

# SOIL NITROGEN CYCLING AND ENVIRONMENTAL IMPACTS IN THE SUBTROPICAL HILLY REGION OF CHINA: EVIDENCE FROM MEASUREMENTS AND MODELING

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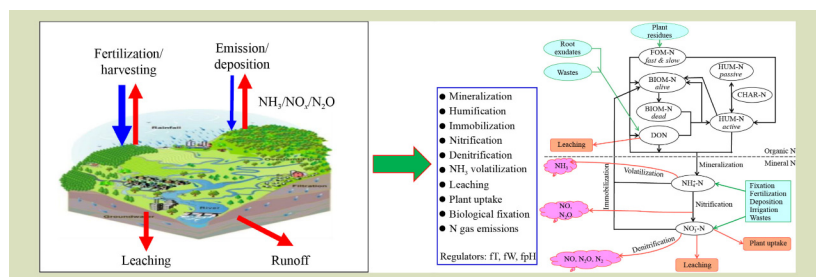
## KEYWORDS

nitrogen cycling, soil nitrogen, nitrogen deposition, greenhouse gases emission, non-point source pollution, nitrogen use efficiency

## HIGHLIGHTS

- Soil nitrogen fluxes and influencing factors were reviewed in the subtropical hilly regions.
- Fertilizer application and atmospheric deposition contributed largely to soil nitrogen input.
- High gaseous, runoff and leaching losses of soil nitrogen were measured.
- Soil nitrogen cycles are well modelled with the Catchment Nutrients Management Model.

## GRAPHICAL ABSTRACT



## ABSTRACT

The subtropical hilly region of China is a region with intensive crop and livestock production, which has resulted in serious N pollution in soil, water and air. This review summarizes the major soil N cycling processes and their influencing factors in rice paddies and uplands in the subtropical hilly region of China. The major N cycling processes include the N fertilizer application in croplands, atmospheric N deposition, biological N fixation, crop N uptake, ammonia volatilization,  $N_2O/NO$  emissions, nitrogen runoff and leaching losses. The catchment nutrients management model for N cycle modeling and its case studies in the subtropical hilly region were also introduced. Finally, N management practices for improving N use efficiency in cropland, as well as catchment scales are summarized.

Received November 30, 2021;

Accepted April 11, 2022.

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## 1 INTRODUCTION

Nitrogen is an essential element for all the organisms on the earth. For providing enough food for the increasing human population in the world, N fertilizer is extensively used to increase crop yields. Globally, nitrogen fertilizer application has increased from 60 Tg in 1980 to 106 Tg in 2015<sup>[1]</sup>. In China, the increase of N fertilizer application was rapid from 12 Tg in 1980 to 31 Tg in 2015, although the N fertilizer application is relatively stable in recent years<sup>[1]</sup>. When the N fertilizers are applied to soil, a series of reactions occur, such as ammonia (NH<sub>3</sub>) volatilization, nitrification and denitrification, leaching and runoff losses. All of these reactions or processes cause considerable nitrogen losses, which also cause serious N pollution in soil, air and water. For example, excessive N fertilizer application has caused significant soil acidification in croplands in China, with an average decrease of 0.5 units in the 20 years from the 1980s to the 2000s<sup>[2]</sup>. High NH<sub>3</sub> emissions from agricultural production also caused PM<sub>2.5</sub> pollution in many cities. According to Gu et al.<sup>[3]</sup>, NH<sub>3</sub> emission reduction can be more cost-effective than nitrogen oxides for reducing PM<sub>2.5</sub> pollution in the air. For the water pollution, as reported by Yu et al.<sup>[4]</sup>, the N losses from agricultural sources were an important source of the water N pollution in China, accounting for 59% of the total N load in water.

The subtropical hilly region of China is a region with high intensity of agricultural production. It is an important region for paddy rice, vegetables, green tea, and pig production. Thus, N fertilizers are commonly used excessively to obtain high yields<sup>[5,6]</sup>. The unsuitable treatment of livestock wastes had caused high NH<sub>3</sub> emissions<sup>[7]</sup>, and the discharge of slurry into streams and rivers directly in the region. This region also has high precipitation, and with mainly low hills, and thus catchments are highly developed in this region<sup>[8,9]</sup>. The N pollution in rivers and lakes was serious in recent years in the subtropical regions, mainly caused by the N fertilizer application, animal wastes and atmospheric N deposition<sup>[4,10]</sup>. N pollution is not only serious in surface water, but also in ground water<sup>[6]</sup>. The water pollution in this region is a serious threat to water ecology and the safety of drinking water. The soil acidification was seriously affected by N fertilizer application<sup>[2]</sup> due to that the soil pH in the region is normally low caused by cations leaching under the weather with high temperature and high precipitation. The high N fertilizer application also causes high NH<sub>3</sub><sup>[11]</sup> as well as N<sub>2</sub>O emissions<sup>[12]</sup>, and thus induced PM<sub>2.5</sub> pollution and climate change effects.

In the recent years, many studies have been conducted in the

subtropical hilly region to understand the soil N cycling processes and their environmental impacts. Many of these studies were conducted by measuring N fluxes in field and catchment scales, which are important to understand the processes and influencing factors of N cycles in the subtropical hilly region. As an extension of the N fluxes measurements in the field scale to the region scale, it is important to model soil N cycles using models. There were still few works conducted to model soil N cycles in the subtropical hilly region. In this review, the progresses of the soil N cycle and their environmental impacts based on measuring and modeling studies in the subtropical hilly region are summarized. The regional N management measurements are also reviewed for improving N use efficiency (NUE) and mitigating N pollution in the subtropical hilly region.

## 2 MAJOR SOIL N CYCLING PROCESSES IN THE SUBTROPICAL HILLY REGION

### 2.1 Nitrogen fertilizer application

The subtropical region of China, accounting for one-quarter of the land area of China, is important for the crop production (e.g., paddy rice, tea and vegetables). With the growth in human population and improvement of living standards, the demands for rice, vegetables and fruits have been dramatically increasing. To increase crop production, N fertilizers have been applied excessively in recent decades in the subtropical hilly region of China. For example, the application of mineral fertilizers had increased from 923 kt in 1980 to 1.42 Mt in 2014 in Hunan Province. In the case of rice cultivation, an average of 180 kg·ha<sup>-1</sup> N is applied in single rice-cropping systems of subtropical China<sup>[13,14]</sup> whereas the average N application rates for early rice and late rice were 170 and 190 kg·ha<sup>-1</sup> N in double rice-cropping systems<sup>[13–16]</sup>. The annual N application rate ranges up to an astonishing 2.6 t·ha<sup>-1</sup> with an average of 553 kg·ha<sup>-1</sup> in the main tea producing areas<sup>[12,17]</sup>. The combined application of organic fertilizer and mineral fertilizer is common in tea production, with the organic N fertilizer accounting for 26%–92% of the total applied N fertilizer<sup>[18,19]</sup>. For vegetable fields, the annual N application rates range from 200 kg·ha<sup>-1</sup> to 1.5 t·ha<sup>-1</sup> with an average of 640 kg·ha<sup>-1</sup> with the organic N fertilizer accounting for 10%–75%<sup>[20,21]</sup>. In addition, the N fertilizer application to fruit crops grew most rapidly, with a rise of 1.4 times from 1998 to 2014, and with an average of 570 kg·ha<sup>-1</sup> N in 2014<sup>[22]</sup>. Although N fertilization deep placement can effectively mitigate N loss and increase NUE, surface broadcasting and multiple fertilization are still commonly applied by local farmers.

## 2.2 Atmospheric N deposition

In the recent decades, high rates of N fertilizer application and the fast development of livestock production have caused large quantities of  $\text{NH}_3$  emissions, while the increasing consumption of fossil fuels caused high emissions of nitrogen oxides in the subtropical hilly region. Thus, the N input from atmospheric dry and wet depositions was also high in the region. The annual total N depositions in the paddy field, tea field and forest sites in a typical subtropical hilly area in central south of China were reported to be as high as 22, 34 and 55  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  N, respectively<sup>[23]</sup>. Wang et al.<sup>[24]</sup> reported that the total (dry and wet) N deposition fluxes were estimated to be 21  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  N in 2014 and 16  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  N in 2015 at rural sites, and 31 and 25  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  N at the urban site in the Three Gorges Reservoir Region. Ouyang et al.<sup>[25]</sup> showed that the total deposition of atmospheric nitrogen dioxide, nitric acid, and particulate nitrate was 13  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  N, which is an important N source in rice paddies in subtropical rice regions. Zhu et al.<sup>[26]</sup> showed that the wet N depositions were 26 and 24  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  N in a typical farmland and forest sites in subtropical hilly region, respectively. Zhu et al.<sup>[11]</sup> showed that atmospheric N deposition was 36  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  N in an agricultural catchment in the central south of China. These results showed that the atmospheric N deposition remained high in the subtropical hilly area, which had become one of the important sources of soil nitrogen.

Atmospheric N deposition reduces the concentration of particulate matter and gaseous pollutants in the atmosphere, which is the main self-purification mechanism for maintaining the cleanliness of the atmosphere. Concurrently, this process migrates air pollutants to terrestrial and aquatic ecosystems, seriously affecting the health of terrestrial and aquatic ecosystems<sup>[27]</sup>. Various studies have shown that as a nutrient element required for plant growth, excessive N deposition will reduce the biodiversity of the ecosystem, lead to eutrophication and acidification of water bodies, and leading to negative environmental impacts such as soil acidification and global climate change<sup>[28]</sup>.

Many scholars have also researched the factors affecting N deposition. Zhu et al.<sup>[29]</sup> showed that the atmospheric N deposition was significantly related to precipitation, N fertilizer use and energy consumption. The precipitation is an important driving factor for the spatial pattern of atmospheric N deposition. The soil N losses caused by  $\text{NH}_3$  volatilization and NO emissions from nitrification and denitrification are important sources of atmospheric N deposition. There were high  $\text{NH}_3$  and NO emissions from croplands in the subtropical hilly region, which has caused high N deposition in the region.

As a mineral nutrient, N input from N deposition accounted for 10% of the average annual N application rate of 360  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  N in double rice-cropping production<sup>[11]</sup>, indicating that N deposition is important in N management in farmland. However, for the natural or semi-natural ecosystems, the increase of N input from atmospheric N deposition may cause many negative impacts, such as soil N and cation losses, biodiversity reduction, increased greenhouse gas emissions, and eutrophication of estuaries and lakes in the subtropical region<sup>[23]</sup>.

## 2.3 Biological N fixation

Biological N fixation (BNF) is the major natural process through which atmospheric  $\text{N}_2$  is converted into forms (e.g.,  $\text{NH}_4^+$ ) that can be used by plants and animals. There are two forms of BNF, symbiotic and non-symbiotic N fixation. Symbiotic N fixation is mostly performed by symbiotic bacteria in root nodules of legumes. Non-symbiotic N fixation, also called free-living BNF, is a crucial way for biological N inputs in non-leguminous crops in agricultural systems. Generally, the free-living BNF rates are lower than symbiotic BNF rates. BNF is affected by the fertilization regimes (N fertilizer) and environmental factors (air temperature, precipitation and N deposition)<sup>[30,31]</sup>. Former studies quantified BNF in forest systems<sup>[32,33]</sup> and cropland in subtropics of China<sup>[34,35]</sup>. N addition suppressed the BNF in Chinese fir plantation in the subtropical region, which was significant in legume trees rather than non-legume trees<sup>[33,36]</sup>. The increase of deposition, in recent decades, most likely reduced total BNF in legume trees<sup>[35]</sup>. It should be noticed that some studies suggested that substrate C:N and C:P stoichiometry rather than substrate N or P was a better way to illustrate the variation in N fixation of forest in South China<sup>[36]</sup>. Also, there was no significant difference in the rate of BNF (6.0–6.2  $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  N) between legume and non-legume trees in mature tropical plantations of China, which was inconsistent with results in other studies<sup>[37–39]</sup>. The rates of BNF in rhizosphere soil were significantly higher in legume than non-legume trees.

The composition and diversity of N-fixing bacteria in tea plantations were reported to be affected by tea age, soil properties and climate factors<sup>[40–42]</sup>. However, the rate and amount of BNF of tea plantations was not adequately defined, so further quantifying the contribution of BNF to NUE, tea yield and quality was needed.

BNF contributes greatly to maintaining the fertility of agricultural soils<sup>[43]</sup>. It is assessed that the rate of BNF in paddy soil ranged from 2.2 to 45  $\text{kg}\cdot\text{ha}^{-1}$  N in the subtropics of

China<sup>[44]</sup>. Variation in BNF was found to be associated with the soil properties, water content, rice cultivars, fertilizer and pH. For example, there was a decreasing trend of BNF from north-east to south for rice paddies in China<sup>[33]</sup>. The potential nitrogenase activity in tropical paddy soil was lower than in warm-temperate and subtropical paddy soils<sup>[45]</sup>. The lower potential for BNF in paddy soil in subtropics was related to soil amorphous aluminum oxide, molybdenum (Mo) and acidification<sup>[33,46,47]</sup>. By direct <sup>15</sup>N<sub>2</sub>-labeling, Wang et al. found that the BNF in the paddy soils ranged from 9.4 to 20 kg·ha<sup>-1</sup> N<sup>[34]</sup>, with higher BNF (22–53 kg·ha<sup>-1</sup> N) was reported in the paddy soil after application of Mo<sup>[47]</sup> and manure<sup>[48]</sup>. Also, hybrid rice cultivars enhanced N fixation (22 and 39 kg·ha<sup>-1</sup> N)<sup>[49]</sup>. The BNF was higher during the period of rice planting (9.5 kg·ha<sup>-1</sup>) than that under fallow (2.1 kg·ha<sup>-1</sup>) in acidic paddy soils<sup>[47]</sup>. A change from forest to tea plantation decreased the N fixation bacteria community and diversity<sup>[50]</sup>. Also, the rhizosphere N-fixing bacteria varies with cultivar, cropping year and soil properties. In general, high input of inorganic fertilizers and soil acidification adversely affect N fixation in the tea plantations. Yet now, it is not clear that the rate of N fixation in tea plantation.

## 2.4 Crop N uptake

Increasing N uptake and NUE of crops could reduce the negative effects of N fertilizer on climate change. Crop N uptake is derived to a large degree from native soil N and N fertilizers. The contribution of N uptake by soil and fertilizer is affected by soil properties, management and climate factors<sup>[51]</sup>.

About 60% of paddy soil in China is in the subtropical region. It is estimated that the N uptake of 180–200 kg·ha<sup>-1</sup> is typically required to achieve high rice yield in the subtropics<sup>[52]</sup>. Kong et al. found that N uptake of rice was 124–238 kg·ha<sup>-1</sup><sup>[53]</sup>. The NUE (the proportion of crop N uptake from N fertilizer to the applied N fertilizer) of subtropical rice ranged from 34% to 66%<sup>[54,55]</sup>. In general, the NUE of early rice was lower than that of late rice in the subtropics, which is associated with the lower N mineralization and more N loss in runoff under the condition of lower temperature and high precipitation<sup>[56,57]</sup>. It has been reported that the NUE in the early and late rice season ranged from 24% to 58% and 37% to 55%, respectively, based on long-term field experiments<sup>[6,58]</sup>. Mi et al. found that the single rice NUE ranged from 30% to 45%<sup>[59]</sup>. Soil properties (indigenous soil N, SOC and pH) affect rice N uptake by changing soil microbial metabolism, enzymatic activity, nitrogen availability and rice root growth<sup>[56,60]</sup>. The higher C:N ratio likely limited rice N uptake, which was derived from the competition for N using by microorganisms. Paddy soil

acidification decreases rice N uptake and NUE, which is caused by lower pH-limited soil enzyme activities and microbial population<sup>[61]</sup>. The subtropical rice NUE increased by over 10% with organic treatment in a long-term experiment (25 years), which is likely to be associated with the increased SOC and it ameliorated soil acidity<sup>[55,59]</sup>. Liming interacted with straw retention was reported to have the potential to increase the yield and N uptake in double rice-cropping systems<sup>[62]</sup>.

The better match of N supply with crop demand by adjusting fertilization method and N fertilizer type is a promising practice to enhance N uptake in rice. It has been confirmed that deep placement of N fertilizer can effectively increase N absorbed by rice in subtropical China<sup>[63,64]</sup>, which is likely to be due to less N loss (i.e., NH<sub>3</sub> volatilization, N runoff and leaching) and high soil N content. Also, it was reported that long-term green manuring enhanced rice N uptake by 18%–62% compared to chemical N fertilizer<sup>[65]</sup>. The benefits of enhancing N uptake in rice by application of enhanced efficiency N fertilizers (i.e., nitrification and urease inhibitors, neem, and slow release fertilizers) have been observed. For example, a meta-analysis of 32 field studies found that the enhanced efficiency N fertilizers increased N uptake by 8% in rice production systems, but greater efficiency was limited by soils with low pH<sup>[66]</sup>. The N uptake in response to polymer-coated urea applied was higher than uncoated N fertilizer, while the above enhancing N uptake varied in early and late rice season, which linked with the effect of temperature and precipitation on N release<sup>[67]</sup>.

In comparison to other crops, tea plants need more N to obtain yield with high quality. However, N uptake of tea ranged from 87 to 120 kg·ha<sup>-1</sup> N, and generally the NUE of tea plants is low (10%–53%)<sup>[68,69]</sup>. Significant soil acidification occurs in the tea production systems<sup>[69]</sup>, which further limits N uptake by tea plants.

## 2.5 Ammonia volatilization

NH<sub>3</sub> volatilization is one of the dominant causes of N loss in crop fields, which increases production costs and causes environmental pollution<sup>[70–72]</sup>. After the soluble N fertilizer application such as ammonium bicarbonate and urea in rice paddies, the content of NH<sub>3</sub> and ammonium N (NH<sub>4</sub><sup>+</sup>-N) in the surface water increased rapidly, which is the internal reason for NH<sub>3</sub> volatilization. NH<sub>3</sub> volatilization was dominated by the NH<sub>4</sub><sup>+</sup>-N concentration in the surface water and generally had a significantly linearly positive correlation for the N input rates<sup>[73]</sup>. Also, NH<sub>3</sub> volatilization is affected by a large number of factors, such as fertilizer application rate, climate conditions

(e.g., temperature and wind speed), and soil properties (e.g., pH and soil type)<sup>[74]</sup>. With the increase in N application rate, temperature, soil moisture and pH, the amount of NH<sub>3</sub> volatilization in rice paddies also increases significantly<sup>[70]</sup>. Zhang et al. found that high NH<sub>3</sub> volatilization intensities of above 30–50 kg·ha<sup>-1</sup> N were concentrated across the subtropical hilly region of China, with about 37% coming from croplands<sup>[75]</sup>. The strong daily NH<sub>3</sub> fluxes occurred after the N fertilizer application and NH<sub>3</sub> volatilization was particularly high in 1–5 days, then declined rapidly to relatively low levels. NH<sub>3</sub> volatilization from rice paddies is greater than from other crop systems, and it is generally considered to be the major N loss pathway from rice paddies<sup>[76]</sup>. Many studies have shown that the cumulative NH<sub>3</sub> losses were observed as 19–62 kg·ha<sup>-1</sup> N, accounted for 17% of the total applied N in single rice cropping systems<sup>[77,78]</sup> whereas 33–85 kg·ha<sup>-1</sup> N and 50–140 kg·ha<sup>-1</sup> N, for the early and late rice, respectively, and accounted for 29% and 36% of the N input on two rice season average<sup>[16,79]</sup>. Also, several studies have reported that the cumulative NH<sub>3</sub> losses from vegetable fields ranged from 0.4 to 55 kg·ha<sup>-1</sup> N, accounted for 9% of the total applied N<sup>[80,81]</sup>, in tea plantations ranged from 5.7 to 49 kg·ha<sup>-1</sup> N, accounted for 11% of the total applied N, and under fruit crops fields ranged from 7.7 to 13 kg·ha<sup>-1</sup> N, accounted for 6% of the total applied N (Table 1).

## 2.6 N<sub>2</sub>O and NO emissions

The subtropic is an important source of N<sub>2</sub>O emissions in China. The subtropical plantation is a hotspot for agricultural N<sub>2</sub>O and NO emissions<sup>[88,89]</sup>. Annual N<sub>2</sub>O emissions in tea plantations in the subtropics ranged from 7 to 80 kg·ha<sup>-1</sup>·yr<sup>-1</sup> N<sup>[5,80]</sup>. NO emissions averaged 8.8 kg·ha<sup>-1</sup>·yr<sup>-1</sup> NO-N for the N fertilizer tea plantations. The average fertilizer-induced emission factor of N<sub>2</sub>O, NO, and N<sub>2</sub>O+NO emissions were 2.1%, 0.8%, and 2.9% in a typical subtropical tea plantation of China, respectively<sup>[90]</sup>. High N<sub>2</sub>O emissions

from tea plantations result from anaerobic conditions, low soil pH and high rate of N fertilizer. Besides, in the recent decades, the woodland and rice paddies had been converted to tea plantations in subtropical China, which would enhance N<sub>2</sub>O emissions<sup>[91,92]</sup>. There are some recommendations to decrease N<sub>2</sub>O and NO emissions from tea plantations by using biochar addition, nitrification and urease inhibitor<sup>[93]</sup>.

The N<sub>2</sub>O emissions from paddy soil were relatively low compared with those from tea plantations. The fertilizer-induced N<sub>2</sub>O emission factor (0.02%–0.42%) is also lower than the IPCC default value of 0.30% for subtropical rice paddy soil in China<sup>[94,95]</sup>.

The irrigation regime and N rates significantly influence N<sub>2</sub>O emissions in paddy soils<sup>[96]</sup>. Generally, intermittent irrigation and midseason drainage often cause increased N<sub>2</sub>O emissions in paddy soils, which is attributed to the enhanced nitrification and denitrification<sup>[97]</sup>. The application of straw has the potential to reduce N<sub>2</sub>O emissions, while the opposite result was observed in the treatment of biochar addition in rice double-cropping systems<sup>[5,15]</sup>.

The N<sub>2</sub>O emissions from forests were lower than croplands<sup>[91,98]</sup>, while the N<sub>2</sub>O emissions from subtropical forest are significantly higher than those in temperate regions<sup>[99,100]</sup>. Consequently, forests likely contribute a large amount of N<sub>2</sub>O emissions in this region. Also, the enhanced N deposition increased N<sub>2</sub>O emission in the subtropical forest<sup>[101,102]</sup>. It has been found that heterotrophic nitrification mainly contribute to the N<sub>2</sub>O emissions of subtropical forests in China<sup>[103]</sup>. The N<sub>2</sub>O emissions are influenced by tree species, soil properties, rainfall and temperature.

The pathways of N<sub>2</sub>O production were studied in different land use types in subtropical region of China. Denitrification was the main cause of N<sub>2</sub>O production in tea plantation soil<sup>[104]</sup>,

**Table 1** Nitrogen fertilizer application and ammonia volatilization

| Crops       | N rate (kg·ha <sup>-1</sup> N) | Method | Organic ratio (%) | NH <sub>3</sub> flux (kg·ha <sup>-1</sup> N) | Mean NH <sub>3</sub> loss rate (%) | References   |
|-------------|--------------------------------|--------|-------------------|--|------------------------------------|--------------|
| Single rice | 300–350                        | B, DP  | –                 | 28.7–49.4                                    | 17.2                               | [82–85]      |
| Early rice  | 120–220                        | B, DP  | –                 | 33.2–89.3                                    | 29.4                               | [6,10,16,79] |
| Late rice   | 120–270                        | B, DP  | –                 | 50.4–140.5                                   | 35.6                               | [6,10,16,79] |
| Tea         | 0–2600                         | Furrow | 26–92             | 5.73–48.8                                    | 10.6                               | [12,17–19]   |
| Vegetable   | 200–1500                       | B      | 10–75             | 0.4–55                                       | 9.2                                | [20,21,81]   |
| Fruit       | 136–570                        | DP     | 25–75             | 7.7–13                                       | 5.5                                | [22,86,87]   |

Note: B, broadcasting; and DP, deep placement.



which mainly attribute to low pH, high  $\text{NO}_3^-$  content and soil moisture. Huang et al. and Zhang et al. found that the contribution of denitrification reached up to 70% and 73% to  $\text{N}_2\text{O}$  production in tea plantations in Zhejiang and Jiangxi Provinces, respectively<sup>[105,106]</sup>. A study using a model of WNNM found denitrification contributed to 77% in tea plantations<sup>[12]</sup>. In contrast, Cheng et al. found that denitrification only contributed to 11%–36% of  $\text{N}_2\text{O}$  production in tea plantations where soil pH was 5.3, whereas the contribution of autotrophic nitrification ranged from 50% to 76%<sup>[107]</sup>. The difference between these results is likely due to soil pH.

In a rice field, Yang et al. found that the synergism of nitrifying microorganisms and denitrifying microorganisms dominated  $\text{N}_2\text{O}$  in the topsoil layer<sup>[108]</sup>. It was estimated that  $\text{NO}$  emission from waterlogged rice field was  $0.001\text{--}0.003 \text{ Tg}\cdot\text{yr}^{-1} \text{ N}$ , mainly from denitrification<sup>[109]</sup>. The change from rice paddy to citrus orchards resulted in that nitrification contributed to most of the  $\text{N}_2\text{O}$  production in the hilly red soil of subtropical region<sup>[110]</sup>. Similar,  $\text{N}_2\text{O}$  emissions in wheat of a wheat-rice cropping system were mainly from nitrification<sup>[111]</sup>. Conversely, Zhang et al., using  $^{15}\text{N}$ -tracing method, found that 55% of  $\text{N}_2\text{O}$  production was from denitrification in upland soil<sup>[112]</sup>. The different contribution of nitrification and denitrification to  $\text{N}_2\text{O}$  production in upland soil was associated with soil C content, pH, inorganic N content and soil moisture content. Besides, Zhang et al. found that denitrification accounted for 49%, 52% and 32% for sweet potato farmland, citrus orchard and vegetable growing farmland, respectively<sup>[91]</sup>.

## 2.7 Nitrogen runoff and leaching losses

Driven by rainfall and artificial drainage, N runoff and leaching losses are generated<sup>[67]</sup>. A former study has shown that the N runoff loss with a conventional fertilizer treatment was  $2.9 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1} \text{ N}$  in a double rice-cropping system in the typical subtropical hilly region of China<sup>[6]</sup>. Zheng et al.<sup>[112]</sup> showed that the N runoff and leaching losses were 10.3 and  $29.6 \text{ kg}\cdot\text{ha}^{-1} \text{ N}$ , respectively, in the whole growing season in a typical sloped red soil upland of Jiangxi. Yue et al.<sup>[113]</sup> showed that the net  $\text{NO}_3^-$  runoff loss of a typical subtropical agricultural watershed was  $34.5 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1} \text{ N}$ , accounting for 15% of the annual fertilizer applied ( $229 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1} \text{ N}$ ). Dong et al.<sup>[114]</sup> showed that fertilization promoted  $\text{NO}_3^-$  leaching by 92 and  $58 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1} \text{ N}$  at 20 and 100 cm deep, respectively. Yang et al.<sup>[115]</sup> showed that early rice TN average runoff loss ( $17 \text{ kg}\cdot\text{ha}^{-1} \text{ N}$ ) and loss rate (11%) are higher than late rice ( $11 \text{ kg}\cdot\text{ha}^{-1} \text{ N}$ , 7.2%) and proved different fertilization management measures have different N runoff losses in

subtropical typical double rice-cropping fields. These results indicated that N runoff and leaching losses must not be ignored. The soil and fertilizer N in the croplands, forest and tea plantations can be lost into surrounding water bodies, which will have adverse impacts on the environment and human health, such as water eutrophication, reduction of biodiversity, toxic algae blooms and deterioration of drinking water quality<sup>[4,116]</sup>.

Soil physicochemical properties, including soil particle size, porosity and humus content, affect N runoff and leaching losses. Soils that are rich in organic matter, with smaller soil particle size and denser soils, have a lower rate of N loss<sup>[117]</sup>. On both tea and bamboo hillslopes, soil temperature and precipitation during the previous 7 days were negatively correlated to leachate nitrate N ( $\text{NO}_3^- \text{-N}$ ) concentrations, whereas the ground water table depth was the opposite. Soil water content and its ratio to field capacity negatively influenced leachate  $\text{NO}_3^- \text{-N}$  concentrations on both hillslopes<sup>[118]</sup>. Steep hillslopes promoted more N runoff loss than gentler slopes. On gentle hillslopes, high precipitation (e.g.,  $> 5 \text{ mm}$ ) was the main influential factor for N runoff loss, but as the slope gradient increases, the frequency of rainfall events became the major controlling factor, implying that N runoff loss from steep hillslopes can be sensitive to even small rainfall events<sup>[119]</sup>.

The N runoff and leaching losses are affected by factors such as climatic conditions and fertilization methods. The amount and rate of N loss increased sharply with the increase of rainfall intensity<sup>[117]</sup>. Under different rainfall intensities,  $\text{NH}_4^+ \text{-N}$  was the main form of N loss from surface runoff<sup>[117]</sup>, and  $\text{NO}_3^- \text{-N}$  was the main form of N loss from leaching<sup>[117,120]</sup>. The runoff volume was influenced both by precipitation and irrigation. There is no simple linear relationship between precipitation and runoff of rice paddies. This variation was mainly controlled by the times of precipitation and irrigation. High runoff volume occurred if the precipitation event occurred shortly after irrigation. However, if precipitation was delayed for a week or more, heavy precipitation may not produce large runoff volumes. Abundant regional precipitation leads to intense soil erosion and severe N leaching on red soil sloping uplands<sup>[67]</sup>. Application of controlled-release urea reduced N surface runoff losses<sup>[121]</sup>.

During irrigation,  $\text{NO}_3^- \text{-N}$  tends to move downward with irrigation water and becomes the main form of N leaching<sup>[120,121]</sup>. The concentration of  $\text{NO}_3^- \text{-N}$  in the leaching solution became lower as irrigation proceeded, probably because of the continuous decrease in soil redox potential due

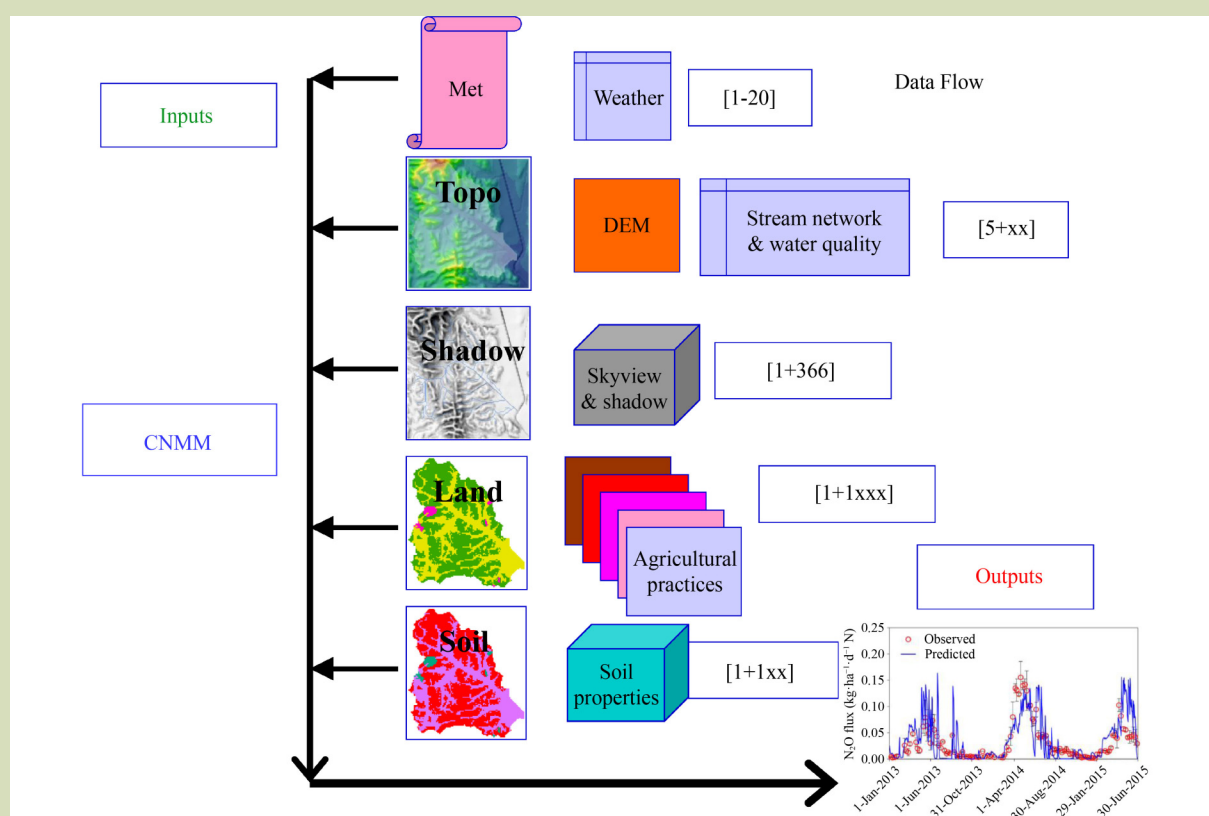
to continuous saturation of the irrigated soil<sup>[121]</sup>. Studies under fertilizer application and clear water irrigation conditions have concluded that the higher the irrigation intensity the higher the  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_3^-\text{-N}$  and total N leaching from the leaching solution and the lower the  $\text{NH}_4^+\text{-N}$  and  $\text{NO}_3^-\text{-N}$  contents in the soil, increasing the risk of N leaching<sup>[120]</sup>. Compared to the conventional flooding system, controlled irrigation saved 34% of the irrigation water and greatly reduced nonproductive water consumption (evaporation + runoff + deep percolation) by 17%–20%.

### 3 NITROGEN CYCLE MODELING

#### 3.1 Model introduction

Given that there have been few modeling studies on soil N cycles in the subtropical regions, the CNMM (catchment

nutrients management model) was developed based on the field measurements in the subtropical hilly region (Fig. 1). The CNMM is a physically-based and spatially-distributed catchment biogeochemical model for simulating energy balance, water, C, N and P cycling in catchment ecosystems (Fig. 2). The CNMM can simulate the complete soil N cycle (including BNF, plant uptake, organic matter mineralization, soil microbial fixation, nitrification, denitrification,  $\text{NH}_3$  volatilization, leaching and runoff loss) at field and catchment scales. It can also simulate the emissions of nitrogen oxides, nitrous oxide and dinitrogen in soil nitrification and denitrification reactions (Fig. 3). BNF is predicted using annual net primary productivity. Nitrification, denitrification and  $\text{NH}_3$  volatilization are simulated based on first-order kinetic equations. Urea hydrolysis process uses first-order kinetics equation description. The crop N uptake process is simulated by WATTS and HANKS methods. Soil ammonium ion adsorption is described by Freundlich. N leaching is described



**Fig. 1** Model structure and data flow of the catchment nutrients management model (CNMM). The structure of the model includes the following parts: (1) hydrology (evaporation and transpiration, snow melting, runoff, infiltration, lateral flow, base flow, stream discharge), (2) soil-water temperature (energy balance), (3) plant growth, (4) plant-soil-water C-N-P Cycling (e.g., SOM decomposition and humification, immobilization, dry/wet N deposition, nitrification, denitrification, plant uptake,  $\text{CO}_2$ - $\text{CH}_4$ - $\text{NH}_3$ - $\text{NO}_x$ - $\text{N}_2\text{O}$ - $\text{N}_2$  emissions, leaching), (5) water and C-N-P transport and loss via runoff, lateral flow, base flow and stream flow, and (6) land management (e.g., planting, harvest, tillage, burning, fertilization, irrigation, wastes treatment).

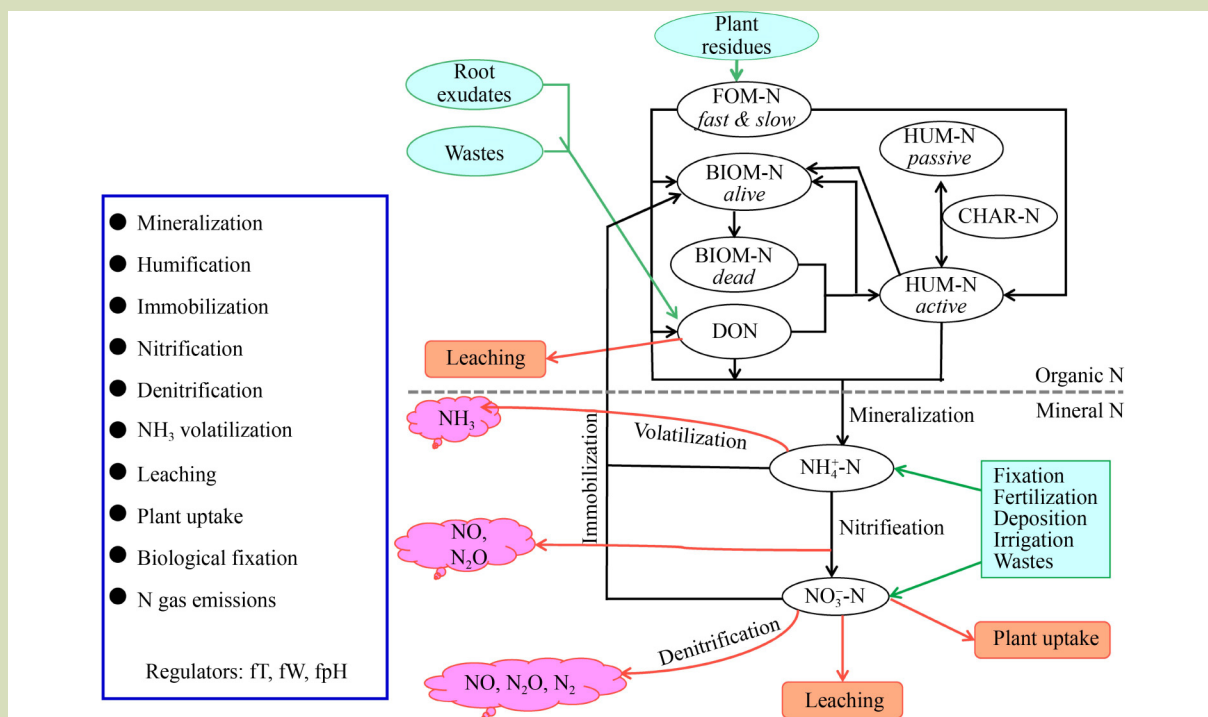


Fig. 2 Nitrogen cycles simulated in the catchment nutrients management model.

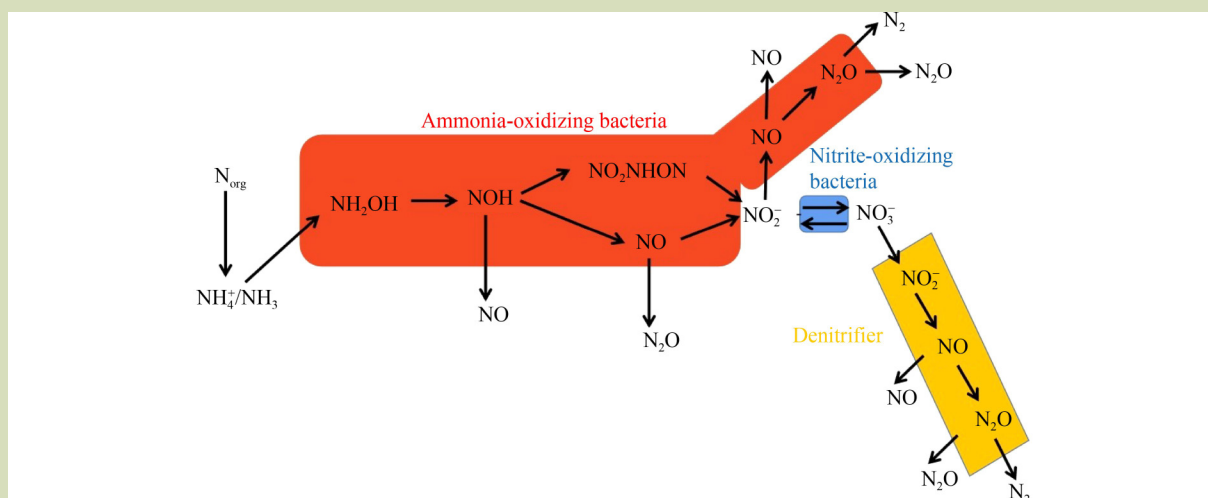


Fig. 3 Production pathways of soil NO,  $\text{N}_2\text{O}$  and  $\text{N}_2$ .

by linear storage capacity transfer equation to describe the vertical or lateral movement of N in soil. Details for soil N cycle modeling are described by Li et al.<sup>[122]</sup>.

CNMM can be applied to simulate the response of stream water quality to land management practices, evaluate climate change impacts on catchment agricultural activities, and

potentially quantify the relationship between the stoichiometric ratio of biogenic elements in the earth's surface materials and agricultural productivity. It runs at the time intervals of several hours, typically 3 h. CNMM was redeveloped from the water and nitrogen management model<sup>[123]</sup>, greatly expanding its structure and function through a close coupling with the distributed hydrological soil vegetation model<sup>[46]</sup> for



catchment hydrology and energy balance, enhanced stream water quality model<sup>[110]</sup> for stream water quality, and inclusion of the Manure-DNDC model<sup>[124]</sup> for waste production and treatment in the catchment. Therefore, CNMM is capable of simultaneously simulating energy balance, hydrology, biogenic elements cycling, stream water quality and land management at catchment scales at hourly intervals. CNMM is mostly coded using C language (ANSI C compatible), with an extendable module-based design, contains approximately 50 thousand lines of source code, and is freely available to the public domain by direct request to the developers.

The CNMM model simulates numerous catchment processes (Fig. 1) in spatial grids and stream networks at any given temporal scales and features with three-dimensional modules of hydrology and solute transport. The hydrology modules run in both the catchment spatial grids and stream networks. The stream water quality modules only run in stream networks. However, the rest modules remain confined to the catchment spatial grids.

The solute transport module simulates soluble C, N and P species, including dissolved organic C, dissolved organic N, dissolved organic P,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and labile P, traveling in soils, surface runoff and stream water. It interacts with a number of modules: plant growth, water, C, N and P cycling (including fresh organic matter decomposition, soil organic matter decomposition and accumulation, wet and dry deposition, nitrification and denitrification, emissions of  $\text{CO}_2$ ,  $\text{NH}_3$ ,  $\text{N}_2\text{O}$ , NO and  $\text{N}_2$  in the plant-soil-water system and sorption/desorption), agricultural practices (including sowing, harvest, tillage, fertilization, irrigation and waste management), and stream water quality (Fig. 2). The hydrology module models snow melting, precipitation interception, canopy evaporation, soil evaporation, plant transpiration, soil infiltration, soil redistribution, surface runoff or overland flow, unsaturated soil water flow, saturated shallow groundwater flow and channel flow.

CNMM is able to simulate the complete soil nitrogen cycle of a watershed system: BNF, plant uptake, organic matter mineralization, soil microbial fixation, nitrification, denitrification,  $\text{NH}_3$  volatilization, leaching, surface loss, and lateral seepage loss (Fig. 1). Meanwhile,  $\text{N}_2\text{O}$ , NO and  $\text{N}_2$  emissions from soil nitrification and denitrification reactions are simulated. The inorganic forms of N ( $\text{NH}_4^+$ -N and  $\text{NO}_3^-$ -N) are very active in the soils and water bodies of the watershed system, and can migrate in the soils and water bodies of the watershed system together with dissolved organic N, especially the fast movement of  $\text{NO}_3^-$ -N.

CNMM can simulate catchments with spatial areas from 1 to 500  $\text{km}^2$ , at soil depths of 1–10 m (typically 4 m, at which soil temperature is approximately constant to the regional annual mean air temperature), time duration of 1–100 years (typically 30, 60 and 90 years), time intervals of 1–24 h (typically 3 h), and spatial grid size of 1–100 m (typically 10–20 m). The upper boundary in the CNMM is set to be on the top of the vegetation canopy, and the lower boundary is at the bottom of shallow groundwater affected by precipitation. The horizontal water and mass flow at any single grid can exchange with adjacent grids in four directions of  $0^\circ$ ,  $90^\circ$ ,  $180^\circ$  and  $270^\circ$ .

CNMM input data include meteorological variables, plant physiologic parameters, soil properties, land management practices, digital elevation model, topographical shadow, stream networks, water quality and waste treatment site information. Its output data contain model state and flux variables in spatial grids and stream networks at the simulation time intervals and a daily, monthly and yearly basis (Fig. 1). The simulation data at user-interested sites in spatial grids and nodes (such as stream junctions and outlets) in stream networks can also be exported for the model calibration and validation of the observed ecological processes.

### 3.2 Model application

The CNMM has been applied mainly to simulate the transformation processes of water and nitrogen in the soil in Feiyue catchment in the subtropical hilly region. Continuous observations of NO and  $\text{N}_2\text{O}$  emissions in tea plantation from 2013 to 2015 were conducted in Feiyue catchment. In the CNMM, the soil NO and  $\text{N}_2\text{O}$  emissions simulation module mainly calculates the NO emission from two processes: nitrification and nitrite chemical decomposition, and two  $\text{N}_2\text{O}$  emission processes: nitrification and denitrification (Fig. 3).

As shown in Fig. 3, CNMM simulation of 0–15 cm soil moisture content,  $\text{NH}_4^+$ -N content,  $\text{NO}_3^-$ -N content and 5 cm soil temperature of tea plantation soil is consistent with the observed values. The variable with a great difference between simulated and observed values is the  $\text{NH}_4^+$ -N content in 0–15 cm soil, which mainly occur in the spring of 2013 (March–May). The dominant factor could be the spatial variability of fertilization or the difference between the actual fertilization amount and the planned fertilization amount.

NO and  $\text{N}_2\text{O}$  emissions from the nitrification process mainly occur in ammonia-oxidizing archaea/ammonia-oxidizing bacteria action stage (or  $\text{NH}_4^+$ -N to nitrite nitrogen stage). The comparison between CNMM simulation results and observed

values are shown in Fig. 4. The simulation results of NO emission are acceptable ( $R^2 = 0.44$ ,  $P < 0.05$ ). In addition, nitrite is unstable and produces NO emission by chemical decomposition under acidic conditions and high temperatures. This simulation showed that this is the main process, accounting for more than 55% of the total NO emission. In addition, the simulation results of CNMM on  $N_2O$  emission ( $R^2 = 0.52$ ,  $P < 0.001$ ) from tea plantations are better than that of NO, and it is calculated that the  $N_2O$  emission contribution of the denitrification process accounts for about 75% of the total emission.

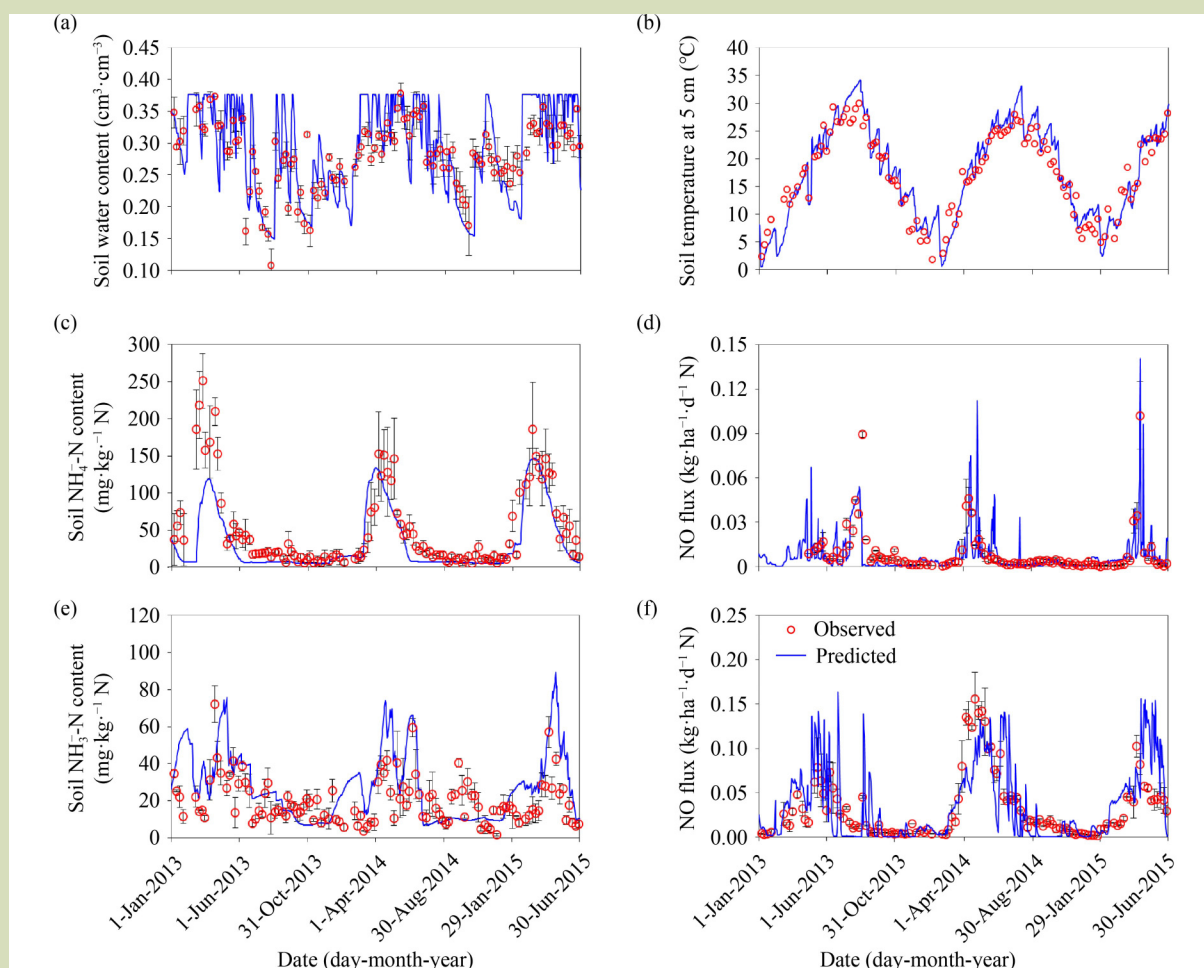
CNMM was applied to simulate Nanyue catchment from January 2011 to December 2013 (the warm-up period is the whole year of 2011). The comparison between the simulated and observed river flow at the main outlet of the catchment is shown in Fig. 5, and the simulated results are consistent with

the observed results, especially CNMM can reliably simulate each flood peak process. CNMM showed good simulation results on the total N ( $1.2\text{--}8.1\text{ mg}\cdot\text{L}^{-1}\text{ N}$ ) and total P ( $0.005\text{--}0.175\text{ mg}\cdot\text{L}^{-1}\text{ P}$ ) of the main outlet of the catchment, especially the total N. However, the simulation of total P was overestimated in the second half of 2013 for undetermined reasons. In addition, CNMM can also satisfactorily simulate the dynamics of groundwater level (Fig. 5).

## 4 REGIONAL NITROGEN MANAGEMENT

### 4.1 Cropland N management

Given the negative effects of soil N cycles and the high application rate of N fertilizer in croplands in the subtropical



**Fig. 4** Simulated and observed values of soil water content (a), temperature at 5 cm (b), ammonium nitrogen content (c), NO (d), nitrate nitrogen content (e), and  $N_2O$  (f) emission fluxes of tea plantation soils in the Feiyue catchment.

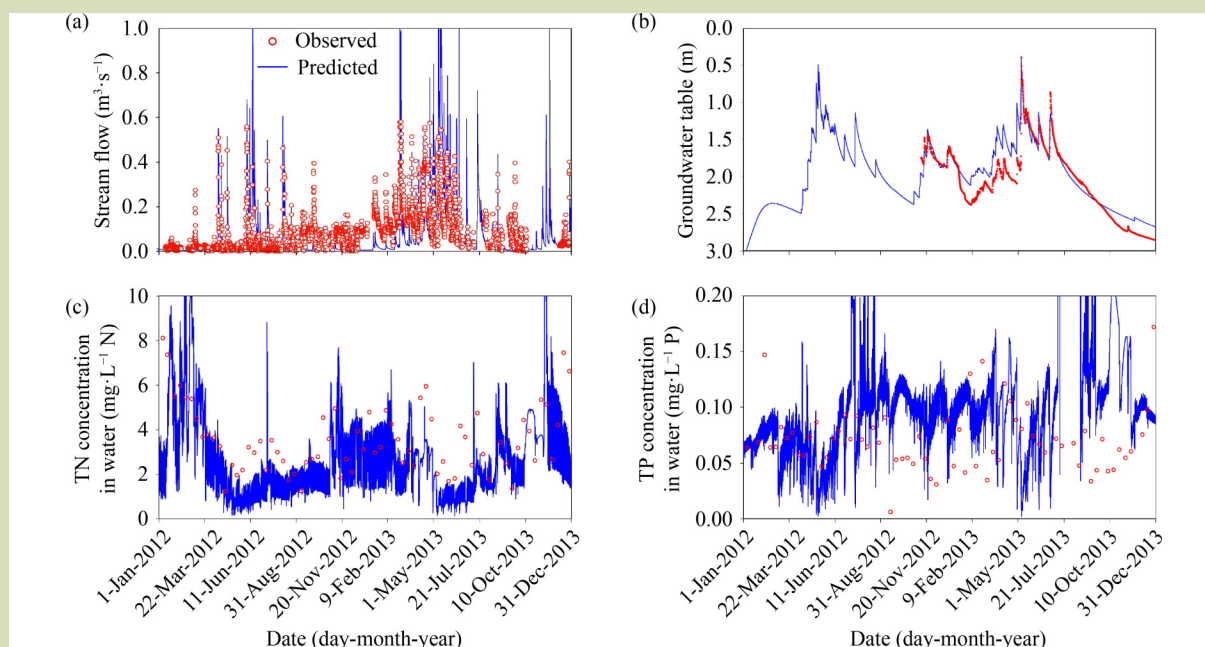


Fig. 5 Catchment nutrients management model simulated and observed values of stream flow (a), groundwater table (b), TN (c) and TP (d) concentrations at the main outlet of Nanyue catchment.

hilly region, it is important to optimize N fertilizer application in croplands in order to reduce the negative impacts<sup>[64,65]</sup>. With the carbon peak and carbon neutrality goals for the Chinese government in 2030 and 2060, respectively, limiting N fertilizer application rates is crucial to reduce the  $\text{CO}_2$  emissions during fertilizer production and  $\text{N}_2\text{O}$  emissions induced by soil nitrification and denitrification<sup>[15,125]</sup>.

#### 4.1.1 Optimize N fertilizer type

Currently, urea is the major N fertilizer used in the subtropical hilly region. Although urea is convenient to apply and is relatively cheap, it also suffers from a high proportion of loss by  $\text{NH}_3$  volatilization, nitrification and denitrification, as well as leaching and runoff losses. For improving NUE, some new types of N fertilizers have shown better performance. For example, the control-release N fertilizers were used to reduce N fertilizer application rate by up to 33%<sup>[126]</sup>, increase NUE by up to 150%<sup>[127]</sup>, reduce  $\text{NH}_3$  volatilization by up to 80%<sup>[128]</sup>, reduce  $\text{N}_2\text{O}$  emissions by up to 27%, reduce N runoff loss by up to 24%<sup>[129]</sup>. The combined application of N fertilizer with urea inhibitor or nitrification inhibitor can also increase NUE while reducing N losses<sup>[130,131]</sup>.

#### 4.1.2 Reduce top dressing N fertilizer

Due to the common use of urea usually by top dressing, a large

portion of N applied is lost. Deep application of N fertilizer has been shown to largely avoid  $\text{NH}_3$  volatilization, reduce N application rate and increase NUE. Thus, deep application of N should be encouraged. In recent years, some new machines have been developed for deep application of N fertilizers.

#### 4.1.3 Recycle of organic fertilizers

Organic fertilizers are usually slow-released, and thus have a lower N loss rate than mineral N fertilizer. The use of organic fertilizer can also recycle nutrients, thus can avoid resource consumption and reduce greenhouse gas emissions during fertilizer production. The application of organic fertilizer can also increase soil organic matter and favor soil productivity.

#### 4.1.4 Use of biological N fixation

Nitrogen-fixing crops (such as soybean and alfalfa) can take advantage of symbiosis with rhizobium to fix N from the atmosphere, and thus can largely reduce the N application rate. Intercropping of legumes with non-legumes has also resulted in increased NUE and decreased N losses. The incorporation of legumes as green manure into soils can also be useful for reducing N application rate as well as improving soil quality. Also, soil N-fixing microorganisms by fixing N from the atmosphere contribute a substantial portion to the soil N pool.

It is also an important objective to increase soil microbial N fixation to reduce the application of mineral N fertilizer.

## 4.2 Catchment N management

Based on the lessons of past failures in the prevention and control of N pollution in large downstream and lakes, considerable research on catchment N management has shifted from the aquatic N pollutant removal to control of terrestrial N emission<sup>[132,133]</sup>, and controlling terrestrial N pollutant emission and removing aquatic N pollutant has now been widely recognized as the prevention and control measures optimizing catchment N management.

Decreasing the total N input, and reducing N surface and subsurface losses from farmland, livestock production and households are the primary means for controlling terrestrial N pollution in catchments. According to recent research in a typical subtropical agricultural catchment, the catchment N input intensity was as high as 202 kg·ha<sup>-1</sup>·yr<sup>-1</sup>, in which fertilizer and animal feed N, as the largest source of N input, contributed to 46%–75% of total catchment N input<sup>[134]</sup>. The higher catchment N input intensity led to 78% water sample categorized as N pollution (> 1.0 mg·L<sup>-1</sup>, Chinese surface water quality standard of GB3838-2002)<sup>[135]</sup>. Controlling total N input can be effectively achieved through the optimization and adjustment of agricultural industrial structure. In the agricultural catchment, the excessive total N input was mainly induced by the low N recycling efficiency caused by the disconnection between planting and breeding industries, and then constructing a recycling agriculture system of crop production and livestock production helps to control total N input at the catchment scale<sup>[136]</sup>. The reduction of N surface and subsurface emission was aimed to lower N loss per unit area across the landscape through improving agronomic efficiencies of applied N. In addition, intercepting and absorbing water N is important for optimized catchment N management. The most common technologies are to adopt ecological engineering measures, such as artificial wetlands, ecological ditches, ecological buffering and interception zone, appropriate to local conditions, to intercept N transport in stream networks, and absorb N pollutants in downstream receiving water bodies<sup>[137–139]</sup>. In subtropical hilly China, the ecological wetland technology with planting *Myriophyllum elatinoides* was developed, and showed an N removal efficiency in water reaches as high as 98%<sup>[135]</sup>. This plant has been widely used to intercept and absorb N pollutants for prevention and control of non-point source N pollution in the subtropical

catchments.

Another problem for catchment N management in the subtropical hilly China is the lack of systematic management strategies and tools. In terms of strategies, although provincial and municipal governments have formulated a series of mandatory standards for the treatment of livestock and poultry wastes, there are few regulations limiting the type, amount, application period and mode for the fertilizers used in the farmlands. In terms of tools, the subtropical hilly China does not have scientific decision-making tools for formulating a catchment N management plan, therefore selecting and setting the types and intensities of technical intervention of catchment N management are still in the stage of qualitative empirical judgment. Some scientists have recognized these problems, and distributed catchment models (e.g., CNMM) based on N cycling processes and the watershed non-point source pollution decision support system based on an artificial intelligence algorithm are now available to help address these problems<sup>[140]</sup>.

## 5 CONCLUSIONS

In the subtropical hilly region of China, due to the high N fertilizer application rates, high N deposition and relatively low NUE in the croplands, the soil N cycle processes (e.g., NH<sub>3</sub> volatilization, nitrification and denitrification, N runoff and leaching) have caused the serious N pollution in air and water. By conducting field measurements, the fluxes of major soil N cycle processes have been quantified and their influencing factors clarified. A process-based model, CNMM, has been developed to simulate soil N cycles in the subtropical hilly region, and has shown sound agreement between simulation and observation at field and catchment scales. For mitigating N pollution in this region, cost-effective measurements in the field and catchment scale should be used as suggested by various studies.

In the future, considering the carbon peak and carbon neutrality policies of China, promoting NUE and mitigating N pollution will be increasingly important in the subtropical hilly region. Suitable measures of N management need to be implemented in the catchment for achieving the goal of low carbon emissions and green development. The effects of these measures in both the short and long-term need to be urgently evaluated based on N cycle models developed for the subtropical hilly region.

## Acknowledgements

The work was supported by the National Natural Science Foundation of China (41771336, 41471267, 4211101081, 42161144002), Youth Innovation Promotion Association of the Chinese Academy of Sciences (Y2021102), Key Research and Development Program of Hunan Province (2020NK2011), and Chinese Academy of Science and Technology Service Network Initiative Project (KFJ-STS-QYZD-2021-22-002).

## Compliance with ethics guidelines

Jianlin Shen, Yong Li, Yi Wang, Yanyan Li, Xiao Zhu, Wenqian Jiang, Yuyuan Li, and Jinshui Wu 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.

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