips, vegetated channels, and water sediment control basins were critically analyzed, examining their typology characteristics, operation principles, process parameters, and operating conditions in regard to N removal from water. Moreover, possible methods of N recovery that would enable its later reuse in agriculture are discussed.

2. Nitrogen Removal Mechanisms

N removal from water can be achieved by means of biological (ammonification, nitrification-denitrification, and plant uptake) and physicochemical (ammonia adsorption and sedimentation) processes [21]. The removal is likely to occur in the same way regardless of whether CWs, buffer strips, or vegetated channels are implemented, whereas minor removal effects can be detected in water sediment control basins, which are mostly used to obtain the lamination of flow rates and thus ensure a more uniform distribution of N concentrations over time [22]. Nevertheless, for all NBS systems, N removal mechanisms involve the interactions among soil, vegetation, and microbial communities.

Organic N, which forms living cells, is biologically converted into ammonia (NH_4-N) through the ammonification process. Then, NH_4-N can be removed mainly via nitrification/denitrification processes, and to a lesser extent via other processes, namely, adsorption, plant uptake, and volatilization [23]. The nitrification process involves the oxidation of NH_4-N by means of nitrifying bacteria (i.e., *Nitrosomonas*), leading to the formation of nitrite (NO_2^-) (Equation (1)), which is further oxidized by other denitrifying bacteria (i.e., *Nitrobacter*) and thus transformed into nitrate (NO_3^-) (Equation (2)). These reactions consume significant amounts of oxygen, eventually leading to a progressive decrease in pH. If it falls below the threshold value of 7.0, an inhibition of the nitrification process may occur. In these cases, the addition of chemicals could be necessary to restore the optimal process conditions.

$$NH_4^+ + 1.5O_2 \rightarrow NO_2^- + H_2O + 2H^+ \tag{1}$$

$$NO_2^- + 0.5O_2 \rightarrow NO_3^- \tag{2}$$

NO_3^-, the most common form of nitrogen in the context of agricultural pollution, can be converted to the gaseous state N_2 through the denitrification process (Equation (3)). This exploits some heterotrophic microorganisms (i.e., *Micrococcus*, *Pseudomonas*, *Bacillus*, and *Achromobactin*), under anoxic or anaerobic conditions.

$$NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2 \tag{3}$$

Different ways to improve and fasten the denitrification process in NBS have been recently studied in order to increase N removal in NBS systems. For example, Zhu et al. [24] have investigated the "anaerobic ammonium oxidation" (Anammox) process, in which NH_4-N oxidation to N_2 occurs by means of autotrophic bacteria (i.e., *Brocadiales*), which use NO_2^- or NO_3^- as electron acceptors [25], without oxygen consumption and other external carbon sources, thus overcoming some limits of the nitrification/denitrification process, such as variations in alkalinity and N_2O production [26].

Plants, an important component of NBSs, can also remove nitrogen through uptake processes, in which NH_4-N and NO_3^- are assimilated and stored by them, later converting inorganic N forms into organic compounds, namely, tissues and blocks for cells [27]. The amount of absorbed N depends on its form within the natural system, on the species of plants that are adopted, and on nutrient concentrations in plants' tissues. Therefore, the use of plants characterized by higher growth rates and N storage capacities should be preferred when promoting N uptake and storage in NBSs. The literature reports that plants can remove up to 20%–30% of the total N input [23].

Other mechanisms for N removal are adsorption and sedimentation. NH_4-N can be easily adsorbed into soils and subsequently released when boundary conditions are changed. Thus, under aerobic conditions, NH_4-N oxidation can take place, resulting in NO_3^- generation. The adsorption process can be influenced by different factors, especially soil typology, the organic matter content in soil, and the presence of water and vegetation. Instead, the sedimentation process can remove most of the particulate organic N [28], which can settle or adhere to the surface of plant stems.

3. Nature-Based Solutions

In the following sections, the influence of various factors on N removal processes in four NBS types will be analyzed, together with their potential to remove this contaminant from agricultural drainage water.

The data used were gathered from the scientific literature dealing with NBSs and factors affecting N removal in these systems. Among different classes of factors that can influence N removal, those for which substantial information was available are discussed in more detail (Table 1).

Table 1. Qualitative and quantitative analysis of scientific studies.

Influencing Factors	NBS Type			
	CWs	Buffer Strips	Vegetated Channels	Water Sediment Control Basins
Vegetation typology	✔	✔	✔	✔
Substrate materials	✔	X	X	X
Microbial communities	✔	X	✔	X
Operating conditions	✔	✔	✔	✔
Total studies (n.)	87	71	7	4

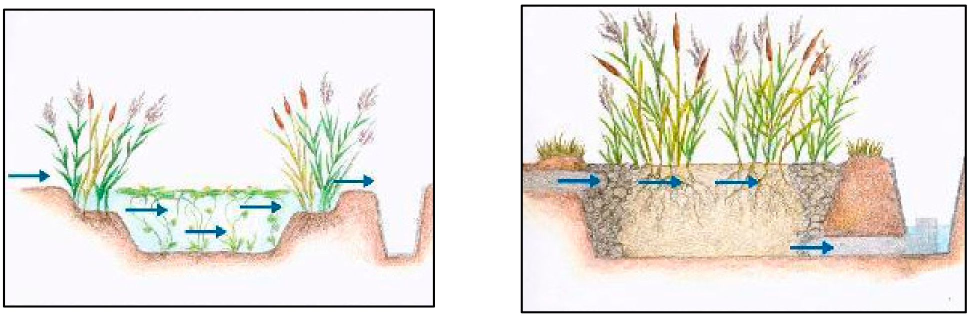
As can be seen in Table 1, the majority of the consulted literature focused on CWs and buffer strips, whereas other systems were given less importance and therefore limited data were available for vegetated channels and water sediment control basins.

3.1. Constructed Wetlands

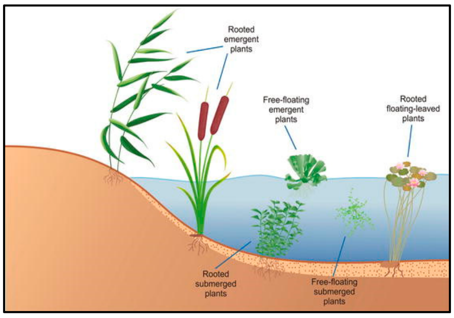
CWs are emerging green technologies, which perfectly describe an example of an NBS. They are also known as "natural bioreactors" due to the complexity of processes that can take place in these systems, as well as the high number of components that are usually involved. In recent decades, CWs have been increasingly adopted as a sustainable treatment technique for the removal of conventional pollutants within agricultural effluents (i.e., sediments, nutrients, etc.).

CWs consist of planted beds filled with appropriate substrates and occupied by microbial communities needed for their operation. They can be classified considering different factors, as shown in Figure 3.

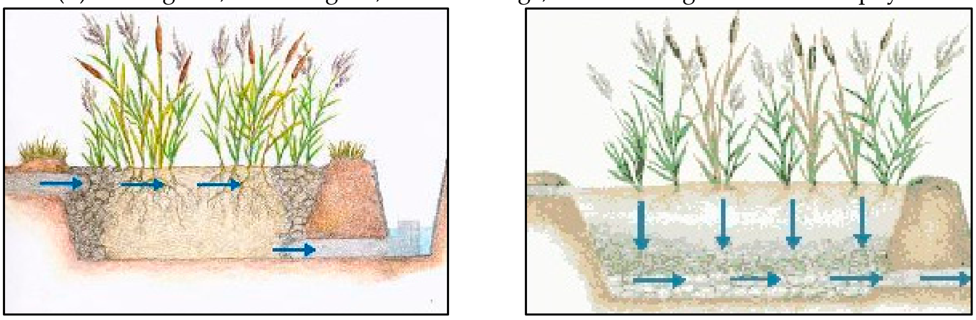
However, according to hydraulic water flow, they can be categorized into surface flow (SF) and sub-surface flow (SSF) CWs, which can later be divided into horizontal (HF-SSF) and vertical flow (VF-SSF) CWs [29].



(**a**) "Surface flow" and "Sub-surface flow" systems



(**b**) "Emergent", "submerged", "free-floating", and "floating-leaved" macrophyte



(**c**) "Horizontal sub-surface flow" and "Vertical sub-surface flow" systems

Figure 3. CW classification based on (**a**) hydraulic water flow characteristics ("surface flow" (SF) and "sub-surface flow" (SSF) systems); (**b**) the type of macrophytic growth ("emergent", "submerged", "free-floating", and "floating-leaved"); (**c**) the flow path in SSF systems ("horizontal" (HF-SSF) and "vertical" (VF-SSF) systems) [26]; IRIDRA—http://www.iridra.eu/en/, accessed on 16 May 2021.

In SF CWs, water flows horizontally over the wetland support medium, between the stems of plants and through any surface debris. The water depths are generally in the range between 0.2 and 0.6 m (in any case <1 m) [30]. The shallow water basin within SF CWs let sunlight permeate down to the bottom, allowing faster algal growth rates and the activation of photosynthesis reactions. Shallow depth can also increase water aeration by the overlying atmosphere. On the contrary, anaerobic conditions can be detected approaching the bottom of the system. Even though they have generally showed lower contaminant removal efficiencies if compared to SSF CW systems, SF CWs have been widely used to treat agricultural run-off and drainage waters, due to the low costs associated with their implementation and maintenance [31].

SSF CWs are sealed basins filled with permeable substrates, namely, rocks, gravel, and soil or a combination thereof. In addition to its filtering action, the CW's medium has the function of assisting the growth of emergent plants. The nomenclature "hybrid systems" instead refers to the combination of HF-SSF and VF-SSF CWs, which have been used to enhance treatment efficiency [30], especially for the removal of N from water [32].

Our literature review revealed that CWs are generally associated with a great variability in their N removal efficiencies. The removal of N has been observed to vary between 11 and 13,026 kg N ha^{-1} year^{-1}, with an average value of 426 kg N ha^{-1} year^{-1} [18]. In order to reduce this variability, it is important to investigate the influence of different geometric configurations, physical components, and operating conditions, which can affect the performance of CWs. These aspects are analyzed in the next sub-sections, Sections 3.1.1–3.1.4.

3.1.1. Vegetation Typology

Plants are the essential component of CWs, and the selection of the appropriate typology can contribute to achieving the desired CW treatment performance. The required plant characteristics are good adaptability to the local climate, pollutants, hypertrophic waterlogged conditions, rapid spread and growth, and they have to show a high removal capacity through the direct absorption and storage of nutrients, or indirectly through the improvement of microbial processes [33].

Plants come under the category of macrophytes, which include four different types, namely, "emergent", "submerged", "free-floating", and "floating-leaved" macrophytes (Figure 3b). They can effectively use nutrients after they have been converted to reactive forms such as NO_3^- or NH_4-N. There are essentially three ways to achieve this: (i) the decay of organic matter by means of microbial communities in soils (dead organisms, leaves, etc.); (ii) biological N fixation by means of N-fixing organisms (i.e., bacteria), which transform atmospheric N_2 into biologically available forms of reactive N; (iii) exploitation of reactive N derived from agricultural practices (i.e., the use of fertilizers). There is a wide assortment of plants that can be used in CWs (Figure 4). For example, the use of a mixture of floating, submergent, and emergent macrophytes and grasses in SFCWs has been suggested in order to increase nitrate removal [34].

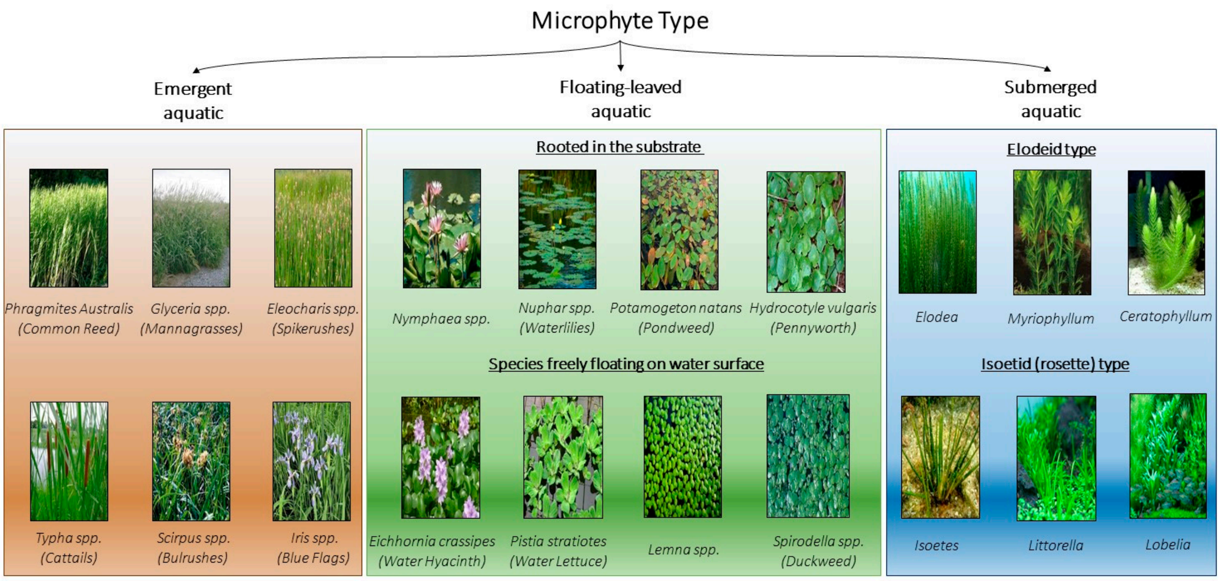


Figure 4. Macrophyte classification [35].

Phragmites australis, *Typha*, *Scirpus*, and *Iris* are the most common species [36], but not all of them have the same treatment efficiency in terms of N removal. This has been proven by different studies, which compared the nutrient removal efficiency associated with the use of various plants under the same boundary conditions. For instance, eight different emergent species (*Schoenoplectus validus, Phragmites australis, Glyceria maxima, Baumea articulata, Bolboschoenus fluviatilis, Cyperus involucratus, Juncus effuses*, and *Zizania latifolia*) in CWs were investigated by Tanner [37], mainly comparing nutrient levels in plant tissues and the potential of the root zone for aeration. *Zizania*, *Glyceria*, and *Phragmites* were the most efficient plants among those investigated [37].

Similarly, Ge et al. [38] studied the influence of eight different species within VF-SSF systems on N removal. The species belonged to three functional groups: grasses (*Arundo donax*, *Phragmites australis*, *Imperata cylindrica*, *Coix lacryma-jobi*), legumes (*Campylotropis* *macrocarpa*, *Aeschynomene indica*), and forbs (*Canna indica*, *Lythrum salicaria*). The authors observed that plants improved the N removal efficiency, although with different percentages. However, it was noted that the biomass of *Coix lacryma-job* increased when both NO_3^- and NH_4-N were present in wastewater. On the contrary, a reduction in biomass was detected for *Aeschynomene indica* when it was exposed to both of these forms of nitrogen.

In the literature it has been demonstrated that the N removal efficiency in CWs can range from 25% to 85% [39]. About 60%–70% of the total N is removed by the denitrification process, whereas a percentage of about 20%–30% is due to plant uptake [32]. Different types of plants might also favor, through their stems, the disaggregation of previously accumulated suspended solids on the surface of CWs, mainly due to wind-induced movement [40]. This limits the effects of surface clogging and promotes the transport of oxygen within CW mediums, resulting in an enhancement of the N removal processes. However, the microbial effect on N removal is usually considered to be more important than that of plant uptake [41].

These aspects, i.e., the selection of the most suitable vegetation, should be taken into consideration when designing CWs.

3.1.2. Substrate Materials

Different CW configurations (i.e., SF or SSF systems) involve the use of various filling materials (also known as substrates, mediums, etc.) for their implementation. Substrates cover different roles, mainly offering the growth medium for plants and microorganisms, providing hydraulic conditions for water flow, and also endowing the necessary conditions for the establishment of most of the physical, chemical, and biological reactions that occur in CWs [35]. The main removal mechanisms promoted by substrates are filtration/sedimentation [42], sorption [43], precipitation [44], ion exchange [45], and microbial degradation [46]. Although in SF systems the function of substrates is mostly related to preventing high infiltration and the consequent contamination of groundwater, in SSF systems they can further provide a filtering action and promote the ideal conditions for the development of microorganisms [35]. When designing CWs, the selection of the most suitable substrates is essential, since they can have a considerable influence on construction costs, purification capabilities, and operating conditions. Substrates have different origins, since they can be derived from natural, agricultural, industrial, and artificial sources. As a consequence, they are characterized by diverse physical and chemical properties, which can lead to different removal rates.

Among natural substrates, gravel, sand, and shale have been the most used substrates in CWs [47]. However, gravel and shale have shown a low capability to adsorb N [48,49]. On the contrary, sand substrates have allowed higher N removal rates [50], probably due to the higher filtering capacity of sand, which is characterized by a smaller particle size distribution if compared to gravel and shale. As an alternative, the implementation of natural soil or sediment has involved the enhancement of nitrification/denitrification and plant uptake processes [48]. Innovative materials have been implemented as well, i.e., zeolite, which is formed through the interaction of volcanic rocks with ash layers together with alkaline groundwater, involving high N removal due to its adsorption into zeolite mesopores and micropores [51,52]. Other natural materials such as limestone and volcanic rocks have been used less frequently, due to their limited efficiency in removing N [53,54].

Among substrates derived from waste from industrial processes, alum sludge has been one of the most used. It is a waste by-product of drinking water treatment plants, generated through the precipitation of aluminum salts, which are commonly used as coagulants. The main process for N removal by alum sludge is adsorption [55]. Higher N removal efficiencies have been detected for fly ash, which can be generated in flue gases after the combustion of coal, due to its high specific surface area, as well as other intrinsic characteristics of this particular material [56]. Slag is another industrial by-product from the blast furnace ironmaking process that has been used as a substrate in CWs. However, N removal rates associated with its use are quite low [57].

Among the artificial materials that have been used as a medium in CWs for N removal, ceramsite [58], activated carbon [59], synthetic fiber [60,61], cement clinker [62], recycled concrete [63], and modified clays [64] can be mentioned.

However, the majority of substrates cited so far may not ensure high N removal since they generally do not provide any organic carbon source, thereby limiting the denitrification process promoted by heterotrophic bacteria. For this reason, naturally occurring materials from vegetation, marine environments, and agricultural wastes have been recently investigated, since they might be potential providers of carbon sources, promoting an enhancement of the denitrification process (proper C/N ratios should be ensured). With this in mind, various researchers have observed that the addition of external carbon sources can favor the modification of extracellular enzyme activities [65] and improve the regulation of microbial biomass and heterotrophic production. Consequently, they enhance the treatment efficiency of CWs, until the entire organic substance is completely used [66].

One of the substrates that was proposed to amend this problem is biochar, a novel carbon-rich material that is produced by the pyrolysis of agricultural biomass waste. It is characterized by a highly porous structure and a high specific surface area, which can allow the adsorption and immobilization of pollutants from contaminated waters. The addition of biochar to a conventional substrate was reported to have beneficial effects for the enhancement of soil fertility [67], the alteration of microbial communities, and the promotion of plant growth [68]. Wood-chips are another natural material that can be easily found near agricultural activities and could be exploited for the leaching out of organic compounds, resulting in an increase of the organic concentration in CW influents, and in the enhancement of both nitrification and denitrification processes [69]. Among other materials available in agriculture, plant waste has also been used for increasing the organic content in CWs, mainly due to the intrinsic characteristics of plants, such as their light weight and porous structure, which have led to significant decreases in N concentrations in agricultural drainage water [70].

Concerning the wetland substrate, it is also important to discuss nitrous oxide (N_2O) emissions, one of the intermediate products of the denitrification process and greenhouse gases. Formed in sediments, its emission into the atmosphere was found to be correlated with the portion of inundated areas in the system. For example, Groh et al. have found that most of the N_2O loss occurred from the terrestrial portions of the wetlands, whereas little N_2O was emitted from inundated areas since it is probable that they weer under anaerobic conditions [71]. On the other hand, some studies have shown that most of the N_2O produced is further reduced to N_2 [72], whereas for a CW located in France it was calculated that the N_2O emitted into the atmosphere was only 9% of the potential emission in sediments [73].

3.1.3. Microbial Communities

Microorganisms play a key role in the removal or the conversion of N in CWs [74]. They can develop in the soil matrix, in which aerobic and anaerobic zones can be further distinguished. As reported in Section 2, N can exist in different forms in wastewater within CWs, contributing to an increased abundance of microorganisms in CWs. Phylogenetic groups connected to N removal are various and can include ammonia-oxidizing microbial communities (*Nitrosomonas* and *Nitrosospira spp.*), denitrifying microbial communities (with the involvement of a wide range of bacterial groups such as *Bacillus*, *Enterobacter*, *Micrococcus*, *Pseudomonas*, *Spirillum*, *Proteus*, *Aerobacter*, and *Flavobacterium*), and anammox microbial communities (i.e., planctomycete-like bacteria *Candidatus Brocadia anammoxidans* in wastewater treatment and *Candidatus Kuenenia stuttgartiensis* in bacterial biofilms, respectively) [75]. Some heterotrophic bacteria in CWs use oxygen as a terminal acceptor. Since these bacteria can provide the highest energy yield, they will dominate whenever oxygen is accessible. However, the absence of oxygen in CWs does not necessarily represent an issue for N removal, since most heterotrophic bacteria are facultatively anaerobic, indicating that they are capable of using NO_3^- or NO_2^- as electron acceptors when oxygen is not available. Hence, it is important to characterize the type of bacteria present in CWs. Several studies have been undertaken to determine the microbial diversity in CWs, analyzing the structure and composition of soil bacteria [76], considering also that different bacteria may favor the selective removal of some contaminants over others. In recent decades, the characterization of bacterial communities has been carried out using molecular techniques and more recently with the application of -omics tools [77]. Bacterial heterogeneity can imply the exploitation of the different functions covered by them. For instance, plant growth-promoting (PGP) bacteria can directly promote the growth of plants, increasing the bioavailability of key nutrients, including N and P, in addition to sustaining the production and the regulation of phytohormones in plants [78]. In addition, bacterial communities that live within plants are shaped as a result of physical, chemical, and biological reactions that can take place in CWs, influencing plants in the N removal process [79].

However, when designing CWs, it should be kept in mind that although the addition of bacteria into CWs can lead to beneficial effects, excessive inoculation could give rise to problems such as clogging [80], which would compromise the correct functioning of CWs, thus reducing their removal efficiency.

3.1.4. Improved Systems and the Influence of Different Operating Conditions

As described in the previous sub-sections (Sections 3.1.1–3.1.3), the N removal process in CWs can be influenced by the adoption of different substrates, plant species, and microbial communities. However, it can be also depend on wastewater properties, CWs' geometric configurations, hydraulic conditions, and other environmental factors.

When the N concentration varies in influents, different removal efficiencies can be detected. This is the case, for instance, of a study conducted by Sengorur and Ozdemir [81], in which different N concentrations in the form of NH_4-N in CW influents were investigated. The authors observed the NH_4-N removal was characterized by a gradual increase over time, mainly due to plant uptake and biodegradation at the beginning, whereas lower removal efficiencies were connected to a lower plant biomass. However, it was also noted that the NH_4-N concentration and removal efficiencies were negatively correlated.

Furthermore, different CWs configurations can lead to different removal efficiencies, mainly due to the variable percentages of oxygen available in these systems, thus affecting the occurrence of nitrification/denitrification processes. Usually, HF-SSF CWs are completely saturated, and the wastewater level is maintained a few centimeters below the surface. In these systems, there is a limited availability of oxygen sources, namely, inputs from the influent, plant release through their root apparatus, and physical surface re-aeration [82]. However, the oxygen required for the purification of conventional polluted waters from agriculture is considerably higher than the sum of all the aforementioned inputs, thereby causing the occurrence of predominantly anaerobic conditions. The conventional feeding modes of HF-SSF CWs, often continuous or in batches, certainly do not help to overcome the limits related to the establishment of anaerobic conditions. On the contrary, VF-SSF are better performing in terms of N removal, since most of these systems are operated with intermittent surface loadings, allowing the aeration of the bulk water and favoring the detection of aerobic conditions.

In order to enhance nutrient removal efficiency by exploiting the advantages of different CW types, hybrid systems have been introduced that combine horizontal and/or vertical systems in series. However, the results obtained were not always the expected ones. For instance, in their study, Kantawanichkul and Somprasert [83] have observed that a horizontal flow sand bed combined with a vertical flow vegetated bed, and vice versa, did not show significant differences in terms of N removal.

Another possibility for the improvement of CWs is provided by the administration of air to the systems. Artificial aeration can be performed in a continuous mode, 24 h per day, although this arrangement does not lead to the maximum achievable levels of N removal due to the absence of an alternation between nitrification and denitrification phases [84]. Moreover, intermittent artificial aeration, in which an alternation of aerobic and anaerobic conditions occurs, can involve the simultaneous reduction of the organic matter as well as the removal of N [85].

In addition to the administration of air, the feeding of influents into CWs can have different effects on the nitrification/denitrification processes. The intermittent feeding mode was proven to favor aerobic conditions within CWs [86], since during the resting period oxygen can be conveyed through the CW medium via convention and diffusion, which mainly depend on the gradient air pressure produced by the effluent water and the effective resting time, respectively. Furthermore, the intermittent feeding mode might be useful to prevent operational issues such as clogging by regulating the equilibrium of the bacteria that are present [87].

Several hydraulic properties, including the hydraulic loading rate (HLR) and hydraulic retention time (HRT), can influence the remediation effects of CWs in terms of N removal. Usually, a higher HLR involves a lower HRT and implies a reduction in the N removal efficiency [83]. Therefore, during the design phase of the CW, either HLR should be selected in order to allow a suitable HRT, or the geometric characteristics of the CW should be adjusted to increase the volume capacity, resulting in a higher HRT. In this sense, the maintenance and operation periods are very important factors, since it has been shown that the long-term operation of CWs can lead to substrate clogging and the presence of of preferential flows, which could reduce HRT [88].

Although the presence of bacteria above certain values could also cause clogging issues [89], N removal in CWs can be enhanced by promoting the microbial activity in soils. Comparing different types of CWs, the total activity of microbial communities is largely in the same range, but the growth of heterotrophic bacteria, commonly responsible for denitrification, has usually been higher in VF-SSF CWs than in SF CWs [90]. pH is an influencing factor that can be modulated in order to favor microbial growth [91]. Furthermore, temperature can play a regulatory role in the activation/deactivation of various microbially-mediated processes in CWs. In particular, it has been observed that denitrification can cease at 5 °C, since only reduced activities have been detected at temperatures lower than 5 °C, and in any case these have shown very low rates [92]. Although the minimal temperature for stable nitrification is 10 °C, low nitrification rates at lower temperatures might be compensated for by an increased HRT, without affecting overall N removal [93].

3.2. Buffer Strips

Buffer strips are considered to be best-management practices, since they exploit green processes and structures to intercept contaminated agricultural water streams and prevent the pollution of aquatic environments. In addition to the term "buffer strips", several expressions can be used to indicate the type of conservation buffer, such as "buffer zone", "vegetative filter strips", "grass filters", "filter strips", and "vegetative buffer strips". These natural systems are characterized by the presence of herbaceous or tree vegetation adjacent to the agricultural land or near the receiving water bodies [20,94].

Buffer strips have an important ecological role in reducing nutrient, pollutant, and sediment concentrations in runoff streams from agricultural fields [71,95,96], decreasing gully erosion phenomena [97], promoting carbon sequestration [98,99], and providing a source for biomass production [100]. Buffer strips can be composed of either pre-existent native vegetation or artificially implanted vegetation, especially for areas where original spontaneous vegetation is absent [20]. Both surface runoff and sub-surface drainage flows might contribute to N transport within catchment basins, and therefore, when designing a buffer strip, it should be ascertained that these systems are not by-passed, thus avoiding the direct discharge of agricultural polluted waters into the streams [101].

Plants can reduce flow velocities, providing resistance to water flow, and also causing the deposition of suspended particulates, to which N may be bound. Buffer strips can contribute to the N transport reduction, mainly through plant uptake and other removal mechanisms (i.e., ammonification, nitrification-denitrification, ammonia adsorption, and sedimentation), basically following the principles that were described at the beginning of Section 2 [102].

Several studies on N removal in buffer strips have demonstrated that denitrification is probably more efficient in N removal than plant uptake because the latter can remove N only temporarily, allowing its availability again once it has been mineralized [103]. On the contrary, denitrification can permanently remove N from the soil and transfer it to the atmosphere. However, plants can accumulate N in perennial tissue, leading to a lag time between plant N uptake and N release by decomposition. Additionally, N can be immobilized in litter during the decomposition of plants, which can generate a temporary reduction of inorganic N in water. Decomposition is very important in winter because it can compensate for the lowest denitrification activity due to low temperature and coincide with the period with the highest runoff and discharge of agricultural drainage water which is rich in nutrients [100].

Similarly to CWs, the literature on buffer strips reports a wide variability in their treatment performance in terms of N removal. Buffer strips have been observed to be capable of removing N from 5 up to 130 kg N ha^{-1} year^{-1} [104]. This variability mainly depends on the width (which can be even more than 200 m) of the system, but it is also correlated with the other factors analyzed in the following Sections 3.2.1 and 3.2.2.

3.2.1. Vegetation Typology

The efficacy of a buffer strip can also depend on the careful selection of the plant species to be used for its settlement, especially considering the functions required for these species. A strategy to enhance the efficacy of buffer strips is to use a combination of different plant species with diverse growth rates and lifetimes, either directly or in succession so as to have more chances for plant or species survival and to promote vegetation diversity [105]. This was confirmed in a study by Schultz et al. [106], in which the effectiveness of buffer strips was enhanced by planting three different zones with diverse vegetation, each parallel to the stream. The zone closest to the stream was planted with trees (four to five rows), in the middle zone there were shrubs (one or two rows), and furthest from the stream, next to cropland, there was a native warm-season grass. Trees near the stream (namely, silver maple, willow, cottonwood, green ash, and box elder) were selected since the quick development of deep roots was required to increase bank stability. For the mentioned zones, the contribution related to the adoption of each single type of plant was investigated, confirming that some plants showed a greater capability to remove contaminants. In the warm-grass season zone it was observed that the selection of the appropriate width was important for the mitigation of issues related to surface runoff events from agricultural fields, promoting water infiltration and the deposition of transported sediments. On the contrary, other permanent warm-season grasses such as Indian grass, big bluestem, and little bluestem can be used in cases where surface runoff is not so relevant. Cool-season grasses, such as brome and fescue, have been observed not to be useful because they did not improve soil quality and did not remain upright under the flow of water [106].

To reduce the runoff of contaminants, in an area not adjacent to water bodies, through the infiltration of water into the soil, it is preferable to resort to a purely herbaceous cover. Shrubs or trees are preferred when the strip is near a water body [107]. The herbaceous species that show high sediment retention and best exhibit valid resistance to surface runoff are grasses with robust culms and rigid leaves, which show a quick settlement capability, long persistence over time, high regrowth capacity after each cut, uniformity of vegetation, and a good ability to attract insects. To limit surface runoff, it is better to plant trees and shrubs that allow a good development of the underlying herbaceous cover. To act as an anti-drift barrier or with a windbreak function, plants with an adequate optical porosity of their foliage are used. Shrub and tree bands can also constitute important ecological networks for natural fauna, as well as being elements of high landscape value [107].

Furthermore, the distance of the plants is another important factor to consider, since some of them need the support of other plants or species for their growth [105]. Before creating a buffer strip, it is also important to establish the time required by plants to generate dense and vigorous vegetation in order to obtain the necessary efficiency and to maximize the pollutant-trapping capacity of the buffer strip [102].

Different types of vegetation can have different effects on N removal from polluted agricultural water streams, and in particular on the denitrification process. Moreover, young forests, bushes, and wet grasslands have been observed to be more successful in N removal because their need for nutrients for the growth phase and their microbiological activity are more intensive [101]. On the contrary, the amount of nutrients required by forested buffer strips is lower (since their growth is limited), and thus higher NO_3^- concentrations are more likely to infiltrate soils and cause the eventual pollution of groundwater.

3.2.2. Influence of Dimensional Parameters and Operating Conditions

The suitable design of buffer strips involves the definition of several aspects that can influence the efficiency of sediment and nutrient removal. Physical features such as extension (length and width) are among the first factors to be evaluated for the correct implementation. Buffer strips are typically installed with a fixed width, although it could be useful to find better design solutions where variable widths can be considered, since a complex and varied landscape topography could lead to an uneven nutrient loading distribution [102]. Parkyn et al. [101] have reviewed several studies in which methods were provided by researchers for the correct design of buffer strips. The reviewed studies revealed that an enhancement of sediment and nutrient removal rates can be provided by increasing the width of the buffer strips from 4.6 to 27 m. Furthermore, many researchers have highlighted the capacity of grassed buffer strips to trap sediment, with the width of buffer strips ranging between 5 and 10 m, whereas for larger widths, on the order of 20–30 m, removal efficiencies up to 100% were achieved. Similar results have been observed in terms of NO_3^- removal in forested buffer strips with a width of 10 m, which have shown a NO_3^- retention of about 70%. Based on these considerations, it could be stated that the minimum width should not be lower than 10–20 m in order to ensure the removal of nutrients and, in general, of other contaminants that could affect the quality of receiving water bodies.

Another important parameter which might influence the efficiency of buffer strips is their slope, in cases where these systems are implemented in non-flat areas. Several studies have concluded that in buffer strips with higher slopes, it is necessary to increase the extension of the buffer strips, since higher flow rates entering the natural systems can be detected, involving larger areas required to decrease runoff flow velocities [101,102].

When designing a buffer strip, soil characterization should be carried out. Indeed, the various types of soil can be characterized by different initial water contents, hydraulic conductivities, and infiltration capacities. Free draining soils can reduce sediment and nutrient access to buffer strips because of a decrease in runoff rates, whereas these NBS can be completely bypassed in the case of increased infiltration.

The particle size of sediments, the fall velocity, sediment density, and grass spacing may affect the resistance to overland flow, and thus lead to different trapping efficiencies. Buffer strips show better results in filtering sediment, nutrients, and pollutants in rolling land than in steep hilly terrain, as steep slopes cause high flow velocities and their buffer effectiveness is minimal or patchy at best along the stream length [101].

Buffer strips can have a variable lifetime, particularly when trapping and filtration processes are successful. To preserve saturation and clogging phenomena in buffer strips, there are several strategies to ensure the continuous removal of nutrients. Some of those are periodic harvesting of plants, the selection of appropriate buffer widths, and the implementation of land practices that can reduce the intake of nutrients.

Seasonal variability in nutrient loads can be also a factor influencing the effectiveness of buffer strips for N removal. Osborne et al. [108] have shown that significant seasonal differences may exist, for instance, in grassed and forested buffer strips. Agricultural drainage water can be rich in nutrients, but their concentration depends on agricultural land-use activity. Normally, the year can be divided into three periods, (i) the dormant season (1 November–15 April), (ii) the planting season (16 April–30 May), and (iii) the growing season (1 June–31 October) [108]. The authors observed that higher nutrient concentrations occurred in riparian grass vegetated buffer strips' outflows during the planting season than during the dormant season, whereas no seasonal variability was detected in nutrient concentrations in the forest. Additionally, groundwater nutrient concentrations varied during the year, with the highest nutrient percentages detected during the planting season, probably due to the application of fertilizers. Meanwhile, no significant seasonal variation occurred in NO_3^- concentrations, although denitrification was higher during winter. These results therefore suggest that on an annual basis, grassed buffer strips were less efficient than forested buffer strips [108].

3.3. Other Types of NBSs

As stated previously, the literature review showed that researchers focused less on other NBS types. Of these, vegetated channels and water sediment control basins were considered to be those with more information available, and therefore they are briefly discussed in the following two sub-sections.

3.3.1. Vegetated Channels

Vegetated channels (also known as "grassed waterways", "drainage ditches", etc.) are integral parts of agricultural lands and can be implemented to intercept nutrient loading (including N) from runoff or tile drainage water. The ability of vegetated channels to intercept water polluted by N is related to several design characteristics, such as the location of the channels within the watershed; their length, cross section, slope, sidewall roughness; and their orientation with regard to the slope [19]. In these man-made ecosystems, floating and submerged vegetation plays a key role since it favors N filtration, increases HRT and reduces flow velocity, oxygenizes soil, increases bed stability, maintains aquatic ecosystems, and provides surfaces for microbial biofilms [109]. However, an excess of vegetation can cause flow blockages and subsequently may result in flooding and waterlogging. Hence, it is important to plan periodic maintenance activities to remove excessive vegetation. For this purpose, low-cost and eco-friendly procedures should be performed. Recently, so called "two-stage" ditches have been introduced [110,111]. These systems involve adjustments of a conventional and trapezoidal vegetated channels in order to better reproduce the conformation of a natural stream by means of the inclusion of adjacent floodplains or benches, aiming thus to extend the time of interaction between the different environmental matrix (namely, water, bench vegetation, and bench soil), and favoring higher N uptake and denitrification rates.

The available data on N removal in vegetated channels are scarce. In addition, the few studies that have studied this issue have reported values that vary greatly. For example, the current literature reports values ranging from 150–4300 kg ha^{-1} year^{-1} [112,113]. The variability in N removal can be associated with several factors. The water temperature and geometric components (length, cross section, slope, sidewall roughness and orientation with regard to the slope, etc.) certainly affect these results greatly. However, seasonal differences can also influence the effectiveness of vegetated channels, as N assimilated by plants may be released back into the channels due to senescence in the dormant season [114]. Furthermore, plant uptake of N is strictly related to vegetation growth [114]. In addition, the concentration of N in water can influence removal performance, considering also that vegetated channels are commonly subject to intermittent flows (thus different HLR values) linked to rainfall frequency and intensity or irrigation.

3.3.2. Water Sediment Control Basins

Water sediment control basins are natural systems that are usually implemented near sloping agricultural lands. They can divert the outflow coming from agricultural fields, hold the water temporarily, and release it later through a drainage channel or through infiltration into the soil. In this way, a reduction in erosive land flows can be detected, also resulting in the containment of sediments and nutrients [115–117]. Dry retention basins are the most common typology used. In these systems, the presence of grass-lined depressions or basins obtained from excavations can channel the outflow, causing a slow-down type filtration and therefore an increase in sediment and nutrient interception by means of vegetation. Bio-retention basins represent another typology that can be created through excavation and the subsequent filling of the obtained space with suitable soil, in combination with fertilizer and vegetation. These systems, in addition to ensuring more uniformly distributed filtration over time, can also create the ideal conditions for the establishment of biological and biochemical reactions in the soil matrix and around the root zones of plants, thereby allowing the increased removal of nutrients. Only a few studies on the implementation of water sediment control basins have been reported in the literature. However, it seems that although these natural systems are very effective in limiting sediment transport, their capacity for N removal is much lower [22].

4. Nitrogen Recovery Strategies

Many researchers are currently investigating possible strategies for the recovery of N and nutrients in general after they have been removed from water by means of NBS. In this section we therefore report the most recent and innovative approaches in this area.

In addition to their removal capability, plants might be also exploited for nutrient recovery through several approaches. For instance, aquatic plants can be harvested and sold as a source of nutrients to feed fish and animals [118], and macrophytes can be potentially added into human food supplements and cosmetics and used in the extraction of high-value cellular components [119]. Duckweed, which includes *Lemna sp.*, *Spirodela sp.*, and *Wolffia sp.*, has been commonly used in wastewater treatment systems as it grows successfully on wastewater and efficiently removes nutrients that are directly adsorbed from water, not via a central system but through each of its fronds. These plants do not have a structural tissue. Their lignin and cellulose content are around 2.7% and 10.0% [120], whereas the fiber content is lower. Their high capacity to assimilate nutrients results in a highly nutritional plant, with an elevated concentration of protein, which makes duckweed an optimal source of food for animals. In addition, duckweed could be used as a fertilizer in agriculture, and this is even more feasible since it can be harvested in a straightforward fashion [120].

N accumulation can also occur by means of algae (microalgae and macroalgae). Floating algae are commonly used in ponds and can remove and accumulate nutrients from polluted streams [121]. In the context of nutrient recovery, microalgae have been explored for several uses, such as in the production of biofuels. They can also be involved in animal diets and as a supplementary food resource, due to their high content of proteins, carbohydrates, nutrients, lipids, vitamins, minerals, antioxidants, and other valuable components. However, since the recovery of nutrients in microalgae can induce the biomass growth of algae themselves, microalgae are further used as biofertilizers, biopesticides, and biostimulants in the agricultural sector [121]. Another difficulty in the use of microalgae is due to their small size, which can make their harvesting procedure problematic. In this context, researchers are still working on the development of advanced strategies, which could lead to highly valued by-products [122]. For example, the immobilization of algae can provide a suitable approach to overcome the harvesting issue [123]. This process consists of cell attachment and/or cell inclusion to or into a carrier (biopolymers and synthetic compounds (alginate, agar, polyacrylamide, silica, zeolite, etc.)) as a support mechanism. Although immobilization has been proven to lead to some advantages, such as higher N uptake rates compared to free algal cells, there is also evidence that algal behavior, growth rate, and biomass productivity are negatively influenced by the immobilized bacteria [124].

The employment of compost is another strategy to recover nutrients that are retained by plants. The composting process involves the conversion of organic waste into a biological by-product that can be used as a soil conditioner or organic fertilizer. In this process, biochemical reactions are exploited by microorganisms to degrade the organic matter, which is used as a substrate for their growth. Several types of plant waste may be composted, such as leaves and branches of trees and fruit skins, alone or together with sewage sludge, pig manure, and organic food [125]. Considering that composting is a very common method nowadays, the involvement of plant waste or aquatic plants in this process as a primary source seems to be a promising and sustainable strategy for N recovery from agricultural drainage waters.

Numerous studies have focused on the composting of harvested biomass from CWs. The plant biomass from CWs is usually not composted alone. Sultana et al. [126] co-composted *Phragmites austraulis* from an HF-SSF system with olive mill wastes, resulting in the production of a final product that met EU regulations for organic farming applications [126]. Kouki et al. [127] investigated the potential use of sewage sludge co-composted with plants to be used in CWs as a carbon source, resulting in the production of a suitable compost for impoverished soils. Furthermore, a mixture of fungi, actinomycetes, and bacteria added to the compost was reported by Song et al. [128] to increase both the cellulose and lignin contents. In their study, the authors used *Typha angustifolia* and *Phragmites australis* with microorganisms and sewage sludge for the generation of compost to be used as a fertilizer with a very high N content.

Biochar derived from agricultural biomass waste has been increasingly recognized as a multifunctional material for agricultural and environmental applications. It is the by-product of biomass pyrolysis in the partial or complete absence of oxygen. Different plants and their parts, as well as animal, human, and industrial waste, can be used for biochar production [129]. As previously reported in Section 3.1.2, biochar has been investigated as a novel substrate in CWs, alone or in combination with other conventional substrates, and it has been observed to promote high nutrient removal percentages due to its porous structure that enhances microbial activity. Biochar plays an important role in nutrient recovery. Several studies have demonstrated its ability to recover nutrients from polluted aqueous solutions and the possibility of its employment as a soil conditioner or fertilizer in soils [130]. The production of biochar from waste biomass, including that from NBSs, might provide a solution to disposal issues, avoiding the burning/chopping of residues and the incorporation of residues in soils, which generate a negative effect in terms of air pollution through the emission of greenhouse gases [130].

Anaerobic digestion is a biological process used to manage organic waste materials, with the main advantage of producing energy in the form of biogas in addition to a stabilized residue, which can be therefore reused in agriculture [131]. For this reason, anaerobic digestion is nowadays the most used method in WWTPs to treat solubilized organic biomass [132,133]. Different biogas yields have been produced through anaerobic digestion, depending on type of the organic materials and the biomass employed for the production. Roj-Rojewski et al. [134] have explored the possibility of producing biogas and therefore methane from CW biomass by means of anaerobic digestion. They used different harvested plants, namely, *Phalaris arundinacea, Phragmites australis, Glyceria maxima,* and *Carex elata Carex lasiocarpa*, as well as an inoculum from a post-digestion tank of a mesophilic biogas plant, treating maize silage with swine manure and chicken droppings. Despite the optimized operating conditions used in the CWs, the obtained methane yields were lower than the ones detected using crops such as maize and grassland as substrates. However, *Phragmites australis* has been observed to be the most efficient CW plant in terms of methane production, probably due to its higher size compared with other CW plants. Therefore, although the biogas yield using CW plants is lower, their use as substrates or co-substrates in agricultural biogas plants remains a possible energy recovery strategy [134].

In addition to biogas production, the main by-product of the anaerobic digestion process is digestate, in which the liquid and solid phases can be distinguished. The latter, rich in residual fibers and P, is easier to manage with respect to the liquid fraction, which, on the contrary, contains a large portion of N and K. Therefore, digestate could be used as fertilizer and, more appropriately, as a soil conditioner because it can provide nutrients and microelements in available forms to plants, also increasing the microbiological activity in the soil and thus favoring soil fertility, although attention must be paid to the concentrations of other emerging contaminants [135,136].

5. Conclusions

In recent decades, nitrogen has been increasingly applied to agricultural soils in the form of fertilizer, leading to an intensification of crop production. However, the use of amounts higher than those needed by crops has led to the introduction of nitrogen into water environments, which currently represents a threat to ecosystem health and biodiversity.

In this review, several nature-based solutions (NBSs), namely, constructed wetlands, buffer strips, vegetated channels, and water sediment control basins, have been investigated as environmentally-friendly and sustainable technologies to treat agricultural runoff and drainage water, with the main aim of removing nitrogen from contaminated water.

This work represents the first attempt to contribute to the wider application of NBSs in the agricultural sector. The information summarized by this study can be used as a starting point for future research that should concentrate on better understanding and providing a clearer definition of the nitrogen removal processes, as well as performing experimental comparisons of the NBSs considered for the relevant environment.

In this study, we concluded that the removal of nitrogen occurs through a combination of physical, chemical, and biological processes. Although most of the literature consulted has shown that NBSs are very efficient in terms of water decontamination, there are still certain gaps (e.g., the complete understanding of these processes) in the current knowledge of their fundamental removal mechanisms. Furthermore, the design of successful NBSs for water purification is not an easy task since their effectiveness has been observed to be dependent on several factors and operating conditions, which can deeply influence the natural purifying functions associated with vegetation and microbial communities.

Finally, various studies have referred to several strategies for nitrogen recovery, which may lead to a reduction of the depletion of non-renewable resources and the environmental impact linked to their extraction and manufacture, promoting the concept of a circular economy.

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