

CRITICAL PROCESSES AND MAJOR FACTORS THAT DRIVE NITROGEN TRANSPORT FROM FARMLAND TO SURFACE WATER BODIES

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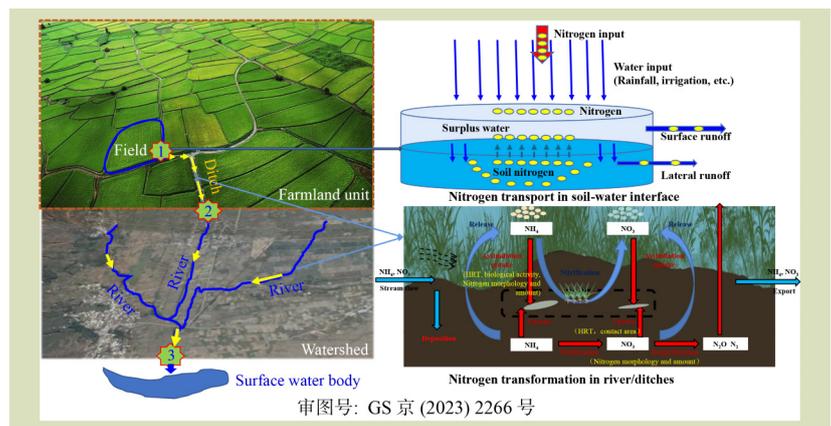
KEYWORDS

nitrogen, loss from soil, transformation, farm field scale, watershed scale

HIGHLIGHTS

- This study clarified the critical processes and major factors that nitrogen transport from farm fields to surface water bodies.
- Soil storage, exogenous inputs and meteorological hydrology were found to influence nitrogen loss from farmland.
- Hydrology, biogeochemistry and nitrogen inputs were found to influence the transformation of nitrogen in the ditches and rivers.

GRAPHICAL ABSTRACT



ABSTRACT

Agricultural non-point source pollution is increasingly an important issue affecting surface water quality. Currently, the majority of the studies on nitrogen loss have focused on the agricultural field scale, however, the response of surface water quality at the watershed scale into the nitrogen loss at the field scale is poorly understood. The present study systematically reviewed the critical processes and major factors that nitrogen transport from farm fields to surface water bodies. The critical processes of farmland nitrogen entering surface water bodies involve the processes of nitrogen transport from

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farmland to ditches and the transformation processes of nitrogen during migration in ditches/streams. Nitrogen transport from farmland to ditches is one of the prerequisites and critical processes for farmland nitrogen transport to surface water bodies. The transformation of nitrogen forms in ditches/streams is an intermediate process in the migration of nitrogen from farmland to surface water bodies. Nitrogen loss from farmland is related to soil storage and exogenous inputs. Therefore, nitrogen input management should not only consider the current input, but also the contribution of soil storage due to the historical surpluses. Ditches/streams have a strong retention capacity for nitrogen, which will significantly affect the process of farmland nitrogen entering surface water bodies. The factors affecting nitrogen transformation in river/ditches can be placed in four categories: (1) factors affecting hydraulic retention time, (2) factors affecting contact area, (3) factors affecting biological activity, and (4) forms and amount of nitrogen loading to river/ditches. Ditch systems are more biologically (including plants and microbes) active than rivers with biological factors having a greater influence on nitrogen transformation. When developing pollution prevention and control strategies, ecological ditches can be constructed to increase biological activity and reduce the amount of surplus nitrogen entering the water body. The present research should be valuable for the evaluation of environment impacts of nitrogen loss and the non-point source pollution control.

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1 INTRODUCTION

Nitrogen is one of the most active nutrients in farmland ecosystems and one of the major factors affecting the environment. The mechanism of nitrogen loss and its environmental impact in farmland is currently a research focus in plant nutrition, soil science, environmental science, ecology and others. Due to the pressing need for food security, increasingly exogenous nitrogen is being added to agricultural ecosystems, but nitrogen use efficiency is declining, resulting in a surplus of nitrogen in farmland^[1–3]. Under some conditions, excessive nitrogen is lost to the atmosphere through ammonia volatilization, denitrification and other processes, or lost to water bodies in runoff or through leaching^[4–10], which cause environmental problems such as air pollution or the water eutrophication. In the 1980s, the problem of nitrogen loss from farmland in China gradually attracted attention from the government because of the growing problem of water eutrophication. Loss of nitrogen from farmland caused by inappropriate fertilizer application (including excessive fertilizer application) and low utilization efficiency has become one of the major reasons for the increasing nitrogen concentration in surface water^[11–14]. Additionally, movement in runoff is the direct way for farmland nitrogen to enter surface water^[12,15]. The problem of nitrogen loss from

farmland has become a serious constraint to green development of agriculture in China. Therefore, research on pollution through nitrogen loss from farmland should be widely prioritized.

In recent years, as non-point source pollution control has been raised to the national strategic height, many studies have been conducted on the status and impact of nitrogen loss from farmland. The characteristics and main factors of nitrogen loss at the field scale have been clarified. However, most of the current studies focus on the farm field scale^[7,16,17], the research on the response of surface water quality at the watershed/regional scale to the nitrogen loss at the field scale is relatively insufficient, furthermore, most the studies are mainly based on empirical analysis^[18–22]. Therefore, it is necessary to understand the environmental effects of nitrogen loss from farm field on the surface water quality at the watershed/regional scale. It is important to sort out the critical processes and major factors that nitrogen transport from farm fields to surface water bodies. The research would be valuable for evaluating the water environment effect of farmland nitrogen loss and the non-point source pollution control.

The critical processes of farmland nitrogen entering surface water bodies involve the processes of nitrogen transport from

farmland to ditches and the transformation processes of nitrogen during migration in ditches/streams. Nitrogen transport from farmland to ditches is one of the prerequisites and critical processes for farmland nitrogen transport to surface water bodies. Ditches/streams are the major channels for nitrogen transport from farmland to surface water bodies. The transformation of nitrogen in ditches/streams is an intermediate process in the migration of nitrogen from farmland to surface water bodies, which is another important factor in the nitrogen transport into the surface water bodies. In summary, this paper reviewed the critical processes and major factors of nitrogen transport from farmland to ditches and transformation in ditches/streams through literature research.

2 CRITICAL PROCESSES OF NITROGEN TRANSPORT FROM FARMLAND TO SURFACE WATER BODY

The nitrogen transport from farmland into the surface receiving water body needs to go through multiple processes such as nitrogen transport in soil-water interface, nitrogen transport farmland to ditches, and nitrogen transformation during ditch during delivery in ditches/streams, corresponding to the three scales of nitrogen transport from farmland to surface bodies, namely, field scale, farmland unit and watershed scale (Fig. 1). The process of farmland-to-ditch export is the initial process of farmland nitrogen entering the surface water body, and nitrogen migration at the soil-water interface is the premise and key link of farmland nitrogen export to ditches^[23].

Only when nitrogen in soil water or runoff is fed into the ditch does it mean that farmland nitrogen actually enters the surface water. A series of physical, chemical and biological transformation processes occur during the nitrogen migration in ditches/streams^[24].

3 CRITICAL PROCESSES AND MAJOR FACTORS OF NITROGEN TRANSPORT FROM FARMLAND TO DITCHES

3.1 The critical processes of nitrogen transport from farmland to ditches

Nitrogen loss at the field scale is a prerequisite for farmland nitrogen transport to the surface body. Among them, the loss of nitrogen at the field scale includes the loss of soil nitrogen and the loss processes of exogenous nitrogen (Fig. 2). The soil-water interaction process is a key link in nitrogen transport from farmland to ditches^[25]. Soil is a complex composite structure. Nitrogen exists in many forms on the surface of soil particles, inside or in soil solution. In different forms and interacts with soil particles and water in different ways. Nitrogen may enter the soil water solution and runoff from the soil by different modes of action such as dissolution, desorption, ion exchange, and suspended particle carryover. Niu et al.^[26] further pointed out that mineral nitrogen was the major loss-prone nitrogen in soils, and that it was also subject to common competition from other processes such as crop utilization and denitrification processes. The correlation

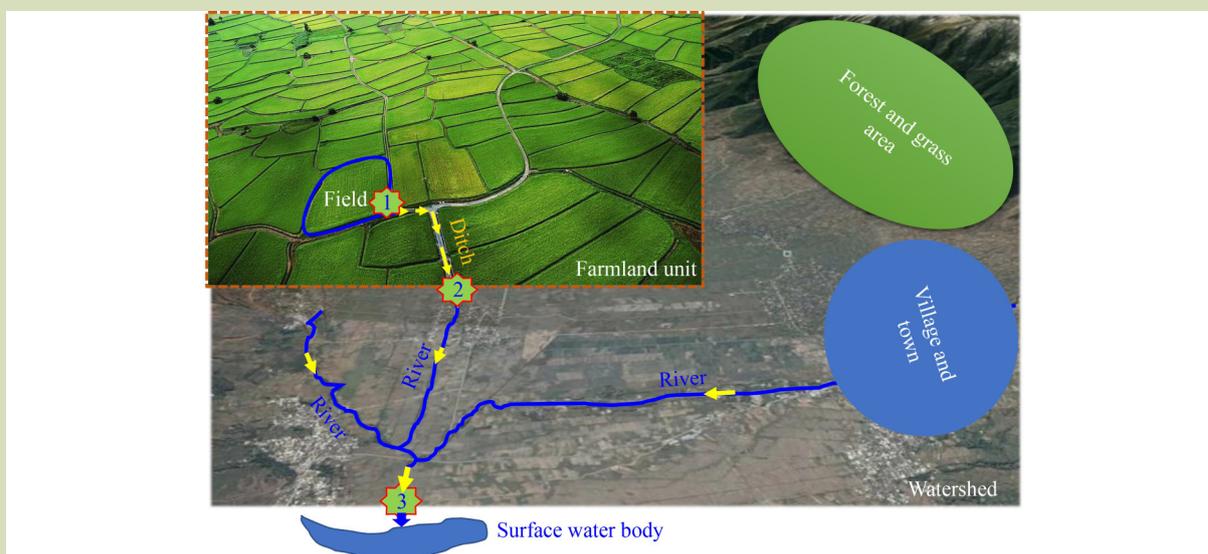


Fig. 1 The critical processes of nitrogen transport from farmland to surface water body (审图号: GS 京 (2023) 2266 号).

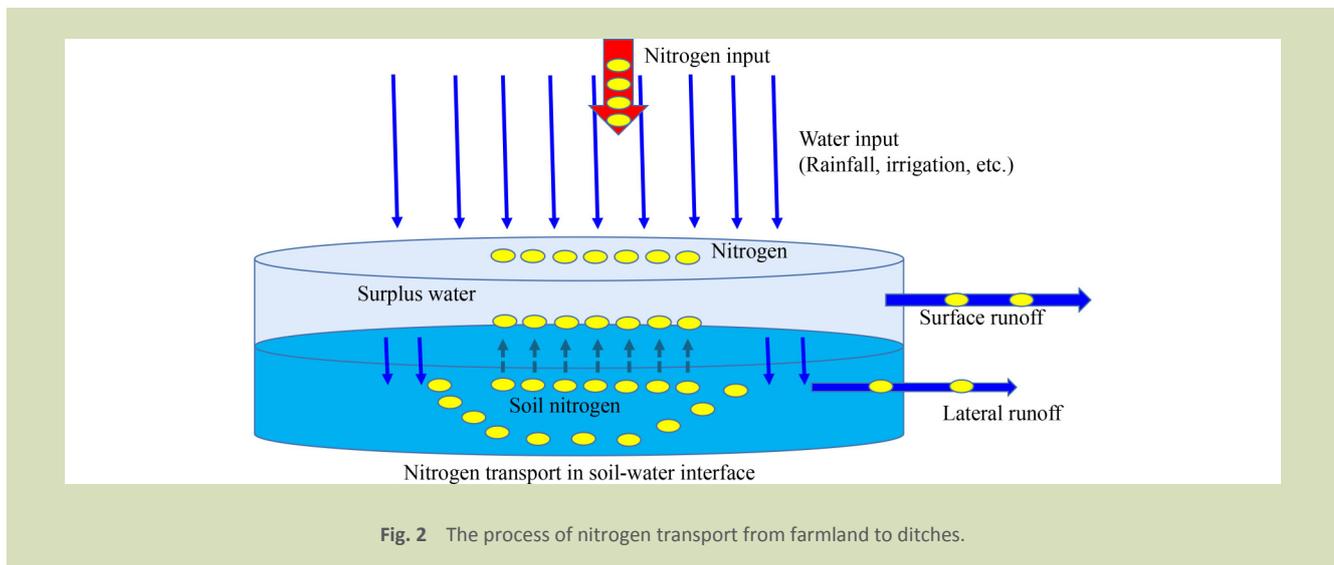


Fig. 2 The process of nitrogen transport from farmland to ditches.

between the loss (leaching) of nitrogen at the field scale and the nitrogen (endogenous nitrogen) produced by soil organic nitrogen mineralization was higher than that of exogenous nitrogen^[27,28]. Mineralization processes promote the increase of nitrogen transport from farmland to ditches. At the same time, nitrogen transport from farmland to ditches has a regulatory effect on the mineralization process of soil nitrogen. When the nitrogen content of easy loss reaches a certain level, the soil mineralization process is inhibited^[26].

Field-scale nitrogen loss also includes exogenous nitrogen loss processes. Sebilo et al.^[29] continuously tracked the whereabouts of ¹⁵N-labeled nitrogen fertilizer for 30 years, revealing the mechanism of nitrogen loss of residual fertilizer nitrogen by soil fixation and then slowly through the mineralization and nitrification process. Ju^[30] noted that the process of fertilizer nitrogen loss through soil fixation, remineralization and nitrification was only the critical loss mechanism of exogenous nitrogen under the conditions of a low nitrogen supply. Under the condition of excessive nitrogen supply, part of the applied fertilizer nitrogen directly entered the leakage water without going through the soil fixation and mineralization process. It also pointed out that the contribution of endogenous and exogenous soil nitrogen to nitrogen loss was limited by the balance between exogenous inputs and crop utilization in the current season: When the crop utilization is higher than the exogenous input (inorganic nitrogen), the inorganic nitrogen produced by the mineralization process of organic nitrogen remaining in the soil will be the main source of nitrogen easily lost in the soil. However, when the exogenous input is higher than the crop utilization, the contribution of exogenous input to nitrogen loss may increase^[30].

3.2 The critical factors of nitrogen transport from farmland to ditches

Related studies pointed out that soil nitrogen stock contributed significantly to nitrogen loss at the field scale. Hou et al.^[31] found that background sources (soil nitrogen) contributed more than 50% to the nitrogen loss from rice fields in China. Based on the isotope tracing technique, Li et al.^[9] found that the nitrogen loss from soils with high nitrogen content was significantly higher than that from soils with low nitrogen content. With the change of soil nitrogen content, the contribution rate of nitrogen loss is multiplied. Therefore, the loss of nitrogen at the field scale is limited by soil nitrogen stocks. As the main source of soil nitrogen, exogenous inputs such as fertilizer application have a significant impact on the level of nitrogen that is easy to lose in soil. Excess nitrogen not taken up by crops remains in the soil, resulting in a short-term increases or long-term accumulation of soil nitrogen stocks^[3,32–34]. Also, fertilizer application affects nitrogen transformations in soil. Mineralization of soil nitrogen is negatively correlated with soil carbon to nitrogen ratio^[35]. Fertilizer application leads to changes in soil carbon to nitrogen ratio, which in turn affects mineralization of soil organic nitrogen^[36]. It has been shown that with the continuous development of agriculture, a large amount of nitrogen enters the farmland ecosystems in the form of reactive nitrogen such as inorganic nitrogen from fertilizers, which changes the nitrogen cycling processes^[37], causing the accumulation of soil nitrogen^[30], and aggravates the loss of nitrogen^[38]. This is an important reason why the intensity of nitrogen loss from farmland ecosystems is much higher than that of natural ecosystems, such as woodlands and grasslands, which are less affected by human activities^[39–41]. In addition, with the increase of exogenous nitrogen input, nitrogen

movement from watersheds to rivers and other water bodies increases, resulting in an increase in water nitrogen concentration, aggravating water pollution^[42–44].

Various studies have found that the output load of nitrogen from farmland is significantly correlated with meteorological and hydrological conditions, including rainfall runoff^[45], and rainfall runoff is the main factor for farmland nitrogen output in most watersheds^[44,45]. In watersheds with different runoff intensities, the ratio of farmland nitrogen output to input was between 10% and 35%, but the ratio of farmland nitrogen output to input in watersheds with large runoff was higher^[39,46]. Li et al.^[15] found that rainfall led to seasonal variation in the migration pathway (runoff/baseflow) of nitrogen from farmland in the watershed. This was mainly related to the control of nitrogen transport from farmland to ditches by runoff factors. Hou et al.^[31] found that rainfall was one of the main factors controlling nitrogen loss from rice fields, and rainfall and soil clay content had the highest contribution to nitrogen loss variation in rice fields. Sinha and Michalak^[20] concluded that rainfall was the main factor leading to spatial differentiation of nitrogen loss and extreme rainfall events have a greater impact on the seasonal variation in nitrogen loss.

In general, as rainfall increases, agricultural runoff increases, nitrogen transport from farmland to ditches increases^[47], the attenuation rate decreases during migration downstream of the watershed^[38] and the nitrogen output load of the watershed increases^[45]. In addition, rainfall conditions alter the pathways of nitrogen loss from farmland. Fu et al.^[48] found that the

nitrogen loss pathway from rice fields in the dry season was dominated by subsurface leaching, while the contribution of the runoff to nitrogen loss increased in the rainy season. Hydrological conditions are limited to meteorological factors, including rainfall, temperature and wind speed. Rainfall type and rainfall intensity all affect nitrogen transport from farmland to ditches^[47,49]. Temperature is also one of the most important factors affecting nitrogen output. The proportion of nitrogen loss from farmland is inversely proportional to temperature within a certain temperature range. It is related to both the increase in evaporation due to the increase in temperature and the decrease in runoff^[50] and the denitrification loss of nitrogen exacerbated by higher temperature^[51,52].

4 CRITICAL PROCESSES AND MAJOR FACTORS OF NITROGEN TRANSFORMATION IN DITCHES AND RIVERS

4.1 Major processes of nitrogen transformation in ditches and rivers

The transformations that occurs when nitrogen migrates in ditches/rivers can be categorized as retention and release (Fig. 3). Retention is the phenomenon in which nitrogen is affected by adsorption, deposition, absorption and gaseous loss (including ammonia volatilization and denitrification) in ditches/rivers, so that the output nitrogen load decreases after migrating through ditches/rivers, that is, the occurrence of

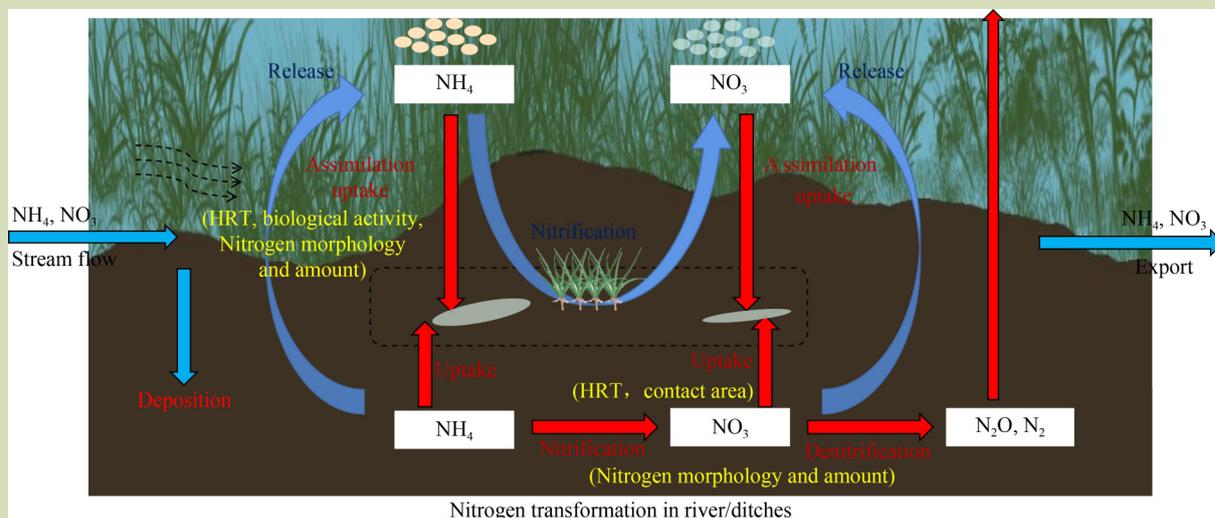


Fig. 3 Processes and factors of nitrogen transformation in ditches/rivers.

attenuation. Release is the phenomenon in which nitrogen is affected by resolution, dissolution, degradation and other effects in ditches/streams, and nitrogen is re-released in ditches/streams, increasing the output load. Retention can be divided into temporary and permanent retention. Temporary retention mainly refers to the transfer of nitrogen from the aqueous phase to other locations of the aquatic ecosystem through various processes (i.e., to sinks), and under certain conditions, it can be released as an endogenous source and re-enter the aqueous phase (i.e., as a source), the temporary retention process is reversible, that is, a process of retention and release, including absorption-degradation processes of aquatic plants, the adsorption-desorption processes of sediment and deposition-dissolution processes. Permanent retention mainly refers to the complete separation of nitrogen from aquatic ecosystems through various complex processes, including as volatilization, absorption by organisms and denitrification.

4.2 Major factors of nitrogen transformation in ditches and rivers

Nitrogen undergoes a series of complex physical, chemical and biological transformations during river migration. These transformations can be divided into two processes: (1) retention with attenuation effects, including adsorption, deposition, absorption, and gaseous loss (including ammonia volatilization and denitrification); and (2) release including resolution, dissolution and degradation^[51]. The above processes are a result of the combined effects of various factors. Generally, according to whether there are organisms involved, each factor can be classified into two categories: biotic and abiotic factors^[53,54]. Biotic factors include aquatic plants (species, cover area, plant community structure, metabolic processes and microbes (including algae, bacteria, fungi). Abiotic factors include river geology (including soil type, texture and structure), topography (including slope, slope length, slope fall), river morphology (including river length, river width and curvature)^[55–57], river location^[58], hydrological conditions (including flow, flow rate, water depth and sedimentation rate)^[59,60], watershed area (scale effect)^[61,62], land use type^[63–65], terrestrial nitrogen input (concentration)^[63,66–68], water temperature^[69], and light.

The mechanisms of different factors on nitrogen transformations differ and can be classified into four categories according to their mode of action: (1) factors affecting hydraulic residence time, (2) factors affecting contact area, (3) factors affecting biological activity, and (4) nitrogen inflow morphology and amount. From the perspective of the reaction

process, the hydraulic retention time^[60] determines the reaction time. The shorter the residence time, the shorter the reaction time and the less complete the reaction. The shorter the residence time of nitrogen in a river, the lower the chance of nitrogen being in contact with sediments, biofilms and the like, so the chance of being absorbed by these is smaller, and the attenuation efficiency decreases accordingly. The contact area of nitrogen with sediments and biofilms determines the area where the attenuation reaction occurs. The larger the contact area, the larger the area of reactive surface, and the higher the attenuation rate. Biotic factors (including the abundance of denitrifying bacteria) determine the responsiveness of the active component. The higher the biotic activity of the active component, the stronger its responsiveness and the greater the reaction rate^[70]. The form and amount of nitrogen entering a river determines the substrate concentration in the reaction. The higher the substrate concentration, the higher the rate of nitrogen transformation. In general, there is a maximum attenuation rate of nitrogen in a river. When the attenuation rate is maximum, a river channel has the greatest ability to remove nitrogen. As the amount of nitrogen entering a river continues to increase, the total amount of nitrogen removed will remain unchanged but the removal rate will decrease^[64,67].

Factors that affect water residence time include river length, flow rate, river curvature and slope. The greater the flow rate for the same river length, the shorter the time the water stays in the river. The longer the distance nitrogen migrates in a river, the more completely it is attenuated^[71,72], that is, the longer a river, the greater the amount of nitrogen removed by the sediment and aquatic ecosystem. River curvature and slope change the residence time of nitrogen in a river channel by affecting the flow rate, which in turn affects the attenuation rate. The more curved the river, the smaller its slope, the longer the water residence time, the more completely the nitrogen is attenuated. Factors affecting the contact area include water depth, river width and geology. Water depth affects the nitrogen conversion process by changing the contact area between the water body and the sediment. With greater water depth, the contact area between a unit water body and the sediment decreases, that is, the contact chance between the nitrogen in the water body and the self-purification active site of the sediment decreases, resulting in a decrease in the nitrogen attenuation rate^[73]. River width also affects the contact area between nitrogen and the sediment. In general, nitrogen has less contact area with the sediment per unit length of a large river than a small river. In addition, the contact area between nitrogen and sediment is related to the geology of the riverbed. The larger the rock grains in the riverbed, the larger

the gap between them, the larger the space for river water, and the larger the contact area between nitrogen and the sediment. Factors affecting biological activity include water temperature and light^[51,52,57,69,74]. This affects the biochemical decay process by affecting the growth and microbial activity of aquatic plants, algae and other organisms^[75]. In general, the photosynthesis of aquatic autotrophs will increase with an increase in water temperature and light, and the nutrients they absorb from the water body will also increase synchronously. Also, increased water temperature and light will increase the activity of microorganisms attached to biofilms and in sediments, accelerating nitrogen attenuation. Numerous studies have shown that the rate of denitrification of river nitrogen is positively correlated with water temperature^[51,52,69].

In addition to these factors, the morphology and amount of nitrogen entering a river influences the attenuation process in a river. The rate of nitrogen removal from rivers is closely related to the concentration of nitrogen in rivers^[64,66,69,76]. In general, as the amount of nitrogen entering a river increases, the amount of nitrogen attenuation increases and the rate of attenuation initially increases and then decreases^[77], which is mostly related to the nitrogen removal capacity of a river. Wollheim et al.^[67] emphasized that there is a saturation point in the removal capacity of rivers for nitrogen. When the amount of incoming nitrogen is less than the maximum removal capacity of a river, the removal rate of nitrogen from the river increases as the amount of incoming nitrogen increases. However, when the amount of incoming nitrogen exceeds the maximum removal capacity of a river, the removal rate of nitrogen from the river then decreases. They also emphasized that flow was an important indicator of how much nitrogen enters a river. It has been shown that the higher the flow rate, the lower the removal rate of nitrogen from a river^[60], which is mainly related to the hydrological dependence of nitrogen loss from the land surface. Various studies have shown that the loss of nitrogen on the land surface is closely related to hydrological factors such as runoff^[78–81]. In addition, the removal efficiency of incoming nitrogen varies depending on the form of the nitrogen entering a river^[24].

Generally, the hydrological factors and biogeochemistry interact to influence nitrogen retention^[82]. The hydrological factors determine the residence time, as well as the nitrogen input loading to river/ditches due to their influence on the nitrogen loss from farmland^[83]. Typically, the effect of biogeochemistry factors depends on the residence time. Therefore, hydrological factors underpin biogeochemistry factors that influence the nitrogen retention. Also, the biogeochemistry factors in large rivers are generally weak, thus

the hydrological factors dominates nitrogen retention in such contexts. However, in the ditches, especially ecological ditches, the biogeochemistry has a more important influence on nitrogen retention.

5 CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

This paper has systematically reviewed the critical processes and major factors in nitrogen transport from farm fields to surface water bodies. The critical processes of farmland nitrogen entering surface water bodies involve the processes of nitrogen transport from farmland to ditches and the transformations of nitrogen during migration in ditches/rivers. Nitrogen transport from farmland to ditches is a prerequisite and critical process for farmland nitrogen transport to surface water bodies. Nitrogen transformations in ditches/rivers are an intermediate link and nexus for lost nitrogen from farmland to enter surface water bodies.

The following is a summary of the article and the application of future prevention and treatment. (1) Nitrogen loss from farmland is related to soil storage, exogenous inputs and meteorological hydrology. Therefore, management practices should be applied to limit nitrogen loss from farmland. Reduction of nitrogen input should not be the only consideration, as the historical contribution nitrogen application resulting in soil storage must also be considered. The effects of climate change on nitrogen loss are also worth of consideration. (2) Ditches/rivers have a substantial retention capacity for nitrogen, which will significantly affects the process of farmland nitrogen entering surface water. So the retention capacity of ditches/rivers needs to be taken into account when assessing the impact of nitrogen from agricultural land on surface water, and for pollution prevention and control. (3) The factors affecting nitrogen transport and transformation can be grouped in four categories: (a) factors affecting hydraulic retention time, (b) factors affecting contact area, (c) factors affecting biological activity, and (d) nitrogen inflow pattern and amount. Ditch systems are more biologically active than rivers. Biotic factors have a greater influence on nitrogen transformation in ditches than abiotic factors. When developing pollution prevention and control strategies, ecological ditches can be constructed to increase biological activity and to reduce the amount of excess nitrogen entering water bodies.

5.2 Recommendations

At present, although there is a clear understanding of the critical processes and major factors of nitrogen migration from farmland into surface waters, with the emergence of some new issues, such as the increase in atmospheric nitrogen deposition and the impact of historical legacy nitrogen on water quality improvement. There is a need for in-depth research on the pathways of nitrogen migration from farmland into surface waters, the relationship between nitrogen from farmland and atmospheric deposition, and the impact of historical legacy nitrogen on the watershed/regional scale environmental effects of nitrogen loss from farmland.

Surface runoff is the main way for nitrogen from farmland to enter surface water bodies. However, increasingly studies have found that in addition to the runoff process, other pathways, such as underground tile drain and regression flow processes, are also important ways for farmland nitrogen to enter surface water bodies^[84,85], and even become the main way in some areas. Fu et al.^[48] found that underground leaching was the main way of nitrogen loss from rice fields, which in turn leads to an increase in nitrogen concentration in the base stream^[86]. Therefore, the processes and mechanisms of nitrogen transport and transformation along pathways other than runoff should also receive attention. In addition, it has been found that atmospheric deposition has become one of the important sources of nitrogen in surface waters^[87–89], and it is closely related to agricultural activities. Ti et al.^[90] found that ammonia emitted by fertilizer application to farmland and livestock farming contributed more than 60% to $\text{NH}_x\text{-N}$ deposition in the region. Studying the critical processes and major factors of nitrogen emissions from agricultural activities that pollute surface water bodies through atmospheric deposition is also one of the future research priorities.

At present, the impact of historical nitrogen on surface water quality is gradually being recognized^[91]. Van Meter et al.^[92]

posited that due to the influence of historical nitrogen in the Mississippi River Basin, even if the nitrogen utilization rate reaches 100%, it will take decades to achieve the expected water quality control target. Due to the limited ability to simulate the historical nitrogen loss process at present, the simulation errors of the existing models are relatively large^[89], and the understanding of its contribution is not yet comprehensive. It is also one of the current research focuses to strengthen the systematic model simulation research on the migration and transformation process of historical nitrogen and to systematically evaluate the impact of exogenous and residual nitrogen on surface water quality. In addition, by integrating soil hydrology and nitrogen cycle models, based on the concept of hydrological soil functional units, constructing the quantitative relationship between key parameters of the nitrogen cycle and hydrology-soil-landscape is one of the important ways to improve understanding of the environmental effects of legacy nitrogen^[23].

The migration and transformation of nitrogen in ditches and rivers is the key link that determines the migration of nitrogen from farmland into surface water bodies. Therefore, quantifying the transformation process of nitrogen migration in ditches and rivers is important for improving the watershed/regional scale of nitrogen loss from farmland. Awareness of environmental effects is crucial. Currently, the methods used to quantify the transport and transformation of nitrogen in ditches and rivers are mainly model based, with fewer studies based on empirical methods to elucidate the processes. With the development of online monitoring technology, it has become possible to use online technology to design segmented multilevel monitoring to study the migration and transformation process of nitrogen in ditches and rivers. Wollheim et al.^[60] installed online nitrate monitoring equipment upstream and downstream of a river and found that the removal of nitrate in a river was closely related to the flow rate. Nitrogen removal rate is higher when the flow rate is low than when the flow rate is high.

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Compliance with ethics guidelines

Wenchao Li, Wen Xu, Gaofei Yin, Xulin Zhang, Zihan Zhang, Bin Xi, Qiuliang Lei, Limei Zhai, Qiang Zhang, Linzhang Yang, and Hongbin Liu declare that they have no conflicts of interest or financial conflicts to disclose. This article does not contain any studies with human or animal subjects performed by any of the authors.

REFERENCES

1. He W, Jiang R, He P, Yang J, Zhou W, Ma J, Liu Y. Estimating soil nitrogen balance at regional scale in China's croplands from 1984 to 2014. *Agricultural Systems*, 2018, **167**: 125–135
2. Yan X, Ti C, Vitousek P, Chen D, Leip A, Cai Z, Zhu Z. Fertilizer nitrogen recovery efficiencies in crop production systems of China with and without consideration of the residual effect of nitrogen. *Environmental Research Letters*, 2014, **9**(9): 095002
3. Zhou J, Gu B, Schlesinger W H, Ju X. Significant accumulation of nitrate in Chinese semi-humid croplands. *Scientific Reports*, 2016, **6**(1): 25088
4. Ju X T, Xing G X, Chen X P, Zhang S L, Zhang L J, Liu X J, Cui Z L, Yin B, Christie P, Zhu Z L, Zhang F S. Reducing environmental risk by improving N management in intensive Chinese agricultural systems. *Proceedings of the National Academy of Sciences of the United States of America*, 2009, **106**(9): 3041–3046
5. Gu B, Ju X, Chang J, Ge Y, Vitousek P M. Integrated reactive nitrogen budgets and future trends in China. *Proceedings of the National Academy of Sciences of the United States of America*, 2015, **112**(28): 8792–8797
6. Zhang D, Wang H, Pan J, Luo J, Liu J, Gu B, Liu S, Zhai L, Lindsey S, Zhang Y, Lei Q, Wu S, Smith P, Liu H. Nitrogen application rates need to be reduced for half of the rice paddy fields in China. *Agriculture, Ecosystems & Environment*, 2018, **265**: 8–14
7. Hou X, Zhan X, Zhou F, Yan X, Gu B, Reis S, Wu Y, Liu H, Piao S, Tang Y. Detection and attribution of nitrogen runoff trend in China's croplands. *Environmental Pollution*, 2018, **234**: 270–278
8. Wang Y, Ying H, Yin Y, Zheng H, Cui Z. Estimating soil nitrate leaching of nitrogen fertilizer from global meta-analysis. *Science of the Total Environment*, 2019, **657**: 96–102
9. Li W, Guo S, Liu H, Zhai L, Wang H, Lei Q. Comprehensive environmental impacts of fertilizer application vary among different crops: implications for the adjustment of agricultural structure aimed to reduce fertilizer use. *Agricultural Water Management*, 2018, **210**: 1–10
10. Yin G, Wang X, Du H, Shen S, Liu C, Zhang K, Li W N. N_2O and CO_2 emissions, nitrogen use efficiency under biogas slurry irrigation: a field study of two consecutive wheat-maize rotation cycles in the North China Plain. *Agricultural Water Management*, 2019, **212**: 232–240
11. Stokal M, Yang H, Zhang Y, Kroeze C, Li L, Luan S, Wang H, Yang S, Zhang Y. Increasing eutrophication in the coastal seas of China from 1970 to 2050. *Marine Pollution Bulletin*, 2014, **85**(1): 123–140
12. Murphy T, Dougall C, Burger P, Carroll C. Runoff water quality from dryland cropping on Vertisols in Central Queensland, Australia. *Agriculture, Ecosystems & Environment*, 2013, **180**: 21–28
13. Zhang X, Davidson E A, Mauzerall D L, Searchinger T D, Dumas P, Shen Y. Managing nitrogen for sustainable development. *Nature*, 2015, **528**(7580): 51–59
14. Zhang Y, Wang H, Lei Q, Luo J, Lindsey S, Zhang J, Zhai L, Wu S, Zhang J, Liu X, Ren T, Liu H. Optimizing the nitrogen application rate for maize and wheat based on yield and environment on the Northern China Plain. *Science of the Total Environment*, 2018, **618**: 1173–1183
15. Li W, Lei Q, Zhai L, Liu H, Hu W, Liu S, Ren T. Seasonal changes of the pathways of nitrogen export from an agricultural watershed in China. *Environmental Sciences*, 2018, **39**(12): 5375–5382 (in Chinese)
16. Gao S, Xu P, Zhou F, Yang H, Zheng C, Cao W, Tao S, Piao S, Zhao Y, Ji X, Shang Z, Chen M. Quantifying nitrogen leaching response to fertilizer additions in China's cropland. *Environmental Pollution*, 2016, **211**: 241–251
17. Yue Q, Ledo A, Cheng K, Albanito F, Lebender U, Sapkota T B, Brentrup F, Stirling C M, Smith P, Sun J, Pan G, Hillier J. Re-assessing nitrous oxide emissions from croplands across Mainland China. *Agriculture, Ecosystems & Environment*, 2018, **268**: 70–78
18. Lian H, Lei Q, Zhang X, Haw Y, Wang H, Zhai L, Liu H, Jr-Chuan H, Ren T, Zhou J, Qiu W. Effects of anthropogenic activities on long-term changes of nitrogen budget in a plain river network region: a case study in the Taihu Basin. *Science of the Total Environment*, 2018, **645**: 1212–1220
19. Hong B, Swaney D P, McCrackin M, Svanbäck A, Humborg C, Gustafsson B, Yershova A, Pakhomau A. Advances in NANI and NAPI accounting for the Baltic drainage basin: spatial and temporal trends and relationships to watershed TN and TP fluxes. *Biogeochemistry*, 2017, **133**(3): 245–261
20. Sinha E, Michalak A M. Precipitation dominates interannual variability of riverine nitrogen loading across the Continental United States. *Environmental Science & Technology*, 2016, **50**(23): 12874–12884
21. Gao W, Howarth R W, Swaney D P, Hong B, Guo H C. Enhanced N input to Lake Dianchi Basin from 1980 to 2010: drivers and consequences. *Science of the Total Environment*, 2015, **505**: 376–384
22. Hong B, Swaney D P, Howarth R W. Estimating net anthropogenic nitrogen inputs to U.S. watersheds: comparison of methodologies. *Environmental Science & Technology*, 2013, **47**(10): 5199–5207
23. Zhu Q, Castellano M J, Yang G. Coupling soil water processes and the nitrogen cycle across spatial scales: potentials, bottlenecks and solutions. *Earth-Science Reviews*, 2018, **187**: 248–258
24. Peterson B J, Wollheim W M, Mulholland P J, Webster J R, Meyer J L, Tank J L, Martí E, Bowden W B, Valett H M, Hershey A E, McDowell W H, Dodds W K, Hamilton S K, Gregory S, Morrall D D. Control of nitrogen export from

- watersheds by headwater streams. *Science*, 2001, **292**(5514): 86–90
25. Gao B, Walter M T, Steenhuis T S, Hogarth W L, Parlange J Y. Rainfall induced chemical transport from soil to runoff: theory and experiments. *Journal of Hydrology*, 2004, **295**(1–4): 291–304
26. Niu S, Classen A T, Dukes J S, Kardol P, Liu L, Luo Y, Rustad L, Sun J, Tang J, Templer P H, Thomas R Q, Tian D, Vicca S, Wang Y P, Xia J, Zaehle S. Global patterns and substrate-based mechanisms of the terrestrial nitrogen cycle. *Ecology Letters*, 2016, **19**(6): 697–709
27. Kopáček J, Hejzlar J, Posch M. Factors controlling the export of nitrogen from agricultural land in a large central European catchment during 1900–2010. *Environmental Science & Technology*, 2013, **47**(12): 6400–6407
28. Chen D, Hu M, Guo Y, Dahlgren R A. Influence of legacy phosphorus, land use, and climate change on anthropogenic phosphorus inputs and riverine export dynamics. *Biogeochemistry*, 2015, **123**(1–2): 99–116
29. Sebilo M, Mayer B, Nicolardot B, Pinay G, Mariotti A. Long-term fate of nitrate fertilizer in agricultural soils. *Proceedings of the National Academy of Sciences of the United States of America*, 2013, **110**(45): 18185–18189
30. Ju X. Direct pathway of nitrate produced from surplus nitrogen inputs to the hydrosphere. *Proceedings of the National Academy of Sciences of the United States of America*, 2014, **111**(4): E416
31. Hou X, Zhou F, Leip A, Fu B, Yang H, Chen Y, Gao S, Shang Z, Ma L. Spatial patterns of nitrogen runoff from Chinese paddy fields. *Agriculture, Ecosystems & Environment*, 2016, **231**: 246–254
32. Wu H, Song X, Zhao X, Peng X, Zhou H, Hallett P D, Hodson M E, Zhang G L. Accumulation of nitrate and dissolved organic nitrogen at depth in a red soil Critical Zone. *Geoderma*, 2019, **337**: 1175–1185
33. Lu J, Bai Z, Velthof G L, Wu Z, Chadwick D, Ma L. Accumulation and leaching of nitrate in soils in wheat-maize production in China. *Agricultural Water Management*, 2019, **212**: 407–415
34. Worrall F, Howden N J K, Burt T P. Evidence for nitrogen accumulation: the total nitrogen budget of the terrestrial biosphere of a lowland agricultural catchment. *Biogeochemistry*, 2015, **123**(3): 411–428
35. Deng Q, Cheng X, Yang Y, Zhang Q, Luo Y. Carbon-nitrogen interactions during afforestation in central China. *Soil Biology & Biochemistry*, 2014, **69**: 119–122
36. Wang J, Zhu B, Zhang J, Müller C, Cai Z. Mechanisms of soil N dynamics following long-term application of organic fertilizers to subtropical rain-fed purple soil in China. *Soil Biology & Biochemistry*, 2015, **91**: 222–231
37. Billen G, Garnier J, Lassaletta L. The nitrogen cascade from agricultural soils to the sea: modelling nitrogen transfers at regional watershed and global scales. *Philosophical Transactions of the Royal Society of London. Series B: Biological Sciences*, 2013, **368**(1621): 20130123
38. Dupas R, Delmas M, Dorioz J M, Garnier J, Moatar F, Gascuel-Oudou C. Assessing the impact of agricultural pressures on N and P loads and eutrophication risk. *Ecological Indicators*, 2015, **48**: 396–407
39. Howarth R, Swaney D, Billen G, Garnier J, Hong B, Humborg C, Johnes P, Mörth C M, Marino R. Nitrogen fluxes from the landscape are controlled by net anthropogenic nitrogen inputs and by climate. *Frontiers in Ecology and the Environment*, 2012, **10**(1): 37–43
40. Kaushal S S, Groffman P M, Band L E, Elliott E M, Shields C A, Kendall C. Tracking nonpoint source nitrogen pollution in human-impacted watersheds. *Environmental Science & Technology*, 2011, **45**(19): 8225–8232
41. Xu Z, Zhang X, Xie J, Yuan G, Tang X, Sun X, Yu G. Total nitrogen concentrations in surface water of typical agro- and forest ecosystems in China, 2004–2009. *PLoS One*, 2014, **9**(3): e92850
42. Swaney D P, Hong B, Ti C, Howarth R W, Humborg C. Net anthropogenic nitrogen inputs to watersheds and riverine N export to coastal waters: a brief overview. *Current Opinion in Environmental Sustainability*, 2012, **4**(2): 203–211
43. Iital A, Pachel K, Loigu E, Pihlak M, Leisk Ü. Recent trends in nutrient concentrations in Estonian rivers as a response to large-scale changes in land-use intensity and life-styles. *Journal of Environmental Monitoring*, 2010, **12**(1): 178–188
44. Li W, Liu H, Zhai L, Yen H, Hu W, Lei Q, Stewart R, Guo S, Ren T. Evaluation of concentration-discharge dynamics and nitrogen export on anthropogenic inputs and stormflow across alternative time-scales. *Ecological Indicators*, 2019, **98**: 879–887
45. Bettez N D, Duncan J M, Groffman P M, Band L E, O’Neil-Dunne J, Kaushal S S, Belt K T, Law N. Climate variation overwhelms efforts to reduce nitrogen delivery to coastal waters. *Ecosystems*, 2015, **18**(8): 1319–1331
46. Howarth R W, Swaney D P, Boyer E W, Marino R, Jaworski N, Goodale C. The influence of climate on average nitrogen export from large watersheds in the Northeastern United States. *Biogeochemistry*, 2006, **79**(1–2): 163–186
47. Yang T, Wang Q, Wu L, Zhao G, Liu Y, Zhang P. A mathematical model for soil solute transfer into surface runoff as influenced by rainfall detachment. *Science of the Total Environment*, 2016, **557–558**: 590–600
48. Fu J, Wu Y, Wang Q, Hu K, Wang S, Zhou M, Hayashi K, Wang H, Zhan X, Jian Y, Cai C, Song M, Liu K, Wang Y, Zhou F, Zhu J. Importance of subsurface fluxes of water, nitrogen and phosphorus from rice paddy fields relative to surface runoff. *Agricultural Water Management*, 2019, **213**: 627–635
49. Wu L, Peng M, Qiao S, Ma X. Assessing impacts of rainfall intensity and slope on dissolved and adsorbed nitrogen loss under bare loessial soil by simulated rainfalls. *Catena*, 2018, **170**: 51–63
50. Hong B, Swaney D P, Mörth C M, Smedberg E, Hägg H E, Humborg C, Howarth R W, Bouraoui F. Evaluating regional variation of net anthropogenic nitrogen and phosphorus

- inputs (NANI/NAPI), major drivers, nutrient retention pattern and management implications in the multinational areas of Baltic Sea basin. *Ecological Modelling*, 2012, **227**: 117–135
51. Schaefer S C, Hollibaugh J T, Alber M. Watershed nitrogen input and riverine export on the west coast of the US. *Biogeochemistry*, 2009, **93**(3): 219–233
52. Kang L J, Xu H, Zhu G W, Zhu M Y, Zhao F. Sediment denitrification potential and its influencing factors in the main rivers of Taihu Lake. *Acta Scientiae Circumstantiae*, 2021, **41**(4): 1393–1400 (in Chinese)
53. Birgand F, Skaggs R W, Chescheir G M, Gilliam J W. Nitrogen removal in streams of agricultural catchments—A literature review. *Critical Reviews in Environmental Science and Technology*, 2007, **37**(5): 381–487
54. Castellano-Hinojosa A, Bedmar E J, Medina-Sánchez J M. Efficiency of reactive nitrogen removal in a model Mediterranean high-mountain lake and its downwater river ecosystem: biotic and abiotic controls. *Science of the Total Environment*, 2022, **858**(Part 2): 159901
55. Alexander R B, Smith R A, Schwarz G E. Effect of stream channel size on the delivery of nitrogen to the Gulf of Mexico. *Nature*, 2000, **403**(6771): 758–761
56. Wollheim W M, Peterson B J, Deegan L A, Hobbie J E, Hooker B, Bowden W B, Edwardson K J, Arscott D B, Hershey A E, Finlay J. Influence of stream size on ammonium and suspended particulate nitrogen processing. *Limnology and Oceanography*, 2001, **46**(1): 1–13
57. Wollheim W M, Vörösmarty C J, Bouwman A F, Green P, Harrison J, Linder E, Peterson B J, Seitzinger S P, Syvitski J P M. Global N removal by freshwater aquatic systems using a spatially distributed, within-basin approach. *Global Biogeochemical Cycles*, 2008, **22**(2): GB2026
58. Stewart R J, Wollheim W M, Gooseff M N, Briggs M A, Jacobs J M, Peterson B J, Hopkinson C S. Separation of river network-scale nitrogen removal among the main channel and two transient storage compartments. *Water Resources Research*, 2011, **47**(10): W00J10
59. Wang Z J, Li S L, Yue F J, Qin C Q, Buckerfield S, Zeng J. Rainfall driven nitrate transport in agricultural karst surface river system: insight from high resolution hydrochemistry and nitrate isotopes. *Agriculture, Ecosystems & Environment*, 2020, **291**: 106787
60. Wollheim W M, Mulukutla G K, Cook C, Carey R O. Aquatic nitrate retention at river network scales across flow conditions determined using nested *in situ* sensors. *Water Resources Research*, 2017, **53**(11): 9740–9756
61. Liu C, Li R, Fu Y. Nutrient retention in agricultural headwater stream: artificial manipulation of main-channel morphology and hydrologic condition. *Environmental Science and Pollution Research International*, 2022, **29**(55): 83004–83019
62. Wollheim W M, Harms T K, Robison A L, Koenig L E, Helton A M, Song C, Bowden W B, Finlay J C. Superlinear scaling of riverine biogeochemical function with watershed size. *Nature Communications*, 2022, **13**(1): 1230
63. Yan X, Han H, Qiu J, Zhang L, Xia Y, Yan X. Suburban agriculture increased N levels but decreased indirect N₂O emissions in an agricultural-urban gradient river. *Water Research*, 2022, **220**: 118639
64. Mulholland P J, Helton A M, Poole G C, Hall R O, Hamilton S K, Peterson B J, Tank J L, Ashkenas L R, Cooper L W, Dahm C N, Dodds W K, Findlay S E G, Gregory S V, Grimm N B, Johnson S L, McDowell W H, Meyer J L, Valett H M, Webster J R, Arango C P, Beaulieu J J, Bernot M J, Burgin A J, Crenshaw C L, Johnson L T, Niederlehner B R, O'Brien J M, Potter J D, Sheibley R W, Sobota D J, Thomas S M. Stream denitrification across biomes and its response to anthropogenic nitrate loading. *Nature*, 2008, **452**(7184): 202–205
65. Wang W, Li Z, Shi P, Zhang Y, Pan B, Li P, Ding S, Li J, Bi Z, Wang X. Vegetation restoration and agricultural management to mitigate nitrogen pollution in the surface waters of the Dan River, China. *Environmental Science and Pollution Research International*, 2021, **28**(34): 47136–47148
66. Zhang Q, Fisher T R, Buchanan C, Gustafson A B, Karrh R R, Murphy R R, Testa J M, Tian R, Tango P J. Nutrient limitation of phytoplankton in three tributaries of Chesapeake Bay: detecting responses following nutrient reductions. *Water Research*, 2022, **226**: 119099
67. Wollheim W M, Bernal S, Burns D A, Czuba J A, Driscoll C T, Hansen A T, Hensley R T, Hosen J D, Inamdar S, Kaushal S S, Koenig L E, Lu Y H, Marzadri A, Raymond P A, Scott D, Stewart R J, Vidon P G, Wohl E. River network saturation concept: factors influencing the balance of biogeochemical supply and demand of river networks. *Biogeochemistry*, 2018, **141**(3): 503–521
68. Marcé R, von Schiller D, Aguilera R, Martí E, Bernal S. Contribution of hydrologic opportunity and biogeochemical reactivity to the variability of nutrient retention in river networks. *Global Biogeochemical Cycles*, 2018, **32**(3): 376–388
69. Zhao Y, Xia Y, Ti C, Shan J, Li B, Xia L, Yan X. Nitrogen removal capacity of the river network in a high nitrogen loading region. *Environmental Science & Technology*, 2015, **49**(3): 1427–1435
70. Xia Y, She D, Zhang W, Liu Z, Wu Y, Yan X. Improving denitrification models by including bacterial and periphytic biofilm in a shallow water-sediment system. *Water Resources Research*, 2018, **54**(10): 8146–8159
71. Li W, Zhai L, Lei Q, Wollheim W M, Liu J, Liu H, Hu W, Ren T, Wang H, Liu S. Influences of agricultural land use composition and distribution on nitrogen export from a subtropical watershed in China. *Science of the Total Environment*, 2018, **642**: 21–32
72. Mineau M M, Wollheim W M, Stewart R J. An index to characterize the spatial distribution of land use within watersheds and implications for river network nutrient removal and export. *Geophysical Research Letters*, 2015, **42**(16): 6688–6695
73. Kellogg D Q, Gold A J, Cox S, Addy K, August P V. A geospatial approach for assessing denitrification sinks within

- lower-order catchments. *Ecological Engineering*, 2010, **36**(11): 1596–1606
74. Chen Z, Zhang Z, Zhang T, Zhou S, Zhang Y, Dong W, Yu M, Zhang Y, Zhang J. Temporal and spatial distribution characteristics and driving factors of denitrification bacterial community structure from landscape water in Hebei province: taking Shijiazhuang as example. *Environmental Sciences*, 2022, **43**(2): 813–825 (in Chinese)
75. Chen L, Zhao Z, Li J, Wang H, Guo G, Wu W. Effects of muddy water irrigation with different sediment particle sizes and sediment concentrations on soil microbial communities in the Yellow River Basin of China. *Agricultural Water Management*, 2022, **270**: 107750
76. Van Breemen N, Boyer E W, Goodale C L, Jaworski N A, Paustian K, Seitzinger S P, Lajtha K, Mayer B, van Dam D, Howarth R W, Nadelhoffer K J, Eve M, Billen G. Where did all the nitrogen go? Fate of nitrogen inputs to large watersheds in the northeastern USA *Biogeochemistry*, 2002, **57**(1): 267–293
77. Marti E, Aumatell J, Godé L, Poch M, Sabater F. Nutrient retention efficiency in streams receiving inputs from wastewater treatment plants. *Journal of Environmental Quality*, 2004, **33**(1): 285–293
78. Shrestha S, Bhatta B, Shrestha M, Shrestha P K. Integrated assessment of the climate and landuse change impact on hydrology and water quality in the Songkhram River Basin, Thailand. *Science of the Total Environment*, 2018, **643**: 1610–1622
79. Garnier J, Ramarson A, Billen G, Théry S, Thiéry D, Thieu V, Minaudo C, Moatar F. Nutrient inputs and hydrology together determine biogeochemical status of the Loire River (France): current situation and possible future scenarios. *Science of the Total Environment*, 2018, **637–638**: 609–624
80. Gao X, Chen N, Yu D, Wu Y, Huang B. Hydrological controls on nitrogen (ammonium versus nitrate) fluxes from river to coast in a subtropical region: observation and modeling. *Journal of Environmental Management*, 2018, **213**: 382–391
81. Zhou Y, Xu J F, Yin W, Ai L, Fang N F, Tan W F, Yan E L, Shi Z H. Hydrological and environmental controls of the stream nitrate concentration and flux in a small agricultural watershed. *Journal of Hydrology*, 2017, **545**: 355–366
82. Zhang Y Y, Shao Q X, Ye A Z, Xing H T, Xia J. Integrated water system simulation by considering hydrological and biogeochemical processes: model development, with parameter sensitivity and autocalibration. *Hydrology and Earth System Sciences*, 2016, **20**(1): 529–553
83. Zhang Y, Xia J, Shao Q, Li L, Yen H, Zhai X, Zhao T, Lin K. Uncertainty analysis for integrated water system simulations using GLUE with different acceptability thresholds. *Science China: Technological Sciences*, 2021, **64**(8): 1791–1804
84. Wang L, Stuart M E, Lewis M A, Ward R S, Skirvin D, Naden P S, Collins A L, Ascott M J. The changing trend in nitrate concentrations in major aquifers due to historical nitrate loading from agricultural land across England and Wales from 1925 to 2150. *Science of the Total Environment*, 2016, **542**(Part A): 694–705
85. Destouni G, Persson K, Prieto C, Jarsjö J. General quantification of catchment-scale nutrient and pollutant transport through the subsurface to surface and coastal waters. *Environmental Science & Technology*, 2010, **44**(6): 2048–2055
86. Wang Y, Liu X, Li Y, Liu F, Shen J, Li Y, Ma Q, Yin J, Wu J. Rice agriculture increases base flow contribution to catchment nitrate loading in subtropical central China. *Agriculture, Ecosystems & Environment*, 2015, **214**: 86–95
87. Ti C, Gao B, Luo Y, Wang S, Chang S X, Yan X. Dry deposition of N has a major impact on surface water quality in the Taihu Lake region in southeast China. *Atmospheric Environment*, 2018, **190**: 1–9
88. Xu W, Liu L, Cheng M, Zhao Y, Zhang L, Pan Y, Zhang X, Gu B, Li Y, Zhang X, Shen J, Lu L, Luo X, Zhao Y, Feng Z, Collett J L Jr, Zhang F, Liu X. Spatial-temporal patterns of inorganic nitrogen air concentrations and deposition in eastern China. *Atmospheric Chemistry and Physics*, 2018, **18**(15): 10931–10954
89. Zhan X, Bo Y, Zhou F, Liu X, Paerl H W, Shen J, Wang R, Li F, Tao S, Dong Y, Tang X. Evidence for the importance of atmospheric nitrogen deposition to eutrophic Lake Dianchi, China. *Environmental Science & Technology*, 2017, **51**(12): 6699–6708
90. Ti C, Gao B, Luo Y, Wang X, Wang S, Yan X. Isotopic characterization of $\text{NH}_x\text{-N}$ in deposition and major emission sources. *Biogeochemistry*, 2018, **138**(1): 85–102
91. Chen D, Shen H, Hu M, Wang J, Zhang Y, Dahlgren R A. Legacy nutrient dynamics at the watershed scale: principles, modeling, and implications. *Advances in Agronomy*, 2018, **149**: 237–313
92. Van Meter K J, Van Cappellen P, Basu N B. Legacy nitrogen may prevent achievement of water quality goals in the Gulf of Mexico. *Science*, 2018, **360**(6387): 427–430