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Long-term changes of nitrogen leaching and their contributions to lake eutrophication dynamics on the Yangtze Plain of China

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Abstract

Over the past half century, drastically increased chemical fertilizer have entered agricultural ecosystems to promote the crop production on the Yangtze Plain, potentially enhancing agricultural nutrient sources for eutrophication in freshwater ecosystems. However, long-term trends of nitrogen dynamics in terrestrial ecosystems and their impacts on eutrophication changes in this region remain poorly studied. Using a process-based ecosystem model, we investigated the temporal and spatial patterns of nitrogen use efficiency (NUE) and nitrogen leaching on the Yangtze Plain from 1979 to 2018. The agricultural NUE for the Yangtze Plain significantly decreased from 50 % in 1979 to 25% in 2018, with the largest decline of NUE in soybean, rice and rapeseed. Simultaneously, the leached nitrogen from cropland and natural land increased with annual rates of 4.5 kg N ha^{-1} yr^{-2} and 0.22 kg N ha^{-1} yr^{-2}, respectively, leading to an overall increase of nitrogen inputs to the fifty large lakes. We further examined the correlations between terrestrial nutrient sources (i.e., the leached nitrogen, total phosphorus sources, and industrial wastewater discharge) and the satellite-observed probability of eutrophication occurrence (PEO) at an annual scale, and showed that PEO was positively correlated with the changes in terrestrial nutrient sources for most lakes. Agricultural nitrogen and phosphorus sources were found to explain the PEO trends in lakes in the western and central part of Yangtze Plain, and industrial wastewater discharge

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was associated with the PEO trends in eastern lakes. Our results revealed the importance of terrestrial nutrient sources for long-term changes in eutrophic status over the fifty lakes of the Yangtze Plain. This calls for sustainable nitrogen applications in agriculture systems to improve water quality of local lake ecosystems.

1 Introduction

For the past half century, China's demand for grain production has increased from 250 Mt in 1960 to 648 Mt in 2010 along with the growing population, industrial development, and human-diet changes (Zhao et al., 2008; Wang and Davis, 1998). Substantial chemical fertilizers (i.e., 35 mega-tons, Mt, nitrogen fertilizers in 2014 (Yu et al., 2019) simultaneously entered agricultural ecosystems for the promotion of crop production. Although national grain production consequently increased from 132 Mt in 1950 to 607 Mt in 2014 (Yu et al., 2019), such a level of fertilization has enhanced nitrogen discharge to terrestrial and freshwater ecosystems, leading to a series of ecological and environmental concerns, such as soil nitrogen pollution, water quality deterioration, and phytoplankton blooms (Zhang et al., 2019; Wang et al., 2021; Qu and Fan, 2010). It was reported that approximately 14.5 Mt N yr^{-1} was discharged to surface water ecosystems over the entire China for the period of 2010-2014, which largely exceeded the national safe level of nitrogen discharge (i.e., 5.2 Mt N yr^{-1}) for the aquatic environment (Yu et al., 2019). Such human-related nutrient enrichment poses a big challenge for China's sustainable development goals (Wang et al., 2022).

The Yangtze Plain, with a human population of 340 million and agricultural area of 100 million hectares (Chen et al., 2020a; Hou et al., 2020), is experiencing unprecedented ecological and environmental issues (Guan et al., 2020; Feng et al., 2019). From 1990 to 2015, total crop production increased by 15 % at the expense of an increase of 89 % in nitrogen fertilizers over the Yangtze Plain (Xu et al., 2019). Consequently, more frequent nitrogen pollution was observed in soil and water. For example, heavy fertilizer usage and intensive livestock contributed to soil nitrogen pollution on the Yangtze River Delta for the past four decades, leading to soil deterioration and nitrogen discharge (Zhao et al., 2022). Nitrogen discharge related to human activities (i.e., fertilizer and manure applications, and human food waste) largely increased the nutrient loading and accelerated the degradation of water quality in the

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Yangtze River since the 1990s (Chen et al., 2020b). Under the recent sustainable development plans proposed by national and local governments, managing nitrogen sources from urban and crop systems is envisaged to mitigate severe soil and water pollutions (Chen et al., 2020b; Zhao et al., 2022; Shi et al., 2020). However, for the Yangtze Plain with a variety of crops and crop managements, the lack of insights into long-term changes of nitrogen dynamics, such as fertilizer application, plant nitrogen uptake, and nitrogen leaching, has limited our solution of proposing effective policies in related to nutrient management.

According to the national surveys and satellite observations, the lakes on the Yangtze Plain experienced eutrophication and algal blooms for the past two decades (Guan et al., 2020; Brunier et al., 2014). Cyanobacteria blooms were reported to frequently occur in Taihu and Chaohu lakes, with the peak expanded extent reported for 2006 (Qin et al., 2019). Since then, the magnitudes of algal blooms significantly decreased from 2006 to 2013, and slightly increased again from 2013 to 2018 (Huang et al., 2020). Satellite observations revealed widespread and serious eutrophication issues in large lakes of the Yangtze Plain for the periods of 2003-2011 and 2017-2018, although significantly decreasing trends were found in 20 out of 50 lakes throughout the periods (Guan et al., 2020). Various laws and guidelines were implemented on regional and national scales to control eutrophication problems, such as the Guidelines on Strengthening Water Environmental Protection for Critical Lakes in 2008 and the Water Pollution Control Action Plan in 2015 (Huang et al., 2019). Nevertheless, the eutrophication issues are still challenging to control and improve under the scarcity of effective strategies for the whole Yangtze Plain.

To understand the primary causes of eutrophication in the lakes of the Yangtze Plain, previous studies have attempted to determine the contributions of riverine nutrient exports and lacustrine nutrient loading to algal blooms in individual lakes, such as Taihu and Chaohu lakes (Tong et al., 2017; Tong et al., 2021; Xu et al., 2015). Wang et al. (2019b) identified that diffuse sources contributed 90% to riverine exports of total dissolved nitrogen, and point sources discharged 52% of riverine phosphorus exports to Taihu Lake. Based on field-measured phytoplankton biomass and nutrient concentrations, algal blooms in Taihu Lake were primarily attributed to excessive nutrient loads from 1993 to 2015 (Zhang et al.,

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2018). Overloaded nutrients, in combination with climatic warming, were found to regulate the seasonal variations of cyanobacteria blooms in Chaohu Lake based on the monthly nutrient monitoring at discrete points (Tong et al., 2021). However, these studies only tracked the primary drivers of algal blooms for individual hyper-eutrophic lakes (i.e., Taihu and Chaohu lakes), which is insufficient to understand the causes of regional eutrophication and support the design of regional management strategies. Additionally, the studies above investigate cyanobacteria dynamics in relation to regional fertilizer use, but do not quantify changes in nitrogen uptake by vegetation.

In this study, we employed a process-based dynamic vegetation model, LPJ-GUESS (Smith et al., 2014), to investigate terrestrial nitrogen dynamics for the past four decades, examining the primary drivers of eutrophication trends in fifty large lakes of the Yangtze Plain (covering 63% of the whole plain). We simulated the vegetation dynamics, nitrogen cycles for agricultural and natural ecosystems from 1979 to 2018, and then assessed the temporal trends of nitrogen use efficiency and nitrogen leaching. The terrestrial nutrient sources were used to examine their linkage with the satellite-derived eutrophication changes for fifty large lakes.

2 Materials and Methods

2.1 Study area

The Yangtze Plain (Fig. 1) is in the middle and lower basin of the Yangtze River. It covers a total area of 7.8 $\times 10^6$ km^2 from Hunan Province to Shanghai City, and accommodates approximately 5000 freshwater lakes, ponds and reservoirs (Hou et al., 2017). Its sub-tropical monsoon climate provides annual mean temperature (~15℃) and precipitation (~1000 mm) conditions favorable for crop cultivations, in particular cereals and oil seeds, making the Yangtze Plain one of the top three food production regions in China. However, since the policy of Reform and Opening-up of China in the 1980s (Zhang et al., 2010), agricultural ecosystems have been confronted with great pressure from urban expansion on the Yangtze Plain. Rapid urban expansion encroached on arable land, mainly on the eastern parts of the Yangtze Plain (Zhang et al., 2021).

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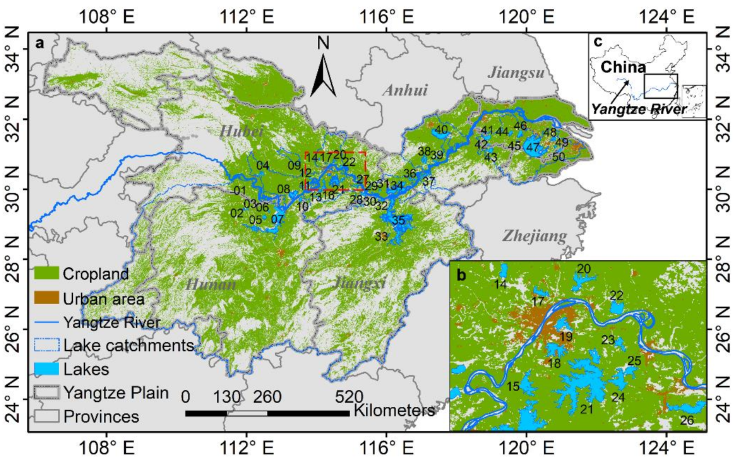

Figure 1. Locations of the Yangtze Plain and the fifty large lakes studied here. (**b**) Detailed overview of Wuhan region (red box in (**a**)) and the surrounding lakes.

2.2 Dynamic vegetation model

We used a dynamic ecosystem model, LPJ-GUESS (Smith et al., 2014; Olin et al., 2015b), to simulate vegetation dynamics (i.e., the establishment, growth, competition, and mortality of plants), soil biogeochemistry, and carbon and nitrogen cycles for different ecosystems on the Yangtze Plain. The model has been widely used to assess ecosystem carbon and nitrogen fluxes at regional and global scales (Smith et al., 2014). Plant functional types (PFTs) and crop functional types (CFTs) are designed to describe the different types of plants and crops with a set of pre-defined bioclimatic and physiological parameters, such as photosynthetic pathways, phenology, growth forms and life history strategies for PFTs, as well as irrigation, fertilization, and rotation schemes for CFTs (Smith et al., 2014; Sitch et al., 2003; Olin et al., 2015a; Lindeskog et al., 2013).

Carbon and nitrogen fluxes between ecosystems and the atmosphere are calculated on a daily basis. For natural PFTs, net primary production (NPP) is accumulated and allocated to different plant compartments (i.e., leaves, roots, and sapwood and heartwood for trees) at the end of each simulation year. Soils are represented by 11 carbon and nitrogen pools with different decomposition rates (Parton et al., 1993; Parton et al., 2010), dependent on soil temperature and texture, water content, and base decay rates (Smith et al., 2014). Atmospheric deposition and plant biological fixation provide nitrogen

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sources for plant growth and development, while the decomposition of soil organic matter can release mineral N into the soils and nitrogen-related gases into the atmosphere. Moreover, soil soluble nitrogen can also leach with the surface runoff in the forms of dissolved organic and inorganic nitrogen (i.e., DON and DIN). In the model, leaching of DON is a function of the decay rates of soil microbial carbon pool and soil percolation, while DIN leaching depends on the available mineral nitrogen in soils and soil percolation, as well as soil water content.

Crop growth starts from a seedling with initial carbon and nitrogen masses at a prescribed sowing date. Chemical fertilizer and livestock manure supply external nitrogen for crop growth. Crop N uptake is simulated as the lesser between crop N demand and accessible mineral N in soils, where the former depends on crop development stages and C:N ratios of leaves and roots, and the latter is affected by soil temperature and fine root biomass (Olin et al., 2015a). Differing from natural PFTs, NPP is allocated to leaves and stems, root, and storage organs for each CFT on a daily basis, according to the daily allocation strategies related to crop development stages (Olin et al., 2015a).

2.3 LPJ-GUESS input, calibration, and evaluation dataset

2.3.1 Input data

We ran LPJ-GUESS separating four land use types (natural land, cropland, pasture and urban) with a 500-year spin-up to simulate the vegetation dynamics and the associated nitrogen fluxes for the Yangtze Plain from 1979 to 2018.

The gridded input data for LPJ-GUESS include climate, fractions of four land use types, total chemical fertilizer and manure application rates, cover fractions of each CFT within the cropland area, and soil properties. We used daily temperature, precipitation, and shortwave radiation provided by the China Meteorological Forcing Dataset (CMFD), with a spatial resolution of 0.1° and a temporal coverage of 1979-2018 (He et al., 2020). The 300-m Climate Change Initiative Land Cover (CCI-LC version 2.0) dataset was regrouped into four different land use types (i.e., urban, cropland, pasture, and natural land) to obtain the cover fractions within each 0.1° grid cell for the period of 1992 to 2018 (Defourny et al., 2012) (see the details about regrouping process in Supplementary S1). Soil properties, i.e., fractions of sand, clay and silt, organic carbon content, C:N, pH, and bulk density were extracted from the World Inventory of Soil Property Estimates (WISE30sec) dataset (Batjes, 2016). Based on the original data with a spatial resolution of 30 sec, we determined the dominant FAO soil type based on their relative area in each grid cell, and used its properties as input data for the grid cell. Gridded chemical fertilizer and manure application data were extracted from global fertilizer usage (Lu and Tian, 2017) and manure data (Zhang et al., 2017), which have spatial resolutions of 0.5° and 0.5', respectively. We resampled the fertilizer and manure application data into the spatial resolution of 0.1° to represent the chemical fertilizer and manure application for each grid cell from 1979 to 2014.

The gridded fractions of CFTs were calculated based on observational data provided by the China Meteorological Data Service Center (https://data.cma.cn/site/subjectDetail/id/101.html). The dataset contains the information about the types, sowing and harvest dates for totally eleven crops at 92 observational sites across the whole Yangtze Plain (listed in Table S1). An adaptive inverse distance weighting method was then used to interpolate the maps of the relative fractions of all crops, and their sowing and harvest dates for the period of 1992-2015 (see the details in Supplementary S2). Due to the limited availability for the period of 1979-1991 and 2016-2018, we used the same crop information (i.e., the fractions of crop types, sowing and harvest dates) from the nearest years.

2.3.2 Model calibration and evaluation data

The model was calibrated based on the observed crop yield collected by the China Meteorological Data Service Center (https://data.cma.cn/site/showSubject/id/102.html). The dataset provides crop yield data for eight main crops collected at different numbers of sites (i.e., winter wheat (number of sites: 37), spring maize (6), summer maize (10), single-season rice (28), early-season rice (30), late-season rice (30), rapeseed (38), and soybean (15)), for the period of 2000-2013. For the Yangtze Plain, hybrid and super-hybrid rice are widely cultivated to obtain high grain yield within short growing seasons due to the enhanced photosynthetic rates associated with leaf-level chlorophyll and rubisco contents (Huang et al., 2016). However, the default parameters for rice CFTs in LPJ-GUESS cannot capture the high yield features of hybrid and super-hybrid rice. Therefore, we calibrated the relationship between the leaf-based nitrogen content and the maximum catalytic capacity of rubisco (see the details in Supplementary S3). We randomly selected five sites with rice yield data from 2000 to 2013 as the calibration data, and the other rice yield data were used as the evaluation data. For parameters of other CFTs (listed in Table S1), the default values performed satisfactory in the comparison with all observed yield data (Fig. 2). It is noted that regional mean yield for each crop was derived from the evaluation data to compare the simulated values on the Yangtze Plain.

Simulated GPP and LAI were further compared with Global Solar-induced Chlorophyll Fluorescence Gross Primary Productivity (GOSIF GPP) and third generation of Global Inventory Modeling and Mapping Studies Leaf Area Index (GIMMS LAI3g) products to evaluate the performance of modelled vegetation variables. The global GOSIF GPP products have a spatial resolution of 0.05° and cover the period of 1992-2018 (Fang et al., 2019). Biweekly GIMMS LAI3g products with a spatial resolution of 0.25° were obtained and then converted to annual mean LAI3g maps from 1982 to 2011 (Zhu et al., 2013).

The modelled responses of nitrogen leaching to different fertilizer applications were evaluated based on an observational dataset published in Gao et al. (2016), where they collected nitrogen leaching for plots with 3 or 4 different levels of nitrogen fertilizer inputs for maize, rice, and wheat. In our study, we selected the observed responses without influences of phosphorus and potash fertilizers on the Yangtze Plain as the evaluation data (two samples for each crop). For these sites, individual simulations were performed by assigning the full coverage of each corresponding crop grow, and prescribing the levels of nitrogen fertilizer applications as in the experimental site. It should be noted that we used the same nitrogen fertilizer applications in the period prior to the field experiment.

2.4 Assessment of long-term changes in nitrogen dynamics

We assessed long-term changes of nitrogen use efficiency (NUE) and nitrogen leaching for the past four decades. A linear regression was conducted for the annual mean NUE and nitrogen leaching for the whole Yangtze Plain to determine the associated change rates (i.e., the regression slopes), and the significance was tested by a *t*-test. The mean leached nitrogen over the drainage area of all examined

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lakes was calculated to explore long-term changes in terrestrial nitrogen sources for lake ecosystems, and the associated temporal trends were assessed by the linear regression and *t*-test.

2.5 Examination of the primary driving forces of eutrophication dynamics

2.5.1 Satellite-derived eutrophication changes

We used satellite-derived PEO data published in Guan et al. (2020) to represent the eutrophication changes for fifty large lakes on the Yangtze Plain. The PEO was defined as the frequency of high chlorophyll-a concentrations (i.e., > 10 mg m^{-3}) or algal bloom occurrences in satellite imagery for each year. The averaged PEO values for pixels within each lake were then obtained to delineate the eutrophication status and changes in fifty large lakes of the Yangtze Plain during the MERIS (i.e., 2003-2011) and OLCI (i.e., 2017-2018) observational periods. However, due to un-availability of crop- and nitrogen-related data for the period of 2017-2018, we only used the PEO data derived from MERIS observations (i.e., 2003-2011) here to examine their primary driving forces.

2.5.2 Examination of the correlations between nutrient and PEO anomalies

To examine the impacts of terrestrial nutrient sources on eutrophication changes in fifty large lakes of the Yangtze Plain, we used the simulated nitrogen leaching (LN) and anthropogenic phosphorus sources (i.e., total phosphorus from chemical fertilizer and manure, TP) representing the agricultural nutrient sources, and industrial wastewater discharge (IW) representing industrial nutrient sources. The gridded phosphorus fertilizer data were extracted from a global dataset developed by Lu and Tian (2017), while the phosphorus content in manure was calculated based on the nitrogen contents of manure products and the associated N:P ratios of different animals' excrement (Table S3). Annual industrial wastewater discharge data were obtained from the China City Statistical Yearbook (https://data.cnki.net/trade/Year-book/Single/N2018050234?zcode=Z011).

The 9-year's mean (2003-2011) of three nutrient-related variables (i.e., LN, TP and IW) were used in a principal component analysis (PCA) followed by a K-means clustering (Hartigan and Wong, 1979) to classify examined fifty lakes based on similarities of terrestrial nutrient sources. In this process, all variables were normalized (across all years and lakes) based on the z-score method to remove the

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influence of different magnitudes in nutrient-related variables. We derived the first two principal components (PCs) from all normalized variables through a PCA, and the lakes were classified into three classes based on the first two PCs through the clustering methods. Finally, the annual anomalies of these nutrient-related variables and PEOs relative to their 9-year means were used to determine the primary drivers of temporal trends in eutrophication for each lake class.

3 Results

3.1 Evaluation of LPJ-GUESS simulation

For the evaluation of LPJ-GUESS simulation for the past four decades, the simulated LAI, GPP and crop yield were compared with observation-based estimates. Mean crop yields agreed well with the observed values, with mean relative errors of < 10% (Fig. 2). The comparison of simulated and observed LAI, and GPP were also satisfactory with overall high accuracy (i.e., a mean relative error of ~20% and the root squared relative errors of < 30%) and spatial distributions consistent with observed patterns (Fig. S1 and S2). Considering the difference in spatial scales between the grid cells and the gridded evaluation data (i.e., the observed LAI and GPP maps), the overall performance of vegetation simulation over the different land use types were considered acceptable. In addition, the simulated responses of nitrogen leaching to different fertilizer applications at the experimental sites showed overall similar trends as the observation ones for all three crops (i.e., maize, rice, and wheat), despite varying magnitudes of differences between the simulated and observed leached nitrogen at certain fertilizer level (Fig. 3).

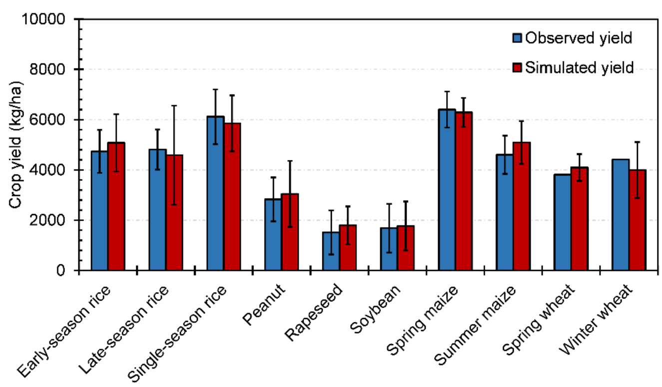

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Figure 2. Comparison between the simulated and observed crop mean yields of different crops on the Yangtze Plain; the mean values were averaged over the period 2000-2015 and across totally 179 sites. Error bars show one standard deviation of crop yield.

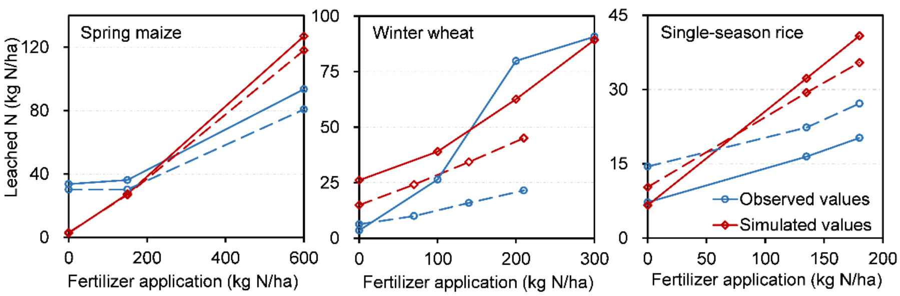

Figure 3. The simulated and observed responses of the nitrogen leaching to different levels of fertilizer application rates for three main crop types (i.e., maize, rice, and wheat) over the Yangtze Plain.

3.2 Long-term changes of nitrogen use efficiency over the Yangtze Plain

The climatological NUE was calculated to examine the spatial patterns of plant nitrogen uptake on the Yangtze Plain from 1979 to 2018. Considerable variations were detected across the entire Yangtze Plain, with NUE values ranging from 5% to 60% (Fig. 4a). Two hotspots of high NUE were in the Hubei and Jiangsu Province (see locations in Fig.1), dominated by cultivations of single-season rice and winter wheat under the moderate levels (i.e., ~200 kg N ha^{-1} yr^{-1}) of fertilizer applications (Fig. S3). The NUE values also differed among different crop types for the past four decades. The largest NUE values were found for soybean (74.0 % ± 11.0 %, Fig 5), while the lowest values were found for late-season rice (15.9% ± 4.3%).

Due to the unprecedented increase of chemical fertilizer application since the 1980s, the crop NUE on the Yangtze Plain has significantly decreased from ca. 50% in 1979 to 25% in 2018 (p < 0.05, in Fig. 4b), with an overall annual change rate of -0.55 % yr^{-1}. Overall, regions with relatively high levels of NUE depicted a moderate or even slight increase for the past four decades, while the regions dominated low-level NUE (i.e., Hubei and Hunan provinces in Fig. 1) experienced strongly declining trends (Fig.

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4a&4c), as a result of the enhanced fertilizer applications. Considerable differences in magnitudes and trends of NUE were also examined among the crop types. Significant decreases (t-test, $p < 0.05$) in the decadal NUEs were found for seven crop types (annotated with "*" in Fig. 5), with the largest decrease for the double cropping of early- and late-season rice (Fig. 4c and S3). In contrast, three crop types experienced increasing trends of NUE, including peanut, spring wheat, and sugarcane (Fig. 5).

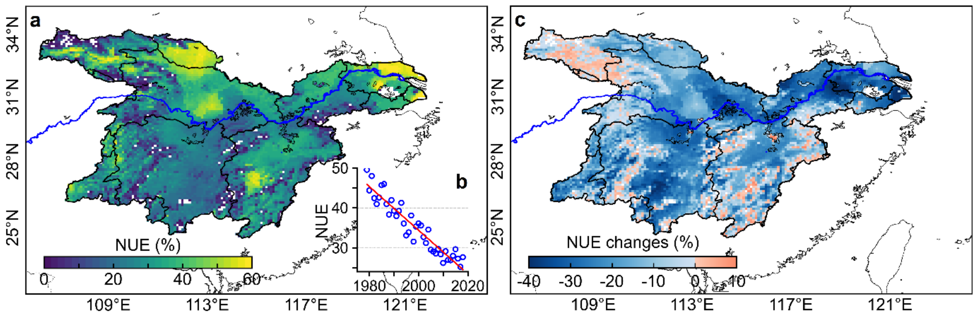

Figure 4. Nitrogen use efficiency (NUE) on the Yangtze Plain from 1979 to 2018. (**a**) Spatial distributions of climatological NUE (1979-2018), and the inset (**b**) shows the long-term trends of area mean NUE; (**c**) Changes in NUE between the first (1979-1988) and the last (2009-2018) decades.

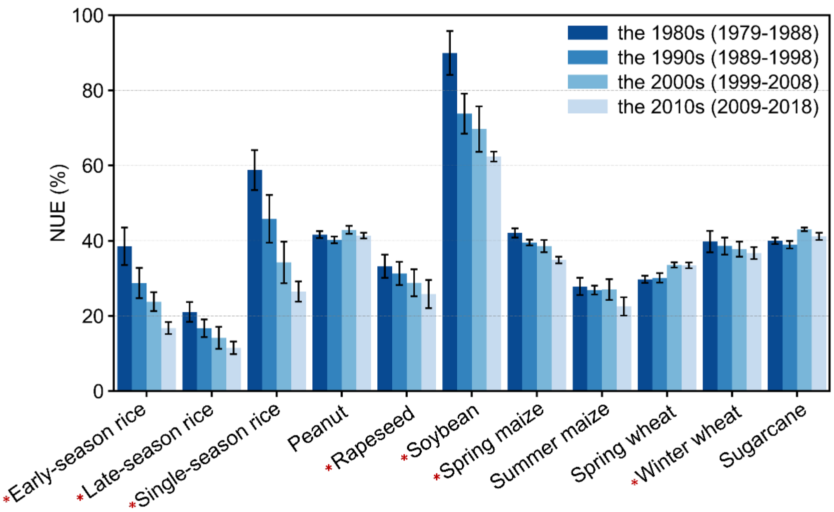

Figure 5. Decadal values of NUE for each crop functional type, averaged over the Yangtze Plain for the past four decades. Significantly decreasing trends ($p < 0.05$) are annotated with * using a t-test.

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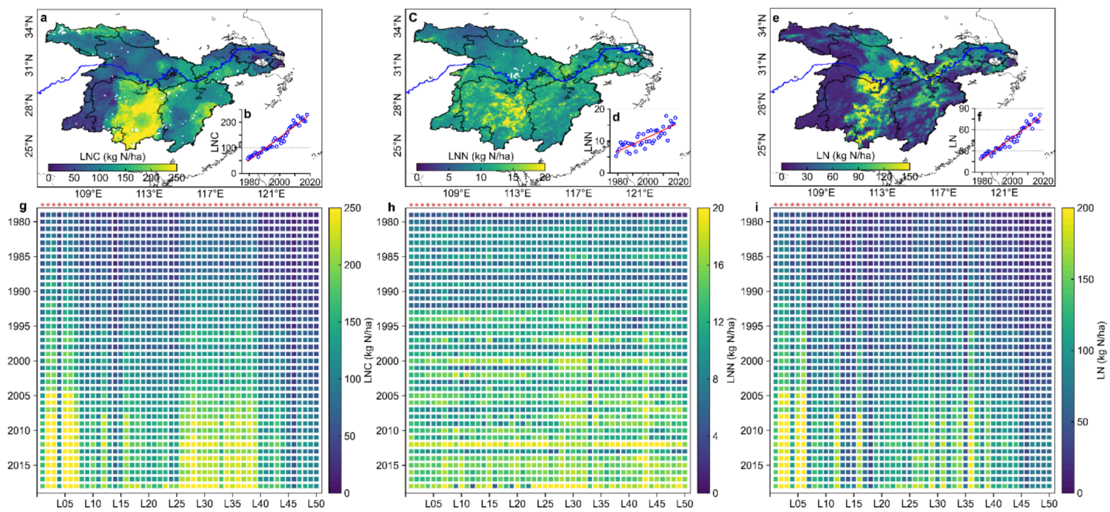

Figure 6. Tempo-spatial patterns of leached nitrogen from cropland (LNC), natural land (LNN), and total leached nitrogen (LN) for the period of 1979-2018. Spatial distributions of climatological (**a**) LNC, (**c**) LNN, and (**e**) LN. The insets (**b**), (**d**) and (**f**) represent the long-term changes of mean LNC, LNN and LN, where the red lines are the linear fitting lines between years and nitrogen leaching. Inter-annual changes of (**g**) LNC, (**h**) LNN, and (**i**) LN for all examined lakes (L01-L50) from 1979 to 2018. Statistically significantly positive trends ($p < 0.05$) are annotated with '*' on top of the panel, and see Fig. 1 for ID numbers of lakes.

3.3 Temporal and spatial patterns of nitrogen leaching for the past four decades

Along with the overall decreases in NUE, the leached nitrogen from agricultural (LNC, averaged across cropland area) and natural systems (LNN, averaged across natural area) experienced statistically significant increase ($p < 0.05$) over the past four decades, with the different rates (4.5 kg N $ha^{-1}yr^{-2}$ and 0.22 kg N/ha /yr^2, respectively in Fig. 6b, 6d). The LNC were an order of magnitude larger than the LNN. The high levels of LNC were found mainly in the Hunan Province (see Fig. 1 and Fig. 6a), with the average LNC value of 149 kg N ha^{-1} yr^{-1}. In contrast, considerable spatial variations in LNN were revealed between the north and south parts of the Yangtze Plain (Fig. 6c).

To understand nitrogen sources for each corresponding lake ecosystems on the Yangtze Plain, we calculated the mean leached nitrogen (LN, averaged across ground area) over the drainage area of each

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studied lake. The LN values ranged from 29 kg N ha^{-1} yr^{-1} in Gehu Lake (L46 in Fig. 6i) to 153 kg N ha^{-1} yr^{-1} in Donghu Lake (L05 in Fig. 6i), indicating the considerable difference between the western lakes in the Hunan Province and the eastern lakes in the Jiangsu Province. All examined lakes experienced statistically significantly increasing trends in the LN (t-test, $p < 0.05$) over the past four decades (Fig. 6i), where the agricultural activities contributed 94 % ± 5 % to the LN changes.

3.4 Driving forces of terrestrial nutrient sources to eutrophication changes

The leached nitrogen, total phosphorus sources, and industrial wastewater discharge were used to represent terrestrial nutrient sources and were further investigated in terms of their linages to the observed PEOs. In the PCA analysis, the first two principal components (PCs) explained 48.7% and 33.6% of variations in terrestrial nutrient sources (Fig. 7), where the first PC primarily depicts positive dependence on IW but negative links with LN, and the second PC reveals negative dependences on TP and IW. All fifty lakes were clustered into three classes based on the first two PCs (Fig. 7). Lakes in class I (n = 22) had the positive loading in the direction of the total phosphorus sources, with the main coverage of the middle Yangtze Plain (i.e., Jiangxi and Anhui Province in Fig. 1), while class II cover the most of lakes (n = 17) in the western regions (i.e., the Hunan Province and the western parts of the Hubei Province in Fig. 1). The lakes of class III (n = 11) are primarily located on the eastern Yangtze Plain, except for two lakes (i.e., Donghu and Tangxun lakes) which locate at the urban area of Wuhan City.

The correlations between annual anomalies of PEO and the three nutrient variables (relative to their means for 2003-2011) were examined for all three lake classes. The PEO anomalies were significantly correlated with different nutrient variables for three lake classes, indicating spatial variations of driving factors for eutrophication changes on the Yangtze Plain (Fig. 8). Specifically, both LN and TP anomalies exhibited significantly positive correlations ($p < 0.001$) with the PEO trends in lakes of class I and II (Fig. 8a&b), indicating that the primary influence of agriculture-related sources to the increasing trends of PEO. In contrast, the annual PEO dynamics in lakes of class III showed a significantly positive correlation ($p < 0.05$) with industrial wastewater discharge (Fig. 7c), meaning that the temporal trends of annual PEO in eastern parts of the Yangtze Plain were mainly associated with industrial wastewater

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discharge. Note that the significantly negative correlations between the PEO and IW anomalies were found for class I and II (Fig. 8c), which might be mechanistically unlikely. In such case, more efforts are required in the future to determine the influence of industrial nutrient sources to eutrophication changes in these lakes.

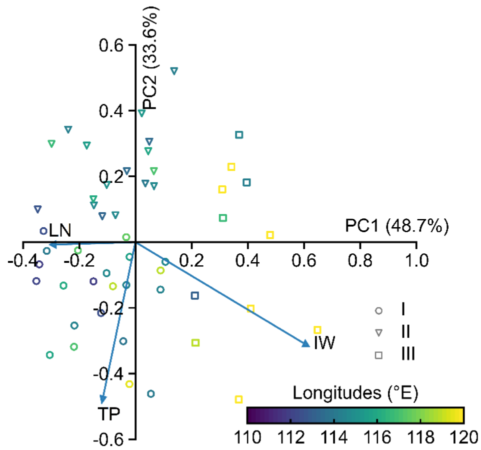

Figure 7. Loading plot of the principal component analysis (PCA) based on three nutrient-related variables. The color of scattering points represent the distributions of lakes in longitudinal order, and the directions of nutrient-related variables (i.e., LN leached nitrogen; TP total phosphorus sources; IW industrial wastewater) were annotated with blue arrows.

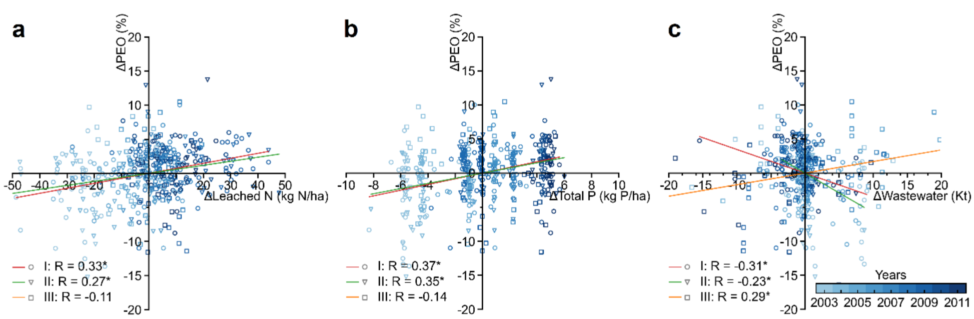

Figure 8. Relationships between the annual anomalies of PEO and nitrogen leaching **(a)**, total phosphorus sources **(b)**, and industrial wastewater discharge **(c)** for fifty studied lakes. The color and symbol of scattering points represent the years and lake classes, and the colored lines (shown for

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significant correlations only) are linear regression between the annual anomalies of PEO and nutrient-related variables for each lake classes.

4 Discussions

4.1 Nitrogen use efficiency and driving forces of eutrophication changes.

The overall low mean NUE (27 %) and declining trends in NUE (-0.55 % yr^{-1}) characterized agricultural ecosystems on the Yangtze Plain for the past four decades. Such decreasing trends are consistent with the previous studies using statistical datasets and numerical modelling (Zhang et al., 2015; Yu et al., 2019). However, the generally low NUE constitutes a vast gap with the targeted sustainable NUE of 60% in China (Zhang et al., 2015), possibly leading to the excessive nitrogen discharge for terrestrial and aquatic ecosystems. The correlation analysis revealed that PEO trends in the western and central parts of the Yangtze Plain were associated with agricultural nutrient sources. As such, improving the NUE can decrease the agricultural nutrient exports to mitigate eutrophication issues in lake ecosystems, especially under the growing food demands in the future.

In the eastern parts of the Yangtze Plain, the correlation between industrial wastewater and PEO indicates that industrial sewage possibly delivered terrestrial nutrients to lakes from the adjacent cities, providing sufficient nutrient conditions for phytoplankton communities (Luan et al., 2007). The Jiangsu Province in the eastern parts of the Yangtze Plain (see the locations in Fig. 1) experienced rapid economic and industrial development since the policy of Reform and Opening-up of China since 1980s (Shen et al., 2020), which might be expected to result in enhanced industrial wastewater production, thereby triggering serious eutrophication in the lake systems. However, the national strategies and policies about green growth of industries encouraged reclamation of wastewater, investment on the advances in wastewater treatment technology and installment of municipal wastewater treatment plants (Li et al., 2013; Lyu et al., 2016). Industrial structures were also required to transform from secondary to tertiary industries under the environment-friendly targets of economic development (Huang et al., 2015). Consequently, industrial sewage showed decreasing trends and attributed to improvement of eutrophication status in the eastern Yangtze lakes.

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4.2 Limitations and Uncertainties

Using the LPJ-GUESS model, we investigated the long-term changes and spatial variations of nitrogen dynamics (i.e., plant nitrogen uptake and nitrogen leaching) over the Yangtze Plain for the past four decades, and then examined the contributions of terrestrial nutrient sources to eutrophication changes in fifty large lakes. However, due to the lacking representation of a phosphorus cycle in the LPJ-GUESS model, we used external phosphorus fertilizer and manure application rates to represent the agricultural phosphorus sources, without consideration of potential impacts from plants and soil processes. Applied phosphorus is taken up through crop roots , or can be retained in soils (Schoumans, 2015) in variable degrees, affecting the export to lake ecosystems, which makes that the use of application data generates an uncertainty in our analysis. Nevertheless, we consider it important to consider the phosphorus sources as potential driving force for eutrophication changes under the low levels of phosphorus use efficiency over the Yangtze Plain (Li et al., 2017; Zheng et al., 2018).

Another source of uncertainty is associated with the potential impacts of terrestrial nutrient losses during the transport processes to surface water ecosystems, as well as the impacts of aquaculture-related nutrient sources. Transport processes depend on soil properties, topography, and hydrological conditions over the drainage area (Solomon et al., 2015; Tang et al., 2014; Tang et al., 2018), which is required to further consider at regional scales to estimate the dynamics of terrestrial nutrient exports for lake ecosystems on the Yangtze Plain. In addition, intensive and widespread freshwater aquaculture across the Yangtze Plain can contribute to accessible nutrient sources for the eutrophication development and phytoplankton growth (Guo and Li, 2003; Wang et al., 2019a). Satellite observations revealed that 17 out of 50 lakes on the Yangtze Plain have established enclosure fishery nets to increase the fish production (Dai et al., 2019). Consequently, substantial nutrients in fish food can directly enter aquaculture zones, promoting the contents of nitrogen and phosphorus in these lakes. These associated drivers are required to be comprehensively assessed to draw a complete picture of accessible nutrient sources for phytoplankton communities and then specify the anthropogenic impacts on water quality and eutrophication deterioration on the Yangtze Plain.

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Uncertainties in the PEO data can originate from the uneven distributions of valid numbers of satellite observations across the fifty large lakes of the Yangtze Plain. Under the influence of observational conditions (i.e., cloud coverage and thick aerosols), the imagery with high-quality observations distributed unevenly across the different years and seasons, which potentially resulted in certain impacts on the derived annual PEOs and their temporal trends. Alternatively, the annual PEOs were calculated based on the quarterly values to minimize such uncertainties. Nevertheless, more frequent satellite observations will still be required to obtain a more accurate assessment of eutrophication changes in lake ecosystems.

5 Conclusions

We used the LPJ-GUESS model to investigate the long-term changes of nitrogen dynamics over the Yangtze Plain for the past four decades, and then examined their potential functions as the driving forces of eutrophication changes in fifty large lakes of the Yangtze Plain. Significant decreases in NUE dominated the whole Yangtze Plain, with the largest decrease in rice, soybean and rapeseed. The leached nitrogen from both cropland and natural land showed statistically significant increasing trends for all fifty examined lakes, indicating increased availability of terrestrial nitrogen sources in lake systems for the past four decades. Two classes of lakes located in the western and central parts of the Yangtze Plain showed significantly positive correlations between anomalies of PEO and agricultural nutrient sources (i.e., the leached nitrogen and total phosphorus sources), and the PEO anomalies in the remaining class (11 eastern lakes) were positively correlated with the industrial wastewater discharge. The impacts of agricultural and industrial nutrient sources to eutrophication changes further emphasize the importance of sustainable management of terrestrial nitrogen and phosphorus to improve water environments.

Data availability. Data used in this study are archived by the authors and are available upon request.

Author contributions. QG, JT, LF and GS designed the framework and methodology of the study. QG drafted a first version of the manuscript and analyzed the results. QG, JT and GS performed the

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calibration of LPJ-GUESS model. All co-authors contributed critically to the manuscript editing and writing processes.

Competing interests. The authors declare that they have no conflict of interest.

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