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Recent advances in control technologies for non-point source pollution with nitrogen and phosphorous from agricultural runoff: current practices and future prospects

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Abstract

Eutrophication of natural water is a universal problem. Nitrogen (N) and phosphorus (P) from agricultural runoff are the main sources of nutrient input, provided that emissions from industrial point sources (IPS) are under control. Therefore, it is of great environmental importance to reduce pollution associated with agricultural runoff as a means of regulating eutrophication levels in natural water. Numerous methods proposed for treating agricultural runoff can be classified into three categories: source control, process control, and end treatment. In this review, major technologies for N and P control from agricultural runoff are summarized along with discussion of newly proposed technologies such as biochar biomimetics and microbial catalyst. Because agricultural runoff (from farmlands to receiving waters) is a complicated pollution process, it is difficult to regulate the nutrients discharged via such process. This review will thus offer a comprehensive understanding on the overall process of agricultural runoff and eutrophication to help establish control strategies against highly complicated agricultural non-point sources.

Keywords: Eutrophication, Phytoremediation, Charcoal, Black carbon, Soil organic matter, Water and wastewater treatment, Sustainable development goals

Introduction

Water is a very important resource for human survival and development. Environmental pollution is the greatest challenge in maintaining safe water sources [1–3]. In recent decades, various technologies were developed to treat industrial effluent and domestic sewage, among others [4–7].

Agricultural non-point source pollution has long been considered an important factor affecting the level of eutrophication [8–10]. For example, agricultural non-point source pollution is estimated to be responsible for

52 and 54% of the total loading of nitrogen (TN) and phosphorus (TP), respectively in Taihu Lake Basin, China [11]. Likewise, they are also found to represent 24% and 71%, respectively in Italy [12]. In USA, agricultural non-point source pollution is considered the dominant source of nutrients in lakes and streams [13]. Appropriate management of agricultural runoff and animal waste is a large concern for the U.S. Environmental Protection Agency (USEPA) and U.S. Department of Agriculture (USDA).

Agricultural runoff is the surface runoff from farmland outflow, which comes from the farmland's surplus water. Its main sources of excess water are from irrigation and rainfall [14]. Agricultural runoff has complex pollutant compositions including nitrates, ammonium, phosphorus compounds, heavy metals, and persistent organic pollutants. N and P, being essential elements in

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amino acids and genetic material, respectively, are vital to the growth of aquatic plants as the key limiting nutrients during eutrophication [15].

Anthropogenic eutrophication has become the primary problem as it is often recognized to have strong potentials to affect the health and security of aquatic ecosystems in the world. The large “cyanobacteria mat” in Taihu Lake (Fig. 1) caused the closing of a drinking water plant in Wuxi, leading to a crisis affecting millions of people [16, 17]. At the same time, continuous input of heavy metals and persistent organic pollutants (POPs) from agricultural runoffs can easily accumulate in organisms to pose various health risks (e.g., pollution of drinking water). Therefore, it is of considerable interest to adequately decrease agricultural non-point source pollution to control eutrophication in lakes and rivers, to protect the water environment, and to secure drinking water quality.

There are three main control strategies for agricultural runoff pollution: source control, process control, and end treatment. Source control works to reduce the application of N and P as well as leaching, such as conservation tillage, fertilization management, and water-saving irrigation [18–20]. Process control aims to eliminate the pollutants by using the space and time of agricultural runoffs from the field to the receiving water, such as ecological ditches [21]. They are usually set in the agricultural ditches. End treatment is the last choice to avoid the damage of the receiving water, if the pollutants does not fall below the safe value [22]. The large storage capacity provides more time for the treatment of agricultural runoffs. Although each approach is based on different principles, they serve to control agricultural runoff pollution to varying degrees. It is difficult to find efforts to integrate the diverse treatment options from source to end. In this



Fig. 1 Cyanobacteria outbreak in Taihu Lake, China

review, we highlight current mainstream technologies along with some promising alternatives. A scenario analysis based on the reference data was also made to provide a comprehensive understanding of the current control techniques for agricultural runoff and their roles in effective control of agricultural runoff.

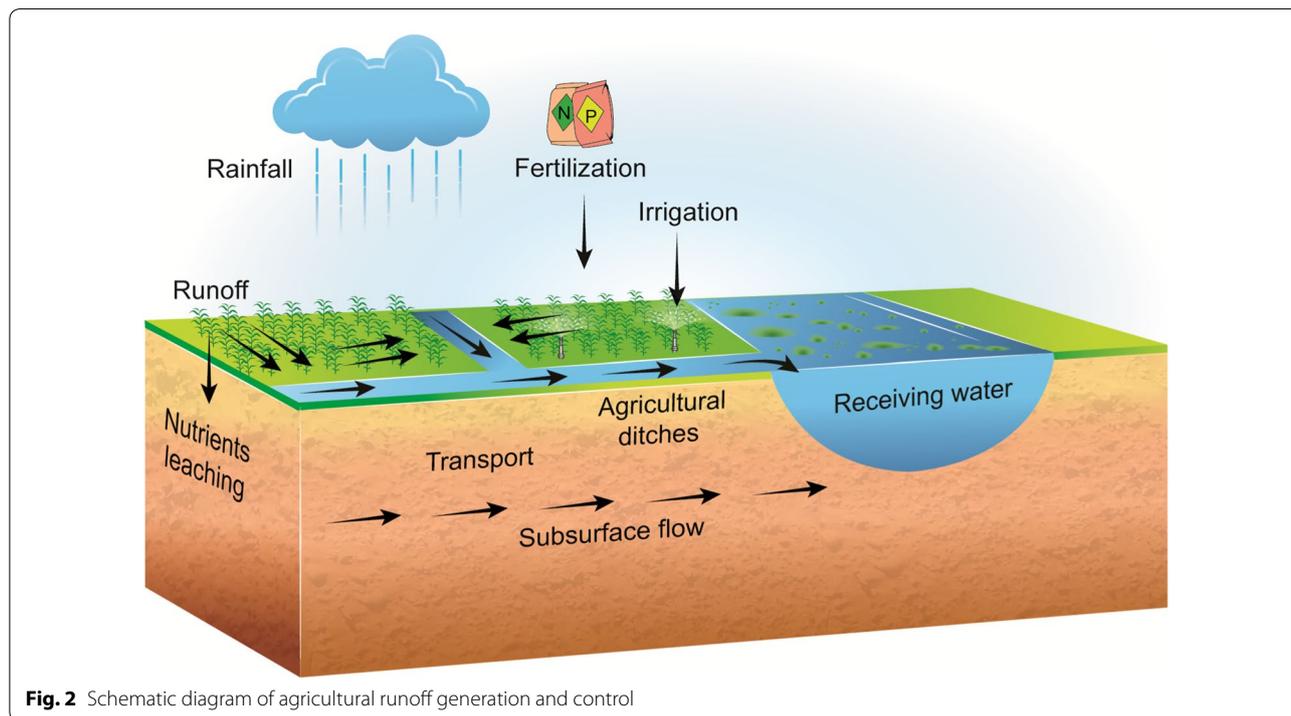
Pollution status of agricultural runoff

Agriculture supports the construction and development of a national economy. It is of particular importance to the most populated countries, such as China, India, and Indonesia [23]. Due to the great demand for food, the use of chemical fertilizers and pesticides has become indispensable over the past decades [24]. The N and P fertilizers have been used most widely in the world. According to statistical data from Food and Agriculture Organization of the United Nations (FAO, Table 1), as of 2015 the world's average use of N fertilizer per cropland area has reached 68.6 kg/ha, and 30.1 kg/ha for P. The USA is still increasing their use of fertilizers. Moreover, China is the largest producer and consumer of fertilizers. Overuse of chemical fertilizers leads to various environmental problems including surface water eutrophication, N-related greenhouse gas emissions, and groundwater pollution [25–27]. Although the application of fertilizer is made to the farmland, the transport of excess N and P takes place by surface water runoff after rainfall and irrigation events. As shown in Fig. 2, the N and P migration process increases the complexity of the whole system, while providing temporal and spatial conditions for effective remediation. As the main component of an agricultural irrigation system, ditches can act as the major pathway of farmland surface runoff. Since agricultural runoff undergoes a certain amount of migration time before discharging to the receiving water, ditches can be an ideal place for controlling on N and P [28].

The diffusivity of N and P differs greatly in soils. Cookson et al. have reported that the diffusion coefficient of H_2PO_4^- in soils was only one thousandth of that of NO_3^- to affect the rate of runoff losses in N and P [29]. In a cropland fertilized with 196 kg N ha^{-1} year $^{-1}$ and 87 kg P ha^{-1} year $^{-1}$, N and P fertilizer runoff loss rates were 9.5% and 3.3%, respectively [30]. In contrast, in paddy soils fertilized with 210 kg N ha^{-1} year $^{-1}$ and 36 kg P ha^{-1} year $^{-1}$, N and P fertilizer runoff loss rates were 5.9% and 0.52%, respectively [31]. In this case, the N and P discharge rates were estimated as 12.39 and 0.18 kg ha^{-1} year $^{-1}$, respectively. Although the loss load of nutrients varies little from year to year, it varies greatly from month to month. For instance, the highest N and P loss concentrations took place over April, June, July, and August in China, which correspond to the high-risk eutrophication period [32]. The unevenness of time distribution of loss load

Table 1 Fertilizer application levels worldwide and by country (data from FAO)

Number	Region	N fertilizer application level (kg/ha/year)			P fertilizer application level (kg/ha/year)		
		2005	2010	2015	2005	2010	2015
1	World	57.55	65.13	68.61	24.98	27.75	30.10
2	USA	65.63	69.75	77.46	24.56	21.21	26.82
3	China	213.5	241.92	228.48	94.73	115.27	116.4
4	India	74.99	97.21	102.51	30.71	48.43	41.18
5	Indonesia	59.05	62.43	61.27	8.03	11.19	17.11
6	Japan	117.98	97.86	79.87	130.25	92.38	76.78
7	Netherlands	244.35	205.82	203.11	42.55	29.02	12.22
8	Spain	51.77	54.65	62.54	28.77	19.55	24.16
9	Thailand	55.43	79.21	80.74	17.12	24.11	16.39
10	Argentina	18.89	19.79	14.52	15.14	17.54	11.62
11	Australia	19.14	22.85	28.04	20.94	19.17	19.98
12	Brazil	27.11	47.36	44.23	37.57	43.58	52.65



greatly increases the difficulty of controlling nutrient loss from agricultural runoff.

As a major form of non-point source agricultural pollution, continuous N and P input leads to their accumulation in the receiving water. Excessive N and P accumulation causes various problems such as algal blooms, water degradation, fish kills, and loss of biodiversity [33]. Due to the lack of effective control on agricultural non-point pollution, N and P pollution has become

a global problem. In case of USA, over-enrichment issues of N and P were observed from about 50% of impaired lake areas and 60% of impaired river reaches [34]. In China, over half of the major lakes are eutrophic while nearly three quarters are continuously deteriorating [35]. Even in Canada, the deterioration of lake Winnipeg is also attributed to excessive N and P nutrient enrichment [36]. Therefore, controlling N and P from agricultural runoff is urgent.

Research progress in source control technologies

Dissolved pesticides, nutrients, and sediments in agricultural runoff cause various problems, including persistent organic translocation, nutrient loss, and soil erosion [37]. Reasonable tillage practices can significantly improve surface roughness and reduce surface runoff, thus reducing runoff emissions and pollution load at the source. As a food staple for 1/3 of the world's population, rice is planted over an area of more than 164 million hectares. Rice requires a great deal of water, which leads to massive agricultural runoff [38]. The dissolved N, P and sediments create a huge pollution load on the surrounding waters [39–41].

Conservation tillage

Although, tillage inevitably disturbs the soil surface, conservation tillage methods (such as reduced tillage and no-tillage) play significant roles in protecting soil from erosion [42]. In addition, conservation tillage improves soil structure and increase organic matter content, which can increase the infiltration to runoff ratio and reduce evaporation [43, 44]. Reduced tillage and no-tillage are both effective methods of conservation tillage. For example, Clausen et al. studied tillage effects on runoff for croplands in Vermont, USA, and found that reduced tillage reduced runoff by 64% [45]. Liang et al. reported that runoff volume from rice-planting watersheds was reduced by 25.9% using no-tillage techniques [46]. Reduced tillage and no-tillage reduce the intensity of tillage practices as well as the impact of rain by the protection of soil surface using crop residues. In recent years, land covers and soil amendments such as biochar, which enhance the soil structure and porosity, are used to protect the soil [47, 48]. Won et al. used rice straw, polyacrylamide, and gypsum to treat with Chinese cabbage field, which resulted in reduction of suspended solids and of total nitrogen (TN) by 86.6% and 34.7%, respectively [49]. Lee et al. have studied the effects of soil amendments on soil loss [50]. Accordingly, the field soils amended with biochar and polyacrylamide reduced soil loss by 70.4% in a 33 mm day⁻¹ natural rainfall, while there was no difference in runoff. Lee et al. found that field soils treated with 4% wood biochar significantly decreased runoff by 16.8% and inorganic N by 41.8% [51]. Biochar is often used in soil remediation, and it also has great potential in agricultural runoff control. The effects of biochar on soil structure and nutrient fixation are worth further studies [52, 53].

Rotation tillage

Conservation tillage is effective for reducing dissolved N in the runoff [54]. However, conservation tillage practices

will inevitably lead to soil compaction during long-term operation, which will lead to P accumulation on the soil surface, and as a consequence, an increase in the runoff loss of P. Tiessen et al. reported that conservation tillage in the Canadian prairies reduced the TN concentration by 41% while the total phosphorus (TP) increased by 42% [36]. Rotation tillage is another choice to control nutrients loss in agricultural runoff. Liu et al. converted conservation tillage to rotation tillage and found that rotation tillage was a better option to decrease various types of P (e.g., either contained in surface soil or released from crop residue) as well as runoff duration [54]. As a result, total dissolved P (TDP) and TP decreased by 46% and 38%, respectively. It is because tillage practices would alleviate soil compaction and decrease P accumulation in surface soil. Crop residues in conservation tillage would capture more water that leads to greater runoff duration time. Therefore, rotation tillage could shorten the contact time between crop residues and surface runoff that reduces the P released from crop residues. Daverede et al. compared P runoff after no-tillage and chisel plow farming practices and found that the latter could reduce the dissolved reactive P load by 60% [55]. Therefore, the selection of tillage practice should be based on local climatic conditions, soil conditions, crops, and dominant eutrophication nutrients.

Fertilization management

Fertilization management is another effective source control method that has been used widely [56, 57]. Fertilizers containing N and P are commonly used in the agricultural industry. N-fertilizer efficiency varies from crop to crop. For example, mean N-fertilizer efficiencies of maize, wheat, and rice are 37%, 18%, and 31%, respectively [58]. In order to fully reflect the global nitrogen use efficiency (NUE), Table 2 listed the world fertilizer N consumption for cereals, N removal in cereals, and estimated nitrogen use efficiency. The estimated NUE is 36%. Once surface runoff is formed, excessive N and P would flow to the receiving water. Therefore, it is critical to deliberately manage fertilizer application. One example of fertilization management is deep placement of fertilizers to lower the risk of discharging N into a body of water. In the Taihu Lake region, it was found that using urea deep placement lowered N loss by 50% in the paddy field [59]. Fertilizer band placement and hole placement can reduce total N loss by 63.6% and 77%, respectively, and total P loss by 42.8% and 53.8%, respectively [60, 61]. This is because band placement can reduce contact with soil microorganisms and slow the nitrification process. Zeng et al. studied the impact of fertilization depth on TN loss [62]. These authors found that a 20 cm fertilization depth reduced TN and TP by 36.2% and 31.4%, respectively

Table 2 World fertilizer N consumption, cereal production, and N use efficiency

Commodities and computations	Amount/ton	Variable
World fertilizer N consumption, 2015 (FAOSTAT)		
Total	108,699,171	
Cereal ^a	65,219,502	C
World cereal production, 2015 (FAOSTAT)		
Barley	147,413,603	
Maize	1,052,097,073	
Millet	28,218,225	
Oats	23,328,079	
Rice	745,337,946	
Rye	13,755,752	
Sorghum	66,006,062	
Wheat	751,863,360	
Total	2,828,020,100	P
World cereal grain N removal, 2015 (Fujihara et al. 2008) ^b		
Barley (N = 21.4 g/kg)	3,154,651	
Maize (N = 13.1 g/kg)	13,782,472	
Millet (N = 20.1 g/kg)	567,186	
Oats (N = 19.1 g/kg)	445,566	
Rice (N = 10.5 g/kg)	7,826,048	
Rye (N = 16.3 g/kg)	224,218	
Sorghum (N = 19.9 g/kg)	1,313,521	
Wheat (N = 26.2 g/kg)	19,698,820	
Total	47,012,482	N
Cereal grain N from soil and rainfall, 2015 ^c		
Nitrogen use efficiency	23,506,241	S
NUE = [(N - S)/C] × 100 = 36%		

^a Fertilizer consumption of cereals calculated from average fertilizer application and cereal acreage in the world

^b Cereal grain N values obtained from the report by Fujihara et al. [110]

^c Cereal grain N from soil and rainfall = N × 0.5 [111]

compared to surface fertilization. Controlled-release of fertilizer is another choice that can lead to slow release of N and P to be adapted to the rate of crop growth while improving nutrient utilization efficiency [63]. Tan et al. have studied the effect of fertilization treatment on N loss in a wheat–maize rotation system [64]. Accordingly, the results indicated that controlled-release N fertilizer performed best in reducing inorganic N concentration in runoff. Controlled-release P fertilizer can reduce P loss by 62% in paddy systems and by 33% loss in corn systems [65]. Optimization of fertilizer timing and application rate is also important variables to control nutrient loss [66]. Because the losses show seasonal characteristics, with higher nutrient loading in summer and autumn. As for rainfall process, nitrate-N loss increased gradually along with ammonia-N loss decreased. Based on these

characteristics, model-based analysis has also been proposed for long-term effects of fertilization management [67].

Water-saving irrigation

Heavy precipitation and field drainage systems can drive surface runoff. During the rice growing season, which is coupled with the rainy season, surface runoff accounts for 86% of cumulative N losses [68]. This is because conventional flooding irrigation (CFI) keeps a high floodwater level in the fields. Water-saving irrigation (WSI) techniques could significantly reduce floodwater levels, improving the buffering capacity of the fields to help reduce runoff and nutrient losses. Furthermore, WSI enhanced root growth with getting more grain yield compared to CFI [69]. Alternate Wetting and Drying (AWD) irrigation has also been employed widely to reduce water inputs and enhance water use efficiency in the rice cropping systems [70–72]. The AWD irrigation was seen to reduce surface runoff by 30.2–36.7% compared to conventional practices [73]. The concentrations of nutrients, however, do not decrease with the decrease of surface runoff if AWD is applied alone. Because the contact time between water and soils will not be decreased. Thus, it is better to integrate irrigation management with tilling practices and fertilization management.

All the above source control techniques can effectively reduce surface runoff and nutrient concentrations. Nevertheless, they cannot prevent runoff from flowing into the receiving water. The concentrations of N and P in agricultural runoff have decreased significantly by source control techniques. However, it is still difficult to achieve the safe discharge concentrations. Because long-term accumulation of nutrients in receiving waters will also increase the risk of eutrophication. Therefore, complete treatment of agricultural runoff still needs additional process control and end treatment technologies.

Research progress in process control technologies

Process control technologies aim to remove pollutants during agricultural runoff transport. Ecological ditches are engineered based on the widely distributed ditches surrounding farmlands. Before the nutrients are discharged into receiving waters, the ditches can reduce pollutants in the runoff by effectively using a similar principle to that of a surface-flow-constructed wetland [74, 75]. They can also significantly reduce the land requisition, investment and operational costs. Therefore, this is considered a promising technology for agricultural runoff control, especially in densely populated areas.

Ecological ditch system

An ecological ditch is an engineered system that has been developed for the removal of agricultural runoff nutrients by sorption, sedimentation, transformation, plant uptake and microbial metabolic activities [76–78]. As an important part of irrigation and drainage system, agricultural ditches are widely distributed among the farmland. Based on traditional agricultural ditches, ecological ditches are helpful to introduce substrates, aquatic plants, and interception facilities by forming a unique sediment-aquatic plant-microorganism system [79].

Periphyton is a key component of ecological ditches. It is widely distributed in natural water bodies and can help remove water pollutants by absorption, adsorption and complexation processes. Periphyton can have a large biomass and is sensitive to water quality and is effective at removing N and P, among other advantages. Table 3 listed the typical ecological ditches with different vegetation and their removal capacities of nutrients. Pierobon et al. have conducted N removal experiments in vegetated (*Phragmites australis* and *Typha latifolia*) and unvegetated ecological ditches in the Po River Basin of Italy [80]. The results showed an average removal capacity of 1.52 kg N km⁻¹ day⁻¹ in the vegetated ditches compared to the unvegetated ditches (0.24 kg N km⁻¹ day⁻¹). This indicated that aquatic plants play a vital role in the sediment-aquatic plant-microorganism system. Vymazal and Březinová

reported that a 200-m-long ecological ditch vegetated with *Epilobium hirsutum*, *Lythrum salicaria*, *Filipendula ulmaria*, *Phragmites australis*, *Typha latifolia*, and *Glyceria maxima* was used to treat the overflow from a fishpond in the Czech Republic [81]. They achieved removal capacities of 5.28 kg N km⁻¹ day⁻¹ and 0.70 kg P km⁻¹ day⁻¹. Flora and Kröger reported a drainage ditch vegetated with *Leersia oryzoides* and *Typha latifolia* in Mississippi State, USA. The nutrients removal capacities were 2.19 kg N km⁻¹ day⁻¹ and 0.58 kg P km⁻¹ day⁻¹ [82]. Li et al. studied the nitrogen removal in an ecological ditch vegetated with *Iris pseudacorus* and *Lythrum salicaria* in Tianjin, China [83]. The removal capacity of nitrogen was 1.73 kg N km⁻¹ day⁻¹. These results indicated that plant diversity has a great influence on the removal capacity of ecological ditches.

Therefore, the selection of highly efficient ditch plants is also important in ecological ditch research. Tyler et al. conducted a mesocosm study on the N removal performance of three plant species, *Leersia oryzoides*, *Typha latifolia*, and *Sparganium americanum* [84]. Using a hydraulic retention time of 48 h, TN removal efficiencies of these three plants were all higher than 50%, while ammonia-N removal efficiencies varied from 33.68% by *S. americanum* to 59.12% by *T. latifolia*. Kumwimba et al. compared six ditch plant species (*Canna indica*, *Acorus calamus*, *Cyperus alternifolius*, *Iris sibirica*, *Colocasia gigantean*, and *Myriophyllum verticillatum*) and found that *Canna indica* exhibited the best performance in N and P absorption and translocation [85]. As a result, 72–99.4% TN, 64–98.7% TP, 75–100% NH₄-N and 100% NO₃-N were removed after treatment.

During the growing seasons, plants can accumulate large amounts of nutrients for self-growth. However, their accumulation ability reduces gradually as senescence starts [86]. Furthermore, plants decomposition will lead to the release of the retained nutrients and, thus, become another source of nutrients [87]. Harvest management is an important aspect of ecological ditch management, though it still needs more in-depth studies. The complete removal of nutrients in ecological ditches is accomplished by plant harvesting. Yu et al. studied the harvest management of an ecological ditch vegetated with *Canna glauca*, *Hydrocotyle vulgaris*, *Sparganium stoloniferum*, *Myriophyllum verticillatum*, and *Juncus effuses* [88]. The removal capacity of TN and TP with multiple harvesting of aboveground plant tissues was 15.74 and 2.29 kg a⁻¹, respectively. In contrast, the removal capacity of TN and TP with annual harvesting was only 4.16 and 0.34 kg a⁻¹, respectively. Therefore, timely harvesting of ecological ditch's

Table 3 Ecological ditches and their vegetation and nutrients removal capacities

Location	Vegetation	Nutrients removal capacity	References
Po River Basin, Italy	<i>Phragmites australis</i>	1.52 kg N/(km day)	[80]
	<i>Typha latifolia</i>		
	Unvegetated	0.24 kg N/(km day)	
South-central Bohemia, Czech	<i>Epilobium hirsutum</i>	5.28 kg N/(km day)	[81]
	<i>Lythrum salicaria</i>	0.70 kg P/(km day)	
	<i>Filipendula ulmaria</i>		
	<i>Phragmites australis</i>		
	<i>Typha latifolia</i>		
Changsha, China	<i>Glyceria maxima</i>	3.20 kg N/(km day)	[28]
	<i>Canna indica</i>		
	<i>Hydrocotyle vulgaris</i>		
	<i>Sparganium stoloniferum</i>		
	<i>Myriophyllum aquaticum</i>		
	<i>Juncus effusus</i>		
Mississippi State, USA	<i>Leersia oryzoides</i>	2.19 kg N/(km day)	[82]
	<i>Typha latifolia</i>	0.58 kg P/(km day)	
Tianjin, China	<i>Iris pseudacorus</i>	1.73 kg N/(km day)	[83]
	<i>Lythrum salicaria</i>		

aquatic plants can effectively promote nutrient removal and plant regeneration. Unfortunately, management of ecological ditches has always been a difficult problem. Large-scale harvesting requires a great deal of labor, which greatly increases the maintenance cost. Smallholders and family farming are dominant forms of agriculture in Asia and Latin America [89]. For small-scale agriculture operators, such costs are usually too high to bear. Therefore, it impedes the large-scale application of ecological ditches in these areas.

Microbial treatment technologies

Due to the low efficiency of phytoremediation, N and P removal using ecological ditches cannot be compared to municipal sewage treatment technology, especially for irrigation and during the rainfall period. Microbial treatment technologies are effective, economical, and environmentally friendly to be used widely for treatment of domestic sewage, dyeing wastewater, and animal wastewater, among others [90, 91]. Therefore, microbial technologies might be the answer for next-generation ecological ditches. Activated sludge methods involving anaerobic-anoxic-aerobic (A2/O) processes have been used in highly concentrated organic wastewater treatments [92]. These microorganisms can simultaneously remove nutrients (e.g., N and P) and heavy metals (e.g., chromium and mercury) in a way that is both highly efficient and environmentally friendly [93, 94]. Wu et al. have proposed an integrated technology using both the A2/O approach and ecological ditches for treatment of heterogeneous non-point source wastewater [95]. This system could not only treat high load N and P wastewater, but also rejuvenate the ecological ditches' microbial habitat. With a hydraulic load of 200 m³ day⁻¹, the removal efficiencies of TP, TDP, TN, NO₃-N and NH₄-N reached 81%, 74%, 82%, 79% and 86%, respectively. It will be a promising research direction to introduce microbial enhancement into traditional ecological ditches.

Research progress in end treatment technologies

Agricultural runoff end treatment is the last barrier before the nutrients enter the downstream receiving water. Constructed wetlands (CW), buffer strips and land infiltration systems are common end treatment technologies.

CW is a unique system of soil-plant-microorganism. It is a transitional zone between the farmland and the receiving water, with good absorption, adsorption, and physical settlement capacity for N, P, particles and organic matter. Díaz et al. suggest that water evaporation, infiltration processes, vegetation characteristics and hydraulic retention time (HRT) are key factors affecting

pollutant load concentrations in CWs [22]. Additionally, the removal efficiency of wetlands is highly seasonal. Valkama et al. studied the seasonal variation of nutrient removal efficiencies and found that TP removal efficiency was highest in June (28%) and lowest in February (5.5%), while TN removal efficiency was highest in July (82%) and lowest in November (3.5%) [96].

Like ecological ditches, CWs also rely on phytoremediation and soil absorption. CW is considered a practical end treatment technology due to its numerous advantages (e.g., low-cost, easy operation, and easy maintenance) [97]. Beutel et al. reported a surface-flow constructed wetland can have a denitrification efficiency as high as 93% at 5 days hydraulic retention time (HRT) [98]. Surface-flow constructed wetland has free water surface and belongs to aerobic wetland, in which wastewater flows horizontally through the substrate surface [99]. The substrate surface formed by sediments and dead leaves of plants is the main site for denitrification. And, P is removed in more shallow oxidized layers. It is suitable for semi-arid environments where warm temperatures and low oxygen levels in the treatment wetland water promotes biological denitrification. The TP removal efficiency in surface-flow wetlands was 41% at 2.2 day HRT [100]. Another popular type of CW is subsurface-flow constructed wetland. Compared with the surface flow constructed wetlands, the water flows under the surface of the wetland bed, which can make full use of the biofilm growing on the surface of the packings, extend the hydraulic residence time, and improve the removal effect and capacity. Chung et al. showed a 62% TN and a 52% TP removal at 5 days HRT in a subsurface-flow constructed wetland [101].

CW also suffers from some intrinsic drawbacks including long HRT, a large footprint, and substrate clogging. These limit its application and long-term stability. Moreover, the oxygen transfer rate may limit the nitrification process while the denitrification process may be limited by organic carbon in water [102]. TN removal in a single-stage constructed wetland is not satisfactory due to its inability to provide both aerobic and anaerobic conditions simultaneously. Vertical flow-constructed wetlands perform well in ammonia-N removal, although they are very limited in TN removal. Subsurface flow-constructed wetlands have a strong TN removal capacity, but their ability to remove ammonia-N is very limited. Many efforts have been made to improve CWs by using different design and operational strategies [103–105]. Sgroi et al. compared four different kinds of wetlands under the same conditions and found that free water surface wetlands have the highest denitrification efficiency, 69%, while unsaturated vertical subsurface flow wetlands have the highest five-day biochemical oxygen demand

(BOD₅), chemical oxygen demand (COD), and total oxygen demand (TOD) removal efficiencies (at 87%, 67% and 72%, respectively) [106]. Attempts have been made to enhance CWs by introducing other proven technologies to synergistically degrade pollutants, such as membrane bio-reactors (MBR), electrochemical oxidation, and MFC. MBR technology has been combined with CWs to simultaneously improve water quality and reduce operational costs. Xiao et al. reported on an integrated system consisting of a submerged membrane bioreactor (SMBR, hollow fiber membrane) and a constructed wetland for the treatment of high load wastewater [107]. The initial concentrations of COD, TN, TP, and NH₄⁺ were as high as 1008.08 mg L⁻¹, 95.22 mg L⁻¹, 5.76 mg L⁻¹ and 62.10 mg L⁻¹, respectively. 98% COD, 96% TP, 80% TN, and 99% NH₄⁺ were removed by the integrated system. Compared with CW, the SMBR contributed most of the degradation capacity, accounting for 95% COD, 74% TP, 68.5% TN and 92% NH₄⁺ [107]. This demonstrates that there is a huge gap in the contaminant degradation rate between MBR and CW. Although the combination of other proven technologies can make up for CW's shortages, they can also weaken its advantages. The management and maintenance of these combined technologies are complex with the increase in the energy demand. Furthermore, CW requires large land requisition, which is becoming more and more difficult, especially in densely populated area. Sensitivity of aquatic plants to temperature can result in great changes in processing efficiency among seasons. Conventional CWs demonstrate difficulty meeting efficiency requirements when confronted with continued environmental deterioration and increasingly stringent emission standards.

Comprehensive control of agricultural runoff

As listed in Table 4, various technologies have been proposed for agricultural runoff control, which can be divided into three categories. Among them, conservation tillage, fertilization management, water saving irrigation, ecological ditch, constructed wetland, and buffer strips have been successfully applied in agricultural runoff control. As mentioned above, most N and P should be removed from agricultural runoff before being discharged into receiving waters. Currently, no single technology can meet such stringent requirements. Therefore, the thorough control of agricultural runoff requires the comprehensive application of various control technologies.

To understand the status of agricultural runoff treatments comprehensively, we conducted a scenario analysis based on reference data (Table 4). Paddy soil is a typical source of agricultural runoff because rice needs a large amount of irrigation water. Thus, paddy soil was selected as the runoff source in this scenario analysis. The

initial concentrations were 10 mg N L⁻¹ and 8 mg P L⁻¹, respectively, and their target output concentrations were set at 0.8 mg N L⁻¹ and 0.06 mg P L⁻¹. It means that the efficiencies of denitrification and phosphorus removal should reach 92% and 99.25%, respectively, as shown by the dotted line in Fig. 3. These levels are in accordance with the safe concentration thresholds of TN and TP in natural water [108]. According to Table 4, no-tillage, controlled-release fertilizer, ecological ditch, and surface-flow wetland are effective in nutrients removal of agricultural runoff from paddy soil. However, no control technique can achieve the target removal rate of nitrogen and phosphorus. Since they belong to source control, process control and end treatment technologies, they were assumed to be applied successively in this hypothetical farmland system. As shown in Fig. 3, the abscissa is the technique used, and the ordinate is the concentrations of nitrogen and phosphorus after the specific technique is used. For example, the application of no-tillage in paddy soil was expected to reduce TN by 8.5% and TP by 7.8% [46]. Then, the concentrations of TN and TP in agricultural runoff were expected to drop to 9.15 and 7.38 mg L⁻¹, respectively. The output concentrations of TN and TP declined to a safe level only when the source control, process control, and end treatment technologies were applied comprehensively. The TN and TP concentrations in agricultural runoff decreased to 0.39 and 0.47 mg L⁻¹ respectively after the successively application of no-tillage, controlled-release fertilizer, ecological ditch and surface-flow constructed wetland [46, 65, 85, 106]. Among these methods, controlled-release fertilizers contributed the most. Coupled with no-tillage, source control technologies played a vital role in agricultural runoff control, which contributed to more than 60% of the N and P reduction. In addition, water saving irrigation can effectively reduce runoff volume to facilitate the reduction of nutrient loads. Ecological ditches served as the connection channel between the farmland and the receiving water. Their natural advantages made full use of the runoff transport time to complete the N and P removal and contributed to 26.4% N and 21.8% P removal. After source and process control, the concentrations of N and P were reduced to 1.02 and 0.98 mg L⁻¹ respectively. These concentrations were very close to the targeted concentrations and indicated that agricultural runoff can be controlled with use of source and process control methods. For countries with limited arable land, this strategy would minimize the amount of land occupied for mitigation techniques.

Scenario analysis in this review was conducted under ideal conditions and without consideration of coupling effects between the different technologies. It still provides a comprehensive understanding of agricultural

Table 4 Current practices on N and P control from agricultural runoff

Number	Strategy	Category	Technology	Source	Runoff decrease (%)	TN decrease (%)	TP decrease (%)	References	
1	Source control	Conservation	Reduced	Cropland	64	7.7	–	[45]	
2			No-tillage	Paddy soil	25.9	8.5	7.8	[46]	
3			SPG cover	Cabbage field	29.4	34.7	7.8	[49]	
4		Amendment	Rotation tillage	Tropical soil	16.8	41.8	39.1	[50]	
5		Rotation tillage		Canadian prairies	–	60	38	[54]	
6		Fertilization management			Orchard	–	36.2	31.4	[62]
7				Band placement	Nursery land	–	61.2	68.1	[60]
8				Hole placement	Nursery land	–	65.1	67.9	[61]
9				Controlled-release N	Wheat-maize	–	30.5	–	[64]
10				Controlled-release	rotation Corn land	–	27.8	34	[65]
11				Controlled-release	Paddy soil	–	–	–	[66]
12	Process control	Water saving irrigation	Alternate wetting and drying	Paddy soil	30.2~36.7	–	–	[73]	
13									
14	End treatment	Ecological ditch	Ecological ditch	Paddy soil	–	72	64	[85]	
15									
16									Microbial treatment
17	Buffer strips		Surface-flow wetland	Agricultural runoff	–	60	–	[98]	
18				Agricultural runoff	–	–	41	[100]	
19			Subsurface-flow wetland	Urban	–	62	52	[106]	
20									
21	Buffer strips		Integrated buffer zone	Agricultural runoff	–	39	50	[112]	
22			Vegetated buffer strips	Maize field	–	52	–	[113]	

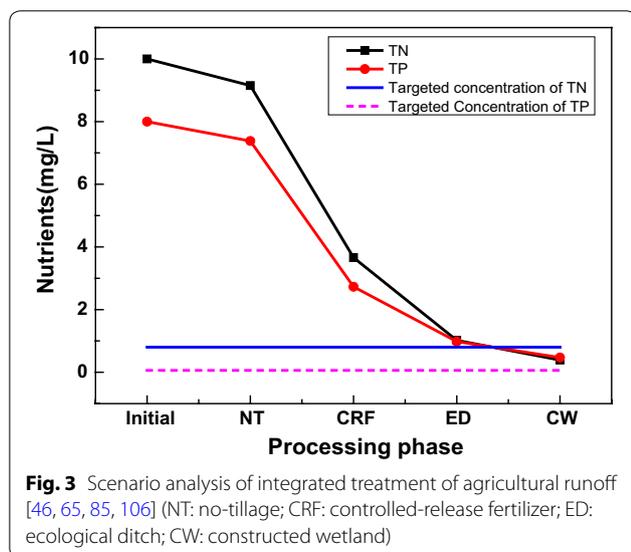


Fig. 3 Scenario analysis of integrated treatment of agricultural runoff [46, 65, 85, 106] (NT: no-tillage; CRF: controlled-release fertilizer; ED: ecological ditch; CW: constructed wetland)

runoff control, however. Since agricultural runoff characteristics vary temporally and spatially based on rainfall and irrigation events [109], the runoff treatment system’s removal efficiency also varies based on flow and concentration. The processing load of control and end treatment methods should be designed to meet peak processing requirements.

Nowadays, researchers have proposed a variety of effective agricultural runoff control techniques from different perspectives. But, neither technology can do the job of controlling agricultural runoff. This review identifies the spatial location of these technologies and their processing capabilities. Aiming at the control of N and P, this review makes full use of the space of farmland system and tries to realize the control of agricultural runoff through the comprehensive application of various technologies based on existing technologies. The ideal treatment technology for agricultural runoff should have

the following characteristics: (1) adaptability for local conditions including climate, geography, type of crops, planting 448 scale, agricultural facilities, and farmer education; (2) simplicity for management without complex operating procedures or technical specifications; (3) low investment and operational costs; (4) flexibility to accommodate big fluctuations in water volume and pollutant concentrations; (5) processability with comprehensive degradation capacities for nutrients and organic matter. To date, no existing technology can satisfy all these conditions.

Based on current technologies, integrated schemes of two or more are considered effective. Source control technologies can decrease both water volume and pollution load through tillage management, fertilization management, and water saving irrigation. They can be well popularized via government administrations. The subsequent processing load will be greatly alleviated after source control. Process control technologies such as ecological ditches can supplement source control technologies. CW has been proven as an effective method for end control in the past. However, confronted with the continual deterioration of the environment and increasingly stringent emission standards, CW systems operating as standalone technologies will have difficulty meeting new environmental requirements. Research is now focused on hybrid CWs, and it may be used in future as an improved method for existing constructed wetlands. Building new constructed wetlands is becoming increasingly difficult due to arable land shortages. Thus, further efforts to develop process control technologies are needed.

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Authors' contributions

XY performed and supported the analysis of technologies for agricultural non-point source pollution control. NG supported data analysis and statistical evaluation. MZ, DCWT, DL, LZ, ADI, PDD, XY and JR contributed in writing and formatting the manuscript. YSO mainly supervised the current study as corresponding author. All authors read and approved the final manuscript.

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All data generated or analyzed during this study are included in this published article and its supplementary information files. References are included for each and every data gathered from the published articles.

Competing interests

The authors declare that they have no competing interests.

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