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Trends in nutrients in the Changjiang River

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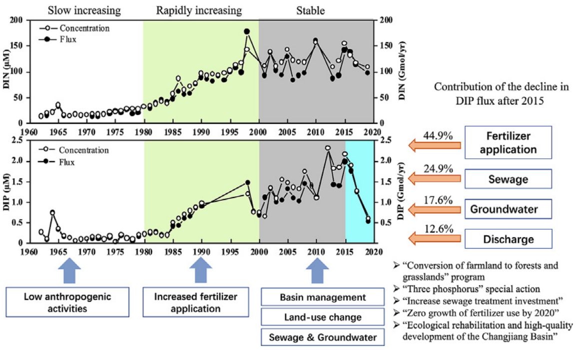
HIGHLIGHTS

- The nitrate flux has increased, while the ammonium flux has decreased.
- Phosphate has decreased in recent 5 years.
- The phosphate decline is due to pollution control and fertilizer reduction.
- Enhanced phosphorus limitations could shift phytoplankton community.

GRAPHICAL ABSTRACT



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ABSTRACT

Better documentation and understanding of long-term temporal dynamics of nutrients in watersheds are necessary to support effective water quality management. We examined the hypothesis that the recent management of fertilizer use and pollution control in the Changjiang River Basin could govern the fluxes of nutrients from the river to the sea. Results based on historical data since 1962 and surveys in recent years show that concentrations of dissolved inorganic nitrogen (DIN) and phosphorus (DIP) in the mid- and downstream reaches were higher than those in the upper reaches due to intensive anthropogenic activities, while dissolved silicate (DSi) was distributed evenly from the up- to downstream reaches. Fluxes of DIN and DIP increased rapidly, and DSi declined during the 1962–1980 and 1980–2000. After the 2000s, concentrations and fluxes of DIN and DSi remained almost unchanged; those of DIP remained stable until the 2010s and slightly decreased afterward. The decline in fertilizer use explains 45 % of the variance in the decline of DIP flux, followed by pollution control, groundwater and water discharge. As a result, the molar ratio of DIN:DIP, DSi:DIP and ammonia:nitrate varied largely during 1962–2020, and the excess DIN relative to DIP and DSi lead to increased limitations of silicon and phosphorus. A turning point probably occurred for nutrient fluxes in the Changjiang River in the 2010s, with the pattern of DIN from continuous increase to stability and DIP from increase to decrease. This decline in phosphorus in the Changjiang River has many similarities with the rivers worldwide. The continued basin nutrient management is likely to have a major influence on river nutrient delivery and therefore may control coastal nutrient budget and ecosystem stability.

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1. Introduction

River transport large amounts of nutrients from land to sea and significantly influence biogeochemical cycle in coastal ecosystems (Malone and Newton, 2020; Xiao et al., 2019; Zhang et al., 2021a, 2021b). Global riverine fluxes of nitrogen (N) and phosphorus (P) have more than doubled between the beginning of the 20th century and 2010 (Beusen et al., 2016). This is largely the consequence of human activities in the watershed, such as the use of fertilizers and detergents (Mayorga et al., 2010). Excessive nutrients (N and P) in aquatic environments may cause eutrophication in surface water (Paczkowska et al., 2020; Wang et al., 2021a; Xu et al., 2010) and nutrient enrichment in groundwater, thereby resulting in the subsequent degradation of estuarine and coastal ecosystems (Liu et al., 2019; Rabalais et al., 2009; Gao and Wang, 2008). While riverine N and P loads have increased during the past decades, dissolved silicate (DSi, an important nutrient form) loads have remained constant or even decreased in many rivers primarily as a result of DSi retention in reservoirs and lakes (Maavara et al., 2020b).

Nutrient retention in reservoirs also influences riverine N and P nutrients transport (Maavara et al., 2020b; Wu et al., 2022). Retention in reservoirs can decrease nutrient fluxes downstream (Wang et al., 2020; Yan et al., 2021) and alter nutrient compositions due to the different nutrient retention efficiencies, generally following the order of P > silicon (Si) > N (Chen et al., 2020a; Maavara et al., 2020a; Maavara et al., 2020b). These unbalanced changes in nutrients due to damming could enhance the P and Si limitations in rivers (Liu et al., 2018; Ran et al., 2019; Wang et al., 2020). To date, >50,000 dams have been built in the Changjiang River Basin (Tian et al., 2022). The Three Gorges Dam (TGD), Xiluodu Dam and Xiangjiaba Dam were fully operated in 2009, 2014 and 2015, respectively, and this trend of damming will continue owing to the increasing construction of cascade dams in the Changjiang River Basin (Yang et al., 2018), thereby resulting in further change of riverine nutrient transport. High N/P ratios of river water may increase the frequency, extent and toxicity of algal blooms in the estuary (Romero et al., 2013; Glibert and Burford, 2017; Yang et al., 2023).

Agricultural activity and urban sewage appear to be major factors affecting the nutrient flux in the river (Chen et al., 2019; Chen et al., 2020b; Guo et al., 2021; Wang et al., 2019c). For example, there has been an enormous increase in N and P fluxes since the 1970s due to increased fertilizer use and sewage inputs in the midstream and downstream subbasins of the Changjiang River (Liu et al., 2018; Liu et al., 2022a, 2022b). The increases in riverine N and P concentrations and loads in Europe and the United States before the 1980s were of great concern (Howarth et al., 2002; Loos et al., 2009; Reder et al., 2013). High levels of N in groundwater systems due to the agricultural activities have also become a major environmental concern all over the world in recent decades (Wakida and Lerner, 2005; Gu et al., 2013; Xin et al., 2019). After the implementation of water pollution control policies in Europe and the United States since 1980s, water quality (N and P concentration) in many rivers had been improved (Bouraoui and Grizzetti, 2011).

In recent years, China has also implemented many policies for environmental conservation and restoration. In the Changjiang River Basin, the cultivated area decreased by 7.5 % and forested area increased by 2.1 % from 2000 to 2015 (Kong et al., 2018) as a result of the initiation of the 'Natural Forest Conservation Program' and the 'Grain to Green Program' policy implemented across China in 1999 to restore degraded ecosystems, stabilize soils, and minimize erosion (Feng et al., 2016). In 2015, China's Ministry of Agriculture issued the action aiming for zero growth in synthetic fertilizer use from 2020 onwards (Ministry of Agriculture of the People's Republic of China, 2015). Recently, the government launched a 'three phosphorus' special action in seven provinces in the Changjiang River Basin (Ministry of Ecology and Environment of the People's Republic of China, 2019) to address the problem of excess total phosphorus in some sections of the Changjiang River due to industrial discharges. The continuous improvement of agricultural cultivation technology has also lowered the demand for chemical fertilizers. In addition, in recent years, several pollution controls have been implemented in the Changjiang River, e.g., 'Increase sewage treatment investment' (National People's Congress of the People's Republic of China, 2011) and 'Ecological rehabilitation and high-quality development of the Changjiang Basin' (National People's Congress of the People's Republic of China, 2016). Those management policy and measures in the recent years would have a major impact on land–sea nutrient fluxes. Currently, there are many studies that use historical river nutrient data to observe long-term river trends (Metson et al., 2020; Stackpoole et al., 2021). However, most studies investigating the effects of human activities on nutrient flux in the Changjiang River Basin use data before the 2010s and there is still a lack of knowledge of nutrients in groundwater (Xu et al., 2013; Strokal et al., 2017; Wang et al., 2019a–c). Thus, the changes in nutrients in the Changjiang River have remained unclear and controversial in recent years. More importantly, the latest observational data and river nutrient trends need to be supplemented in order to help governments better develop river nutrient management policies. Based on cruises conducted at different stations in the Changjiang River Basin and on multiple temporal scales during 2013–2020 and historical observation data during 1962–2016, this study investigated the temporal and spatial changes in the concentrations, fluxes and compositions of nutrients in the mainstream and tributaries of the Changjiang River on a long-term scale over the past six decades, on a monthly scale within a year, and on a daily scale during the flooding season. The influences of human activities on riverine nutrient transport in the Changjiang River were explored.

2. Materials and methods

2.1. Study area

The Changjiang River originates from the eastern Tibetan Plateau and has a length of approximately 6400 km and a catchment area of 1.8×10^6 km^2. The Changjiang River consists of three sections (upstream, midstream and downstream, divided by the Yichang and Hukou stations, Fig. 1) and covers the typical three steps of China's terrain from highlands in the upper valley to the lowland delta plain downstream. Two large lakes, Dongting and Poyang, are connected to the Changjiang River mainstream in the midstream. The Datong hydrological station is located downstream and approximately 600 km upstream of the river mouth. Based on the precipitation and runoff patterns in the catchment, the hydrological year of the Changjiang River is usually divided into flood (June–September), normal (with a moderate flow, April–May and October–November) and dry seasons (December–March).

2.2. Sampling and laboratory analysis

To study spatial patterns, surface water along the mainstream and tributaries and ground water samples were collected in April 2017 (normal season) and July 2018 (flood season) in the Changjiang River Basin (Fig. 1). The multi-year average runoff of the Yangtze River was approximately 900 Gm^3/yr (1960–2020), and the runoff in both 2017 (937 Gm^3/yr) and 2018 (875 Gm^3/yr) was close to it, suggesting a normal flow year in both 2017 and 2018 (Fig. S1). The average water discharge during the dry, normal and flood seasons in 2017/2018 were 44.5/44.6 Gm^3/month, 81.2/62.9 Gm^3/month and 111.3/94.4 Gm^3/month respectively. To study seasonal patterns, weekly to monthly samples were taken at Jiangyin Station in the lower reach of the Changjiang River in May 2013–May 2014, June 2016–December 2017 and May 2018–August 2019. Monthly concentrations of nutrients were averaged from weekly concentrations and monthly fluxes of nutrients were summed from weekly fluxes at Jiangyin Station. Daily samples were also taken at Jiangyin Station during the flood season from July 16 to September 1, 2020. Details on the field samples and laboratory analysis can be found in Text S1 and Table S1.

2.3. Historical observation data

To study long-term change patterns, historical data on nutrients concentration at Datong Station for the 1962–1999 period (Table S1 and Fig. 4)

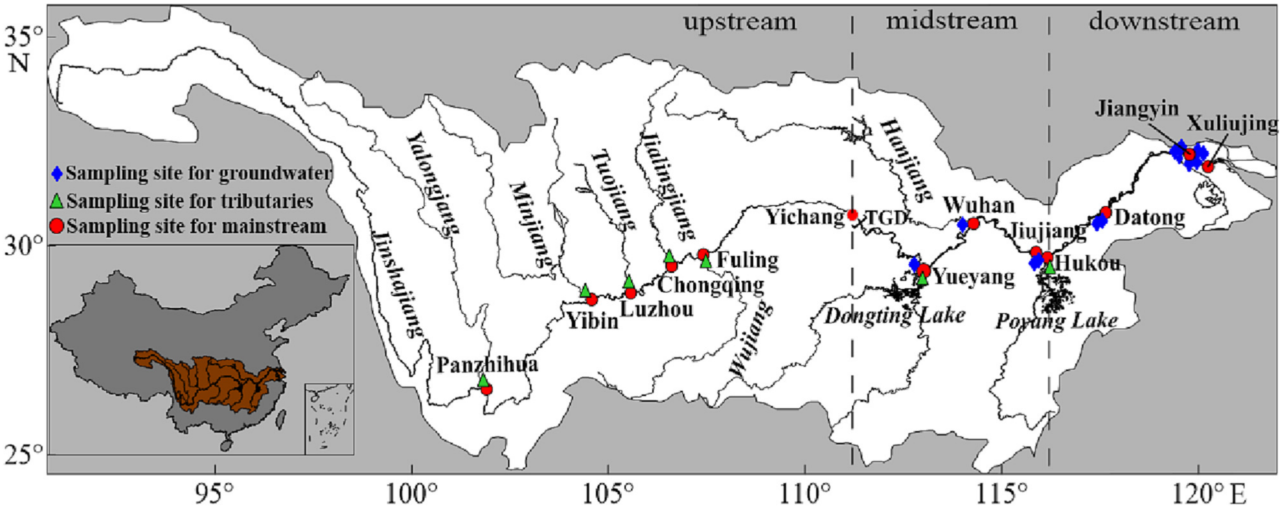

Fig. 1. Sampling stations along the Changjiang River, modified from (Wu et al., 2019a). The drainage area for each tributary is shown in Table S2. Groundwater samples were collected in the areas close to the river channel at Jiangyin, Datong, Jiujiang and Yueyang, and the site information is shown in Table S3.

period were obtained from the Hydrological Yearbooks of the People's Republic of China (Changjiang River Water Resource Committee, 1962-1999). Data on nutrients concentration at Datong Station for the 2000–2017 period were collected from Ge et al. (2020). Nutrient concentration data at Xuliujing Station (combined with data at Datong Station to represent the nutrient export to the estuary) for 2002 and 2008 (Table S1 and Fig. 4) were from Dai et al. (2011). There was no measurement of nutrient from Datong between 2002 and 2008, and the data collected from Xuliujing were thus used for that period, which is reasonable because there is no significant difference in nutrient concentrations and discharges between Datong and Xuliujing (Mei et al., 2019; Zhang et al., 2021a, 2021b). DSi concentration data for 2004–2020 at Datong Station were taken from Ran et al. (2022) and Ge et al. (2020). Water discharge at Datong Station for the 1962–2020 period were also obtained from the Hydrological Yearbooks of the People's Republic of China. For different frequencies of nutrients concentration, annual concentrations were averaged by month or week, and annual fluxes and water discharge were summed by month or week.

2.4. Data analysis

2.4.1. Nutrient fluxes

For each nutrient form, dissolved ammonium (NH_4^+-N), nitrate (NO_3^--N), nitrite (NO_2^--N), dissolved inorganic nitrogen (DIN), dissolved total nitrogen (DTN), dissolved phosphate (DIP), dissolved organic phosphorus (DOP), dissolved total phosphorus (DTP), particulate total nitrogen (PN), particulate inorganic phosphorus (PIP), particulate organic phosphorus (POP), particulate total phosphorus (PP), total nitrogen (TN) and total phosphorus (TP), the nutrient flux F (in mol/yr or mol/month) at a station on the Changjiang River at the measured time was calculated as follows:

$$F = C \times Q \tag{1}$$

where C is the mean concentration of the nutrient form at the station during the measured time (in mol/m^3) and Q is the annual or monthly average discharge (in m^3/yr or $m^3/month$).

2.4.2. Nutrient retention

The nutrient retention was calculated as follows:

$$F_R = F_{IN} + F_T - F_{OUT} \tag{2}$$

where F_R is the reservoir retention. Total inputs are the inflow at Fuling (F_{IN}) and the lateral inputs from the tributaries (F_T) of the TGD, and outputs are the outflow at Yichang (F_{OUT}). And the contribution of tributaries can be simplified as follows:

$$(1+q)F_{IN} - F_{OUT} - F_R = 0 \tag{3}$$

where q is the yearly (outflow-inflow)/inflow ratio of water through TGD to quantify the contribution of tributaries to the transport of nutrient for which no measurements were available. For this study, q equals to 0.05 (Ran et al., 2017).

2.4.3. Uncertainty and excess N

We used relative errors to quantify uncertainty in the fluxes (Runkel et al., 2004). Relative errors (δ) were calculated as follows on the basis of a Fourier series:

$$\delta = \sum_{i=1}^{n} \left| \frac{Q_i}{nQ_a} \frac{C_i}{C_a} \right| - 1 \tag{4}$$

where n is the number of samples in one year, Q_i is the daily discharge of the sampling time (in m^3/s), Q_a is the annual average discharge (in m^3/s), C_i is the nutrient concentration (in μmol/L), and C_a is the annual average nutrient concentration (in μmol/L).

We employed a Redfield ratio of 16 for DIN to DIP (Redfield et al., 1963) to quantify excess N ($N*$) (in μmol/L) (Zheng and Zhai, 2021), and the calculation was as follows:

$$N^* = DIN - 16 \times DIP \tag{5}$$

For nutrient limitation, we used DSi = 2 μmol/L, DIN = 1 μmol/L and P = 0.1 μmol/L as the absolute limits of nutrient concentration (Justić et al., 1995), and DIN:DIP = 16:1 and DSi:DIN = 1:1 as the potential relative limitation thresholds (Redfield et al., 1963).

2.4.4. Shapley Value approach decomposition

The Shapley Value technique (Shapley, 1953) was applied by Shorrocks on distributional analysis to decompose, through a regression-based approach, any inequality index as the sum of its contributing factors. Adopting this decomposition, we can estimate the marginal effect of each contributing factor, alone or grouped among similar variables, taking into account the order in which they can be eliminated. The overall contribution of each factor is obtained averaging the estimated marginal contributions across all the possible elimination sequences. The calculations were done using the Shapley2 package in State software and the equation can be expressed as follows:

$$\varnothing_i(v) = \Sigma_{S\subset N, i\in s} \frac{(|s|-1)!(n-|s|)!}{n!}[v(s) - v(s-\{i\})] \tag{6}$$

where $\varnothing_i(v)$ is the contribution of each variable, $v(s)$ is the total revenue created by the s variable, $v(s\text{-}\{i\})$ is the total revenue created by s-1 variables other than i, *'(|s|-1)!(n-|s|)!/n!'* is the weight.

2.4.5. Rates of change in nutrient concentrations

We employed nutrient change rate to quantify the effect of policy on changes in nutrient concentrations, which was calculated as follows:

$$R_i = \left(\frac{C_i - C_0}{C_0}\right) \times 100\% \Delta T > 0 \tag{7}$$

$$R_i = \left(\frac{C_0 - C_i}{C_0}\right) \times 100\% \Delta T \le 0 \tag{8}$$

where R_i is the rate of nutrient change, C_i is the nutrient concentrations (in μmol/L), C_0 is the nutrient concentration at the beginning of the policy implementation. '$\Delta T = 0$' is the year of policy implementation. For Ebro, Guadalquivir, Seine and Rhine River, we choose 1991 as the year when the policy was implemented due to dramatic decrease in the use of commercial fertilizers that started in the late 1980s and the legislation including the Nitrates Directive (Council EEC, 1991a) and the Urban Waste Water Treatment Directive (Council EEC, 1991b) was embedded for controlling eutrophication and nutrient loading into receiving waters. For Changjiang River and Yellow River, we choose 2015 as the initial year when the policy of fertilizer was implemented.

3. Results

3.1. Spatial-temporal changes in nutrient in the main channel of Changjiang River

3.1.1. Nutrient concentration

Along the Changjiang mainstream, the concentration of NO_3^--N significantly increased from the headwater to downstream, maintained stably high levels in the midstream and downstream, and slightly decreased at Hukou Station (at the confluence of Poyang Lake and the Changjiang mainstream) (Fig. 2). NO_3^--N was the major DIN form (with a fraction of >84 % in the normal season and > 95 % in the flood season), followed by NH_4^+-N (1.28 % ~ 14.2 % in the normal season and 0.19 % ~ 4.03 % in the flood season). DIN accounted for 81 % and 76 % of TN in the normal and flood seasons, respectively. As a result, the spatial distributions of DIN and TN concentrations along the Changjiang River mainstream were similar to those of NO_3^--N concentrations in both the normal and flood seasons. During the normal season, the highest NO_3^--N (123 μmol/L), DIN (131 μmol/L) and TN (151 μmol/L) concentrations were observed at Yueyang, Chongqing and Jiujiang Stations, respectively, and the lowest was at Panzhihua Station. During the flood season, the highest concentrations of NO_3^--N (143 μmol/L at Jiangyin), DIN (144 μmol/L at Jiangyin), TDN (167 μmol/L at Jiujiang) and TN (178 μmol/L at Fuling) were mainly observed in the midstream and downstream regions, while their lowest concentrations were found at Yibin Station upstream (Fig. 2).

In the normal season, the average concentrations of DIP, DTP, and TP along the Changjiang mainstream were 0.77 μmol/L, 1.19 μmol/L, and 1.91 μmol/L, respectively (Fig. 2), with their lowest concentrations of DIP (0.02 μmol/L), DTP (0.12 μmol/L) and TP (0.31 μmol/L) at Panzhihua and the highest concentrations of DIP (1.67 μmol/L), DTP (1.69 μmol/L) and TP (3.29 μmol/L) at Yueyang Station (Fig. 2). PP accounted for 29.4 % of TP at Yichang and 63.0 % at Datong, and POP contributed 0.7 % to TP at Yueyang and 44.7 % at Yichang. In the flood season, the average concentrations of DIP, DTP and TP in the midstream increased by 10.7, 5.1, and 2.5 times, respectively, compared with those in the upstream, and reached their maximums at Yueyang Station. POP dominated PP (73 %–96 %), and PP with concentrations of 0.47–2.76 μmol/L accounted for the largest proportion in TP (Fig. 2). Along the Changjiang mainstream, the concentration of TP remained high downstream, except at the Wuhan station (1.44 μmol/L). DTP increased from Yibin Station (0.49 μmol/L) to Yueyang Station (2.52 μmol/L) and then stabilized downstream. DIP overall increased from upstream to downstream, with the lowest concentration at Yichang Station (0.06 μmol/L) (Fig. 2).

PP was a prominent component of TP in the Changjiang River with a contribution of 30 %–63 % to TP in the normal season and 31 %–80 % in the flood season respectively, while PN only accounts for approximately 1 %–26 % of TN in normal season and 2 %–14 % in flood season. Both PP and PN showed a significant decrease at Yichang station (after TGD) in normal and flood seasons (Fig. 2).

Both in the normal and flood seasons, along the Changjiang mainstream, the DSi concentration decreased from Chongqing to Wuhan Stations, especially in the Three Gorges Dam (TGD) region, and then increased downstream, reaching the highest values at Hukou (130 μmol/L) and Jiujiang Stations (125 μmol/L) (Fig. 2). The spatial variation in nutrients in the major tributaries and groundwater of the Changjiang River can be found in Texts S2 and S3 and Tables S3, S5 and S6, respectively.

According to our preliminary estimates, approximately 11.5 % of TN, 4.2 % of TP, and 9.4 % of DSi were retained in the normal season, respectively, which is lower than the retention rates of 19.7 %, 41.3 %, and 11.2 % in the flood season. Particulate nutrients were more easily retained, with about 15.3 % of PP and 47.2 % of PN being intercepted in the normal season, and even more at 58.9 % and 56.6 % in the flood season.

3.1.2. Nutrient ratios

The molar ratios of DIN:DIP, DSi:DIN and DSi:DIP along the Changjiang River ranged from 76 to 1270, 1–4 and 61–4910 in the normal season, and from 225 to 1790, 1–21 and 79–2070 in the flood season, respectively (Fig. S2). The high DIN:DIP and DSi:DIP ratios were attributed to the extremely low DIP concentrations (0.02–0.14 μmol/L) upstream, such as at Panzhihua and Yibin Stations. At Luzhou Station, a DIN:DIP ratio of 107 in the normal season was much lower than that of 454 in the flood season. The ratios of DIN:DIP (420–1790) and DSi:DIP (417–2070) increased rapidly in the mainstream from Chongqing Station to Yichang Station (upstream of the TGD), especially at Yichang Station (DIN:DIP of 1960 and DSi:DIP of 1950) in the flood season. The ratios of DIN:DIP (183–345) and DSi:DIP (179–332) showed almost no spatial difference at the mid- and downstream stations in the flood season, respectively. In the normal and flood seasons, the DSi:DIN ratio slightly fluctuated and stabilized at approximately 1 downstream after Luzhou Station.

The ratios of DIN:DIP, DSi:DIN, and DSi:DIP in the major tributaries also showed large variation and were generally lower than their values in the corresponding mainstreams. The groundwater of the Changjiang River was enriched with DIN and DSi, while DIP concentration was relatively low, resulting in higher molar ratios of DIN:DIP in comparison with the surface water. The ratios of nutrients in the major tributaries and groundwater of the Changjiang River can be found in Texts S4 and S5.

3.2. Temporal changes in nutrient in the lower reach of Changjiang River

3.2.1. Monthly/seasonal changes in nutrient concentrations and fluxes

At Jiangyin Station during 2013–2019, NH_4^+-N and NO_2^--N concentrations were relatively lower in comparison with NO_3^--N concentrations and accounted for only approximately 3 % and 0.6 % of DIN, respectively, and showed obvious seasonal variations with maximums in the dry season (Fig. 3). DIN, more specifically NO_3^--N, accounted for 70–90 % of TN and was the main form of N in the Changjiang River. The DIN concentrations were negatively correlated with water discharge ($r = -0.41$, $p < 0.01$), indicating that both the lower DIN concentrations in the flood season and higher concentrations in the normal season were related to the seasonal variations in water discharge (Fig. S3).

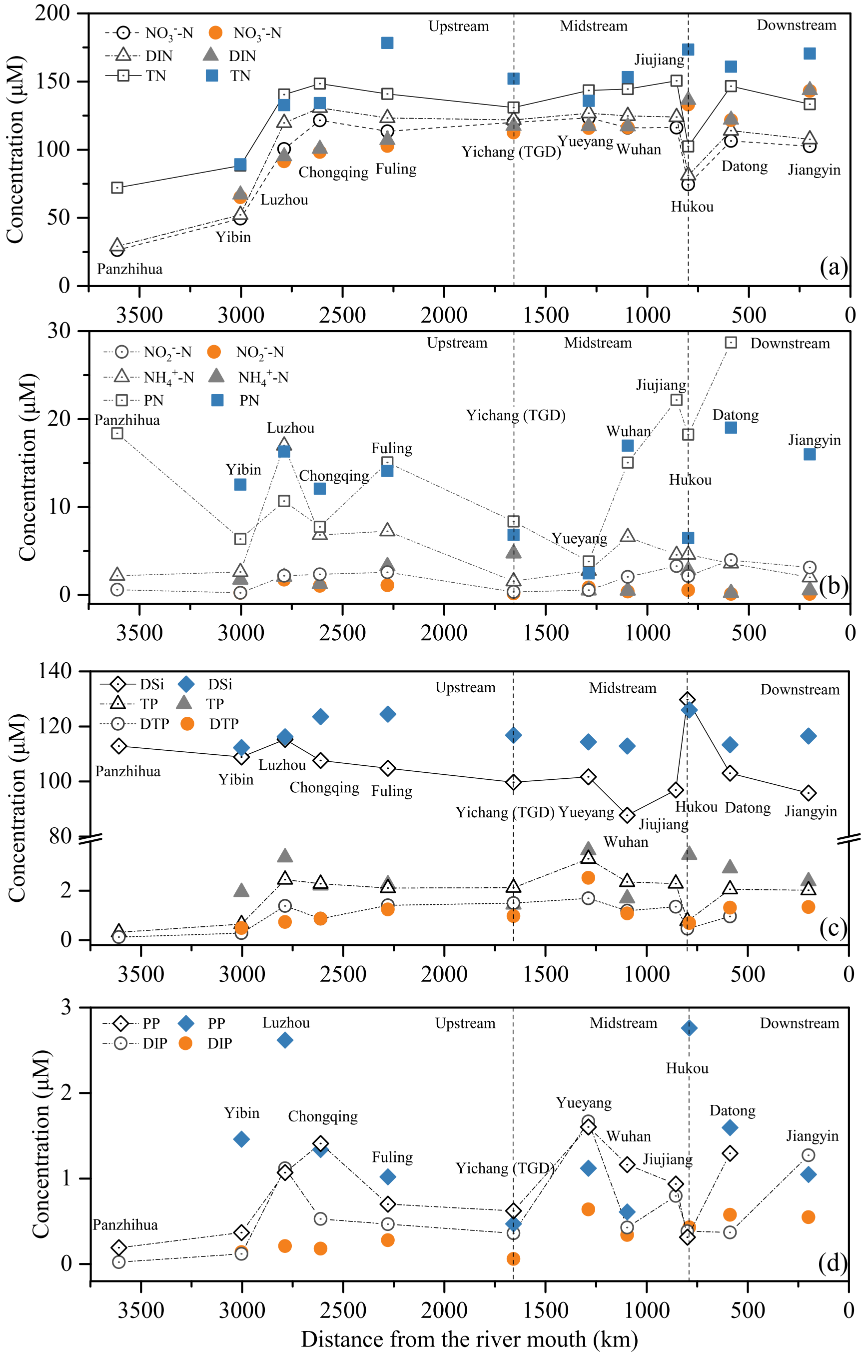

Fig. 2. Longitudinal distributions of nutrient concentrations in the Changjiang mainstream in the normal season in 2017 (open symbols) and flood season in 2018 (solid symbols). The distance represents the length of the river channel of the mainstream from the sampling station to the river mouth.

During 2013–2019, the average concentrations were 1.21 μmol/L for DIP and 1.47 μmol/L for TP downstream. DIP accounted for 82 % of TP and dominated TP abundance in the downstream of the Changjiang River. Similar to the DIN, the DIP concentrations were also negatively correlated with water discharge ($r = -0.46$, $p < 0.01$), and the highest concentration was in the dry period (Fig. 3 and S3).

The downstream DSi concentration ranged from 58 to 156 μmol/L and fluctuated during 2013–2019. And its concentration was higher in the flood and normal season than in the dry season (Fig. 3). Different from the seasonal patterns of N and P, the correlation between DSi concentrations and water discharge ($r = 0.03$, $p < 0.01$) was not obvious (Fig. S3).

The annual DIN, DIP and DSi fluxes were 90.2, 1.27 and 77.6 Gmol/yr during June 2013–May 2014, 109, 1.15 and 122 Gmol/yr during June 2016–May 2017, and 97.5, 0.5 and 79.7 Gmol/yr during June 2018–May 2019, respectively, of the Changjiang River (Table S4). DIN accounted for 74 % of the annual TN flux (122 Gmol/yr), while DIP accounted for 90 % of the TP flux (1.4 Gmol/yr) in the recent 10 years (Fig. 3). High fluxes of nutrient transport mainly occurred in the flood season. For example, during June–August, DIN fluxes in the flood seasons contributed 36 %, 41 % and 35 % to their annual fluxes, respectively. The variations in the fluxes of DIN ($r = 0.96$, $p < 0.01$) and DSi ($r = 0.93$, $p < 0.01$) during these periods were consistent with the water discharge, indicating that the seasonal DIN and DSi fluxes of the Changjiang River were mainly controlled by water discharge (Fig. 3), although the seasonal DIP flux was weakly correlated with water discharge ($r = 0.36$, $p < 0.01$).

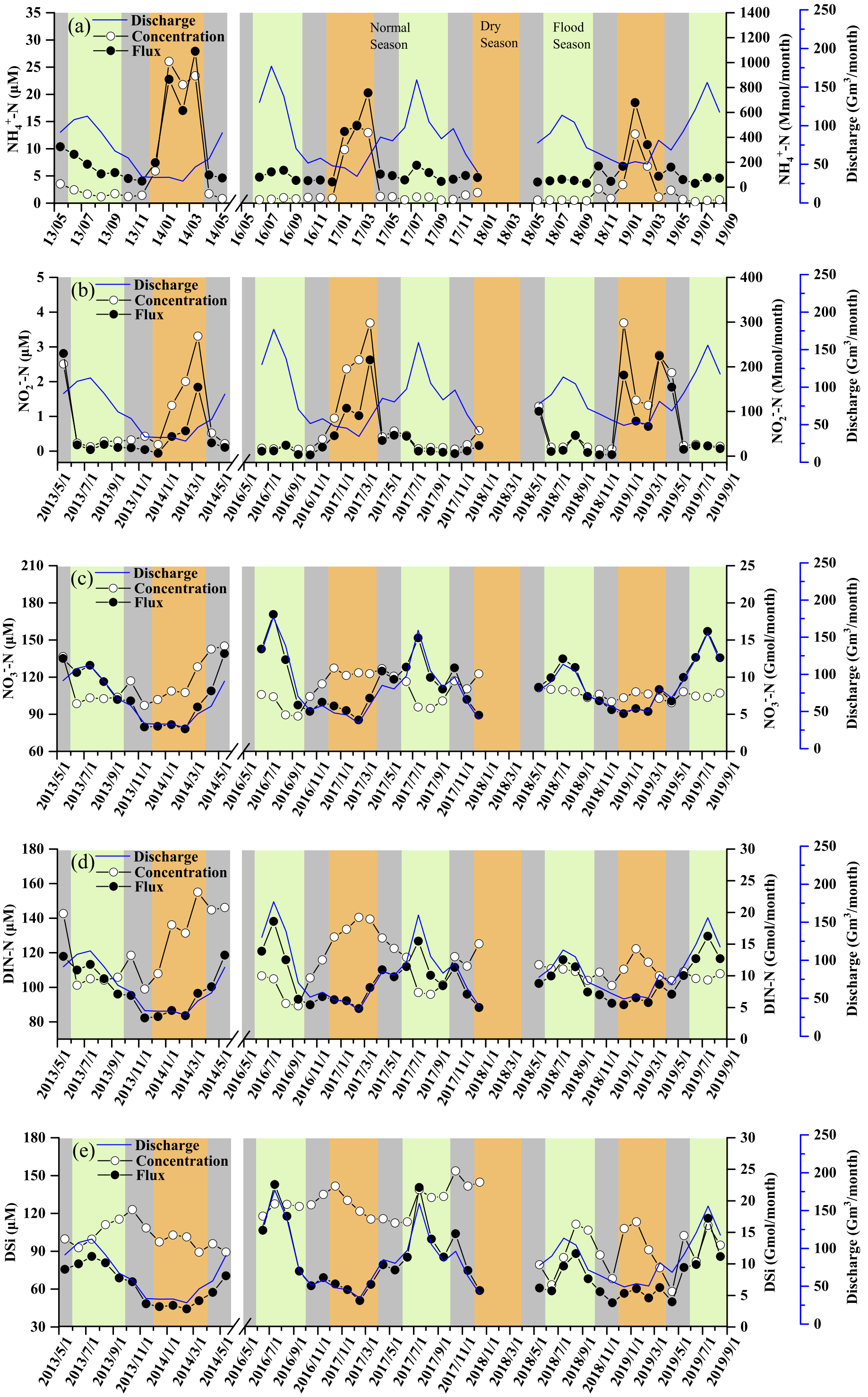

Fig. 3. Concentrations and fluxes of nutrients and water discharge downstream of the Changjiang River during 2013–2019. (a) NH_4^+-N, (b) NO_2^--N, (c) NO_3^--N, (d) DIN, (e) DSi, (f) DIP, (g) TN, and (h) TP. The data enveloped by the orange, green and gray areas were collected in the dry season, flood season and normal season, respectively.

The relative errors of annual fluxes for 2013–2019 of all nutrients except NH_4^+-N and NO_2^--N were <10 % (Table S4). The large monthly variation in NH_4^+-N and NO_2^--N concentrations may cause more uncertain estimates of their annual fluxes, but since NH_4^+-N and NO_2^--N only accounted for a small proportion (totally 0.6 %–2.4 %) of DIN, the estimate of DIN remains reasonable.

3.2.2. Daily changes in the flooding season

During rainstorm event (July 16–September 1, 2020) (Fig. S4), when the water discharge was at high levels, all forms of nutrients concentrations were significantly higher than that in the latter part time of the rainstorm event, especially for NH_4^+-N and NO_2^--N. As the water discharge decreased, the nutrient concentrations and fluxes returned to the normal level. The

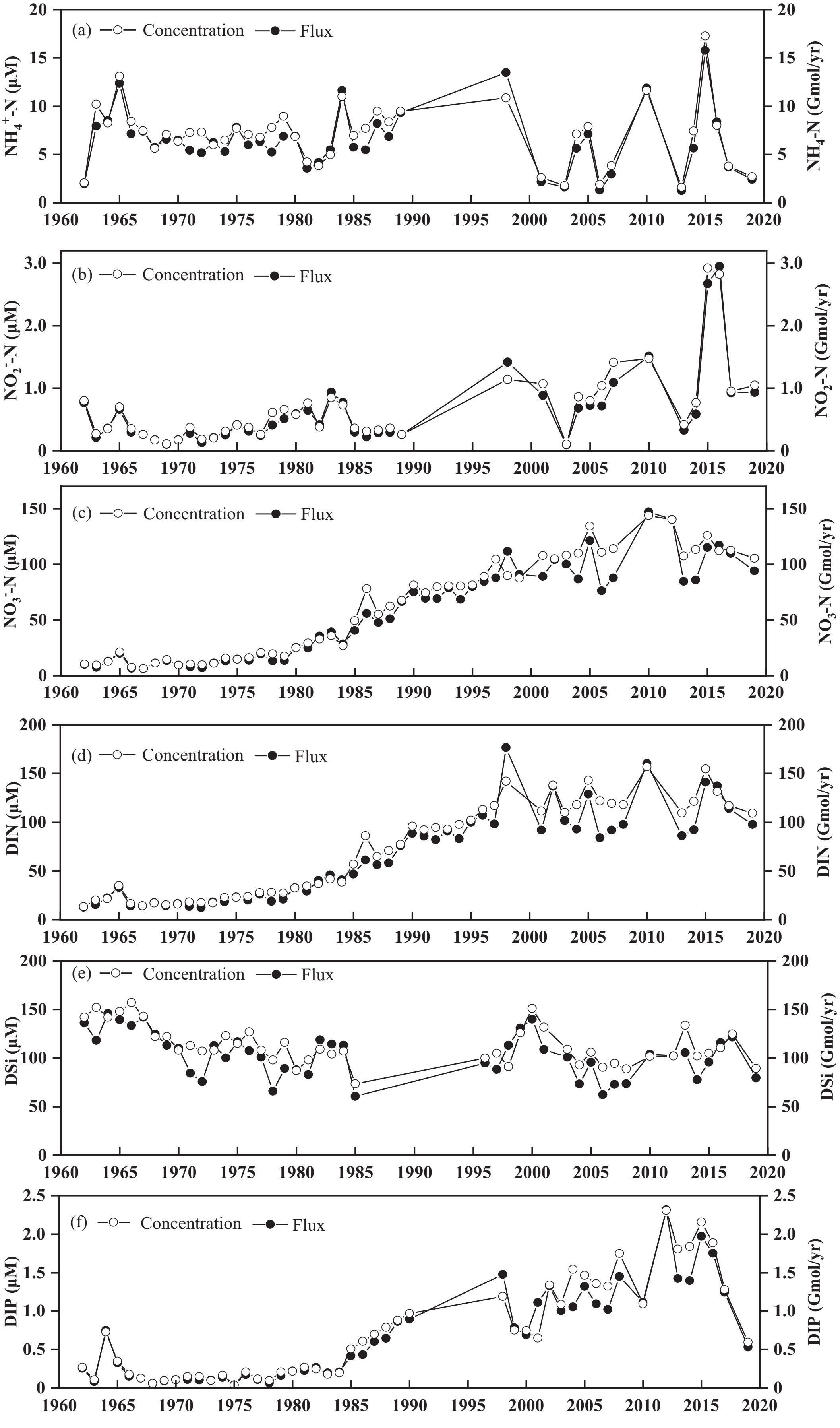

Fig. 4. Long-term changes in nutrient concentrations downstream of the Changjiang River: (a) NH_4^+-N, (b) NO_2^--N, (c) NO_3^--N, (d) DIN, (e) DSi, and (f) DIP.

concentration of TN decreased from 187 μmol/L to 92 μmol/L, and TP decreased from 10.5 μmol/L to 1.6 μmol/L.

3.2.3. Long-term changes in nutrient concentrations, fluxes and ratios

The DIN concentration was quite low before 1960, rapidly increased by approximately 2.5 times during 1960–1980, and reached the fastest growth rate after 1980. By the beginning of the 21st century, the average DIN concentration increased by 10 and 2.5 times compared with those in 1960 and 1980, respectively (Fig. 4). Similar to the DIN concentration, DIP concentration increased from 0.27 μmol/L in 1962 to 1.09 μmol/L in 2010. However, between 2015 and 2019, the DIP concentration decreased by approximately 74 % from 2.16 μmol/L to 0.57 μmol/L, and the DIN concentration decreased by approximately 29 % from 155 μmol/L to 109 μmol/L (Fig. 4). Different from DIN and DIP, the DSi concentration decreased by 36 % from 142 μmol/L in 1962 to 91 μmol/L in 2019.

Also, the long-term changes in the nutrient fluxes at Datong Station could be generally divided into three stages. The first stage was from the 1960–1980, during which the fluxes of DIN and DIP showed steadily slow increasing trends (from 18 to 34 Gmol/yr for DIN and from 0.12 to 0.21 Gmol/yr for DIP). The second stage was from the 1980–2010, during which the DIN and DIP fluxes rapidly increased (by approximately 4 and 6 times, respectively, Table S5). The third stage was the last 10 years, during which the DIN and DIP fluxes tended to be stable and growth tended to decline. Between 2015 and 2019, while the annual average DIN flux was stable, the DIP flux decreased by 0.39 Gmol/yr and 0.25 Gmol/yr in comparison with that in the 2010–2015 and 2010s periods, respectively (Table S5). On the long-term time scale, the annual nutrient export from the Changjiang River seems to have been influenced more by its concentration ($r = 0.97, p < 0.01$) than discharge ($r = 0.20, p < 0.01$).

Multiple regression analysis (ordinary least squares) and decomposed r-squared values show that the reduction in P fertilizers contributed the most to the decline in DIP flux (44.9 %), followed by DIP discharge from sewage (pollution control, 24.9 %), groundwater (17.6 %) and water discharge (12.7 %), while other factors had no significant impact (e.g., damming) on the decline in DIP after 2015. And DIN Shapely values showed that N fertilization (88.2 %) was also the main cause of the variation in DIN concentration, followed by groundwater (6.38 %) and water discharge (5.43 %).

The different changes between N and P were also found in the nutrient concentrations and fluxes in the up- and midstream reaches of the Changjiang River (Fig. 5). From the 1960s to the 1980s, the DIN flux increased in both upstream (by 1.6 times at Panzhihua Station and 4.0 times at Chongqing Station) and midstream (by 2.6 times at Yichang Station and 2.0 times at Wuhan Station). The DIN flux increased by 2–3 times in the upstream and midstream regions during 1980–2000 (Fig. 5). The changes in the DIP flux during the 1960–2000 were similar to those of DIN, but the DIP flux in the up-, mid- and downstream reaches largely decreased in 2017 compared to those in 1997 and 2008, which is different from the continuous steady increase in the DIN flux (Fig. 5). The DSi flux slowly decreased during the 1960s–1980s. Except for anomalously high values in 1998 (flood year), DSi fluxes continued to decline from 1980 to 2000, but at a slower decline rate than in 1960–1980 (Fig. 5). After the 2000s, the change in DSi fluxes was less significant. It is thus clear that the present fluxes of DIN and DIP from the Changjiang River to the estuary have increased, and that of DSi have declined in comparison with the values of 1960s.

There might have been no obvious change in DON and DOP based on our measurements and few historical data (Liu et al., 2003; Fig. S5). Since long-term measurements of DON and DOP in the Changjiang River are

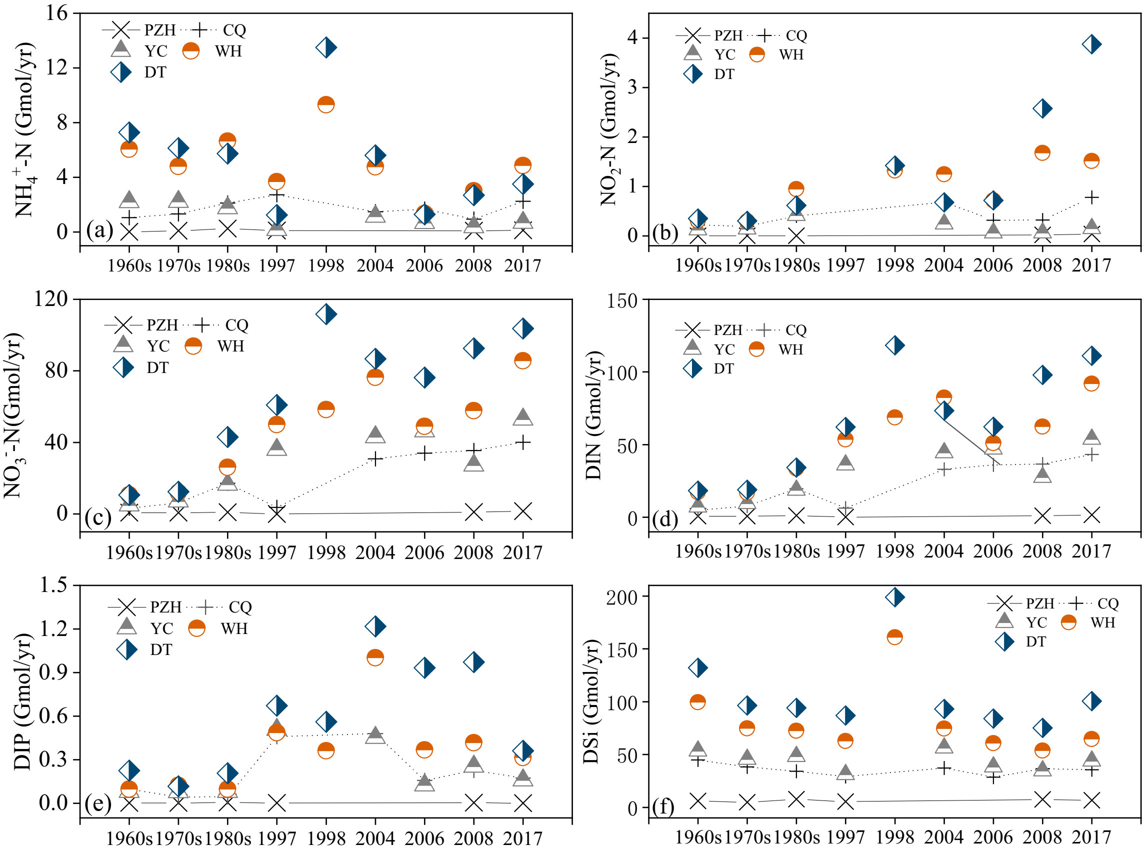

Fig. 5. Long-term changes in nutrient fluxes in the major channel of the Changjiang River: (a) NH_4^+-N, (b) NO_2^--N, (c) NO_3^--N, (d) DIP, (e) DSi, and (f) DIN. PZH represents Panzhihua, CQ represents Chongqing, YC represents Yichang, WH represents Wuhan, and DT represents Datong. Data for 1960s–1980s were obtained from Changjiang River Water Resource Committee, (1962-1989). Data for 1997 and 1998 were obtained from Liu et al. (2003) and Shen and Liu (2009). Data for 2004, 2006, 2008, and 2017 are from the present study.

lacking, the variation in DON and DOP in recent decades remains largely uncertain.

During the 1960s–2000s, the molar ratio of DIN:DIP largely increased from 125 in the 1960s to 201 in the 1970s and decreased to 98 in the 2000s, while that of DSi:DIN and DSi:DIP consistently decreased from 8.0 and 984 in the 1960s to 0.9 and 105 in the 2000s, respectively, in the Changjiang mainstream (Fig. S6), resulting in excessive DIN. Meanwhile, an obvious decline in the NH_4^+-N:NO_3^--N ratio was observed from 1.07 in 1963 to 0.03 in 2019.

4. Discussion

4.1. Change in nutrient limitations

Since the 1960s, the nutrient ratios in the Changjiang River had been much higher than the Redfield–Brzezinski values of N:Si:P in a range of 16:(15– 20):1 (Brzezinski, 2004; Redfield et al., 1963). Although all the observed nutrient concentration exceeded absolute limits of nutrient concentration (Justić et al., 1995), the stoichiometric DSi:DIN, DSi:DIP and DIN:DIP ratios showed obvious decadal variation in the Changjiang River. Our high-resolution data indicated that after rapid increases in previous decades, the variation of the DIN:DIP ratios have stabilized after 2000 (Fig. S6), in contrast to the observations of previous studies using limited data (e.g. Wang, 2006; Liu et al., 2022a, 2022b). This change in nutrient ratios could be largely due to the enhanced human activities, especially the variation in fertilizer uses (Liu et al., 2018) and the construction of dams (Ran et al., 2022). As a result, in the Changjiang River, nutrient limitation has developed toward P and Si limitations (Fig. S7), characterized by high DIN and low DIP and DSi concentrations (Figs. 4 and S2) (Wu et al., 2019b). In recent years, fertilizers have shown a continuous reduction trend, and DIP concentrations have decreased more significantly than DIN (Fig. 4) due to the lack of agricultural groundwater replenishment (Van Meter et al., 2016), which would exacerbate the imbalance of N/P ratio. On the other hand, compared to N, P show preferential retention in lacustrine systems (Wu et al., 2022). As a result, a large amount of TP could be retained in the reservoir area (Tian et al., 2021). This process, however, has contributed minimally to the trapping of DIN in the TGD (Ding et al., 2019; Ran et al., 2017). The DIN:DIP in the Changjiang River could continue to increase in the future, particularly in the mid- and downstream reaches (Fig. S7) and in the estuarine system.

The long-term changes in the NH_4^+-N:NO_3^--N ratio and N* also reflect the influences of recent prevention and control of water pollution and hydropower projects on the Changjiang River during different periods (Xie et al., 2008). The high NH_4^+-N:NO_3^--N ratio at the Luzhou to Yichang stations in both the normal and flood seasons (Fig. S2) could be largely due to water pollution in the upper reach. Since the beginning of the 21st century, some key policies have been implemented in the Changjiang River, including the 'Law of the People's Republic of China on Prevention and Control of Water Pollution' (National People's Congress Standing Committee, 1996) and 'Reduce total ammonia nitrogen' (National People's Congress, 2001), and have greatly alleviated pollution (Fig. S8). Human activities through the improvement of urban sewage (decreased NH_4^+-N:NO_3^--N ratio) (Fig. 6; $R^2 = 0.32$, $p < 0.05$, $n = 15$) and application of agricultural N fertilizer have changed the N source in the basin, causing increased NO_3^--N export, decreased NH_4^+-N export and decreased NH_4^+-N:NO_3^--N ratio and N* ($R^2 = 0.17$, $p < 0.05$, $n = 15$) in recent decades (Fig. 6).

4.2. Drivers of changes in nutrient concentrations and fluxes

4.2.1. Damming effect

The concentrations of NO_3^--N, DIN, TN, PN, DIP and PP decreased from Fuling Station to Yichang Station (40 km downstream of TGD) (Fig. 2), largely due to the retention effect of the TGD. After the implementation of the TGD, the TP flux exported into the Changjiang River estuary decreased by 12 % (Ding et al., 2019), which is close to our estimation. A modeling study also showed that the TGD has contributed annually to 5 % of TN and 7 % of TP basin-wide retention (Liu et al., 2018), which generally agrees with our observations. DIP and TP are more easily trapped in reservoirs and have higher retention rates than DIN and TN. As a rough estimation, the TGD played an important role in the trapping of the incoming PP (0.32×10^4 t/yr PP) due to the trapping of suspended sediment and more remarkable in the flood season (58.9 %) than in the normal season (15.3 %), suggesting a fairly higher retention rate than that of dissolved forms of nutrient. Nevertheless, the transformation of DON, nitrite and ammonia to nitrate in the reservoir (Ran et al., 2017) may also lead to the

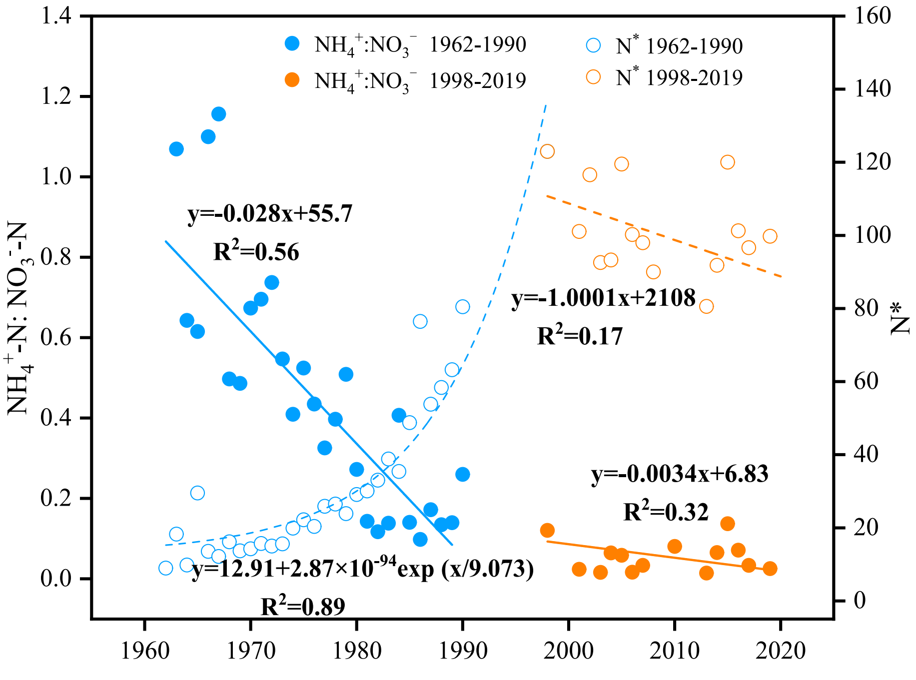

Fig. 6. Scatter diagram of NH_4^+-N:NO_3^--N ratios and N* downstream of the Changjiang River during 1962–2019.

longitudinal increase in NO_3^--N in the downstream reach of the TGD (Fig. 2), which is the balance between formation and retention of NO_3^--N. Since the operation of the TGD, the DSi concentration at Yichang Station (downstream of the TGD) has decreased (Fig. 2 and Table S3), which is consistent with previous observations (Ding et al., 2019). In the reservoir area, the increased diatom biomass (Wang et al., 2022) can remove more nutrients by sinking diatom detritus. However, this additional retention for nutrients, especially for the particulate nutrients, is insufficient to change the overall trend of N and P in the export due to the rapid increase in delivery to the river.

4.2.2. Change in fertilizer application

The rapid increase in the N and P concentrations in the Changjiang River is related to the intensive agricultural activities in the basin. With low population density and agricultural activity (Liu et al., 2003), the NO_3^--N, DIN, TN and DIP concentrations in the Jinshajiang River (headwater of the Changjiang River) upstream were much lower than those in the mid- and downstream reaches (Figs. 2 and 5). The model simulations indicated that agricultural surface runoff has accounted for 40 %–80 % of TN and 70 %–80 % of TP delivered by rivers since 2000 (Liu et al., 2018). The midstream TN and TP concentrations were thus higher in these areas than in other reaches (Fig. 2). Studies have shown that the increases in NO_3^--N and DIN concentrations in the Changjiang River Basin during 1958–1985 have significant positive correlations with the use of chemical fertilizers in the basin (Duan et al., 2007). The rapid increases in the DIN and DIP concentrations were also consistent with the increases in fertilizer application, agricultural area and urban sewage discharge during 1980–2000 in the Changjiang River (Fig. S9). Over the past 40 years, agricultural fertilizers have shown a significant correlation with the concentrations of DIN, DIP and N* (Fig. S8). However, the average fertilizer uses during 2016–2019 decreased by 1.8 Mt./yr compared to 2010–2015. As a general estimation using the same proportion modeled by Liu et al. (2018), the decline in fertilizer use in recent years could result in a reduction of P export by −0.32 - -0.28 Gmol/yr, and the decline rate was approximately −5 %/yr in recent years. A further decline in P flux in the Changjiang River might occur due to the continuous reduction of P fertilizer in the basin (Fig. S10). Similar N and P flux trends were also observed in the Yellow River (Wu et al., 2021).

4.2.3. Contribution of groundwater and wastewater discharge

TN concentrations in groundwater along the Changjiang River in both the normal (Table S6) and flood seasons (Table S7) were 3–4 times higher than that in the river, while the groundwater TP concentration was similar to that in the river, which suggests that excessive N in groundwater may be an important supply of N in the river. P has a much lower movability than N in the river basin due to the difference in chemical activities (Alexander et al., 2008), which regulates the abundance of P in the groundwater. And anthropogenic sources, including agricultural, industrial, and aquacultural activities, can result in the enrichment of N (Shuler et al., 2017). Groundwater receives nutrient inputs mainly from leaching (Puckett et al., 2011). The residence times of groundwater vary from years to decades or even longer, and this means that large N and P amounts are tend to store in aquifers in the river basin. This legacy implies that the surface water concentrations will persist for decades even after the fertilizer inputs have ceased (Van Meter et al., 2016). Although the exchange of groundwater with surface water in the Changjiang River Basin is declining (Fig. S9), groundwater remains an important source of N for surface water.

The DIN and TN concentrations downstream remained high (Fig. 2), which is partly due to industrial production and urban sewage discharge (Liu et al., 2018). In the Changjiang River Basin, the share of wastewater discharge contributed to 9 % for N and 11 % for P (Liu et al., 2018). However, despite the increase in sewage fluxes, total N discharged from sewage was reduced by half during 2016–2019 (0.53 Mt./yr) compared to 2011–2014 (1.17 Mt./yr) (Fig. S9) due to the improvement of wastewater treatment efforts (Liu et al., 2022a, 2022b). Moreover, waste produced in the P industry including tailing pond leakage, ore mining and wastewater discharge are decreased due to the special action launched by government (Xu et al., 2018). In contrast to the decline in DIP flux in recent years, we consider that the N stabilization is largely attributed to the replenishment of excess N in agricultural groundwater. Similar to the Mississippi River in the 1970s, previous rapid fertilizer growth allowed N to enter the groundwater and hindered N decline in the Changjiang River (Liu et al., 2018), thereby contributing to a more stable riverine N flux.

4.2.4. Contribution of changes in basin management

Land-use change is a major factor that influences nutrient export in the Changjiang River Basin (Zhang et al., 2021a, 2021b). In recent years, the decrease in the agricultural land area might reduce the supply of N and P from the basin soil to the river water in the Changjiang River Basin (Fig. S9). And the recent policy change, such as reducing fertilizer use and reducing TP in sewage (Figs. S9 and S10) could alleviate P pollution in the Changjiang River. Nevertheless, these policy strategies of both reduction N and P supply, which have only been implemented in the last 5 years, did not simultaneous reduce the concentrations and fluxes of DIN (Fig. 4 and Fig. 5) and TN due to the significant contribution of agricultural activities to N flux.

The Changjiang River is classically seen as a large high-nutrient river, which previous studies have predicted will further increase (Blaas and Kroeze, 2016; Liu et al., 2018). However, in this study, we observed a probable decrease in the delivered P in the Changjiang River from 2015 to 2019. We consider that the decline in P (mainly DIP) may be related to some recent policies (e.g., decreased agricultural area, increased forestry area, decreased fertilizer application and N and P discharge from sewage (Figs. S9 and S10). Before 2000, N fertilizer consumption has been increasing faster than cereal yield gains, triggering a steady decline of N use efficiency in cereal systems in China (Heffer and Prud'homme, 2016). In developed countries, N use efficiency has undergone steady improvement over the past three decades, driven by the adoption of fertilizer best management practices. Similar trends are observed for N fertilizer applied to maize in the United States, wheat in West Europe or rice in Japan (Fixen et al., 2014). However, a reversal of trend is observed after 2000, reflecting the government's new focus on resource efficiency. From 1998 to 2016, the utilization efficiency of fertilizer in China increased from 29.1 % to 39.8 % (He et al., 2019). Meanwhile, N use efficiency in croplands increased from 22 % to 30 % from 2002 to 2015 (Omara et al., 2019). And, the use of N fertilizer in China (kg yield/kg N) decreased obviously (Tong et al., 2003). It is expected that fertilizer application could be reduced by 30 %–60 % without any loss of crop yields (Ju et al., 2009).

Multiple regression analysis of the above influencing factors with DIP fluxes was performed using a Shorrocks–Shapely decomposition of R-squared values. The results show that the main reason for the decline of DIP flux is the decline in phosphate fertilizer and wastewater discharge, with less effect of natural factors such as changes in groundwater and runoff, while other factors had no significant impact (e.g., damming) on the decline in DIP after 2015. The factor of DIN flux reduction is also mainly the reduction of nitrogen fertilizer, and the effect of sewage discharge on N seems to be more retarded than P. There are still some uncertainties in our assessment of the contributing factors that result in DIP and DIN decline due to the limited amount of data. Our estimation is reasonable because riverine P and N is dominated by fertilizer contributions (Liu et al., 2018), and there was no large reservoir operation during 2015–2019 on the Changjiang River.

4.3. Comparison with other major rivers

Similar variations in riverine fluxes of N and P influenced by anthropogenic inputs have been observed in the major rivers worldwide. Compared with the rapid growth of nutrients in Asian rivers (such as the Changjiang River and Mekong River) in recent years (Li and Bush, 2015), the slow growth of nutrients in European rivers (Ludwig et al., 2010) may be related to a series of N and P bans issued by the European Union to control water eutrophication since the 1980s (Ludwig et al., 2010). Again, in comparison with Asia, the relatively higher wastewater treatment efficiency in Europe was also an important factor that contributed to the decline in nutrient yield. Although the fluxes of nutrients in the Changjiang River are significantly higher than those in other rivers in the world, the fluxes of nutrients per capita area are lower than the world average level (Table S8).

Many other large rivers in the world, including the Rhine, Ebro, Yellow, Guadalquivir and Seine Rivers, have experienced reduction in fertilizers application during the past decades (Fig. S11). We found that the decrease rate of P concentration in these rivers increased in the first 10 years after the implementation of the fertilizer policy (Fig. 7), while the change rate of N concentrations has not change significantly, mainly due to N legacies in the soil and groundwater that can serve as a stable and long-term sources to surface waters. The Changjiang River, which is experiencing what these rivers have experienced, is likely to continue to decrease in P in the future after a series of changes in basin management including fertilizers reductions and land use change, while significant reductions in N may take >10 years (Van Meter et al., 2018). In other words, current reductions in N use may have little effect on short-term changes in N loading at the catchment outlet.

The unbalanced nutrient ratios in the river have become more serious in recent decades, especially for the ratios of DSi:DIN and DSi:DIP in most world rivers. In less disturbed rivers, such as the Amazon and Zaire rivers (Lin et al., 2019;), the DSi:DIN (~8.8:1 and 26:1, respectively) and DSi:DIP ratios (~100:1 and 190:1, respectively) are higher than those in other rivers (Ludwig et al., 2009). For the rivers in developed areas, such as the Mississippi (Chen et al., 2012), Rhine (Loos et al., 2009) and Rhône (Moutin et al., 1998) rivers, the DSi:DIN and DSi:DIP ratios are the lowest and close to the Redfield–Brzezinski ratio. In the Changjiang River and other Chinese rivers (e.g., the Pearl River and Yellow River) compared with other world rivers, the DSi:DIN ratio was lower, whereas the DSi:DIP and DIN:DIP ratios were higher, resulting in relatively high P limitation in these Chinese rivers. The unbalanced DSi:DIP and DIN:DIP ratios and increased DIN fluxes from Chinese rivers might be responsible for many environmental problems in coastal seas (Li et al., 2014). With the projected decreasing demand for fertilizer in the river basin, decline in P and NH_4^+-N will continue in the coming decades. Although implementation of the government's sustainability policies leads to declining DIP loads, the trajectory of high DIN:DIP ratios will persist in the near future. The Changjiang River and its estuary would also encounter fairly high DIN discharge issues with considerable agriculture in the next two decades, compared to other major rivers around the world. Policies to reduce nitrogen export and to improve fertilizer and manure use efficiencies are thus particularly important, especially in the midstream and upstream.

4.4. Implications for the East China Sea

Affected by the incongruous changes in N, P and Si concentrations, the composition of nutrients downstream of the Changjiang River has changed largely during the past 60 years (Fig. 4). These changes in nutrient may further strengthen the P limitation in the Changjiang River in the future. Enhanced P limitation could contribute to changes in the environment of the estuary and coastal areas, such as the enhancement of seasonal hypoxia (Gan et al., 2014). Our results also suggest that increased riverine N and P fluxes could also lead to enhanced deoxygenation based on the relationship between riverine nutrient export and hypoxia, especially the increase in river P export (Fig. S12).

With the increases in nutrient loadings, the frequency and area of red tides in the Changjiang River estuary and adjacent waters have increased obviously in recent decades (Jiang et al., 2014; Xu et al., 2020; Wang et al., 2023). And the imbalance of the nutrient structure in the Changjiang River could also cause important changes in the community structure of phytoplankton in coastal ecosystems (Wang et al., 2021b; Xiao et al., 2018). Our results show that the increase in riverine nutrient fluxes could increase primary productivity but reduce biodiversity in the Changjiang River Estuary (Fig. S12). Both the decreased DSi flux and increased DIN flux in recent years could reduce the number of diatoms and enhance the number of dinoflagellate species (Fig. S12). A strong positive relationship is also observed between the DIN and DIP concentrations of Changjiang River and the frequency or scale of HABs in the Changjiang estuary, indicating that land-based nutrients played a crucial role in the occurrence of HABs.

Our understanding of changes in nutrient ratios and fluxes in a major river is important for understanding riverine and coastal nutrient budgets

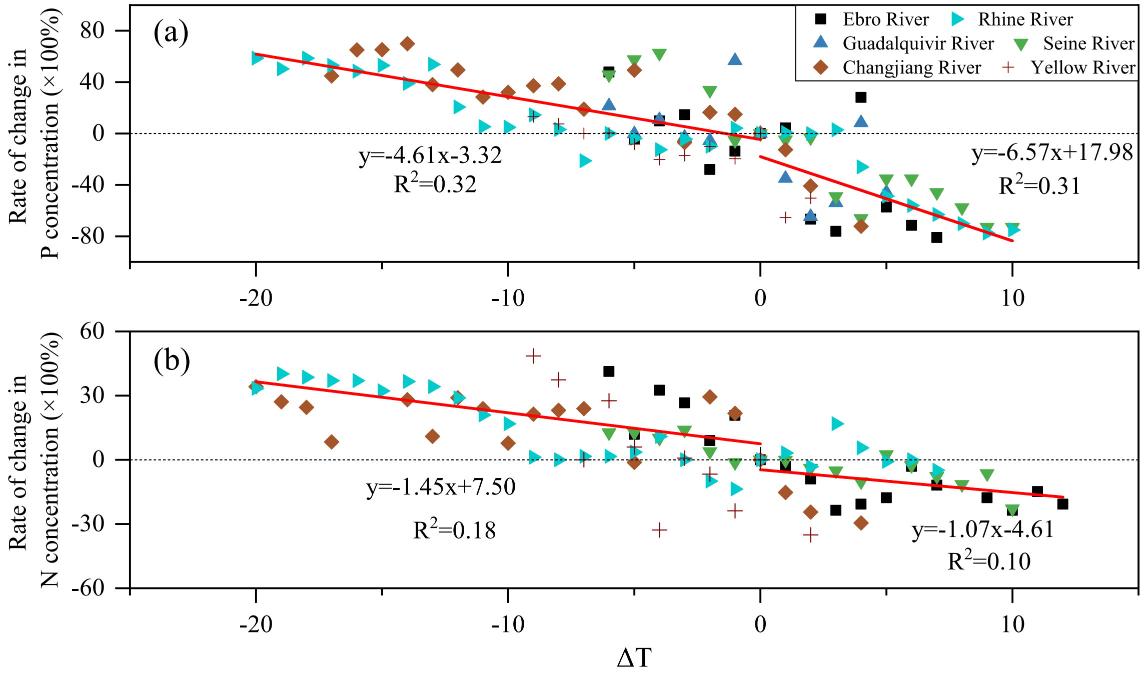

Fig. 7. Rates of change in DIN and DIP concentrations in the Changjiang River and TN and TP in other major rivers worldwide after the implementation of watershed management policies. The horizontal coordinate 'ΔT = 0' is the initial year of policy implementation. Data on the Ebro, Guadalquivir and Seine River are from Bouraoui and Grizzetti (2011); data on the Rhine River are from Grimvall et al. (2000); data on the Yellow River are from Zheng et al. (2022).

and mitigating eutrophication. Several significant conclusions and questions arise from our analysis of nutrient transport, and the effects of human perturbations may help to guide future research. For example, it has only recently become clear that the delivery of P is highly dependent on the variation in human activities in the river basin (Wu et al., 2022). Studies designed to examine the mechanisms and ecosystem influence of these changes in nutrient patterns are needed to assess how future human activities will amplify and dampen nutrient transport from major rivers and its impact.

5. Conclusions and remarks

This study showed an overview of the spatial pattern, temporal variation, and drivers of nutrient concentration and export for the Changjiang River. We explored 60-year time series nutrient to assess how nutrient change in this major river. We saw that nutrient concentration, form and export have varied over time, which would affect the river and coastal ecosystems.

Over the past 60 years, the DIN and DIP concentrations and fluxes of the Changjiang River experienced slow increases during 1960–1980 and rapid increases during 1980–2000; in recent 5 years, P decreased, while N and Si remained stable. Although NO_3^--N and DIP have shown an increasing trend in recent decades, a different trend between DIN and DIP has been observed recently. The relatively slow increase in DIP suggests that a turning point of DIP change since 2015 compared with DIN might have occurred in the Changjiang River. Si and P limitations have been enhanced. The decline in P fertilizer contributed approximately 55 % to the decrease in DIP flux during 2015–2019 in the Changjiang River.

NH_4^+-N rapidly decreased between 2015 and 2020, which implies a change in the N form composition with a much lower NH_4^+-N:NO_3^--N ratio than a few decades ago and an improvement in water quality. This change in N species may be responsible for the recent decreases in fertilizer and pollutants in the Changjiang River Basin.

The rapid alteration of nutrient transport in this large river is taken as an indicator of the complex transformation in nutrient concentrations and fluxes in the river basin. The nutrients in the Changjiang River may change differently due to the continuous promotion of protection policies made by the Chinese government and an increase in public environmental awareness. Recent observations of nutrient concentrations in the major rivers indicate that decreasing P loading could occur in the future if changes in fertilizer use are implemented. These change in nutrient deliver of river basins and estuaries need to be considered when assessing the impact of human activities for coastal ecosystem.

Data availability

All data that support this study are available in the paper and supplementary materials.

Declaration of competing interest

All authors declare no conflict of interests.

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CRediT authorship contribution statement

Wentao Wu and Xiangbin Ran: Conceptualization, Methodology and Investigation. **Wentao Wu**: Data curation, Formal analysis and Writing - original draft. **Xiangbin Ran and Junjie Wang:** Writing - review & editing, Supervision. **Hao Wang**: Investigation, Resources. **Jun Liu, Qingzhen Yao and Zhigang Yu**: Review & editing.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.162268.

References

Alexander, R.B., Smith, R.A., Schwarz, G.E., Boyer, E.W., Nolan, J.V., Brakebill, J.W., 2008. Differences in phosphorus and nitrogen delivery to the Gulf of Mexico from the Mississippi River basin. Environ. Sci. Technol. 42, 822–830.

Beusen, A.H.W., Bouwman, A.F., Van Beek, Mogollón, J.M., Middelburg, J.J., 2016. Global riverine N and P transport to ocean increased during the 20th century despite increased retention along the aquatic continuum. Biogeosciences 13, 2441–2451.

Blaas, H., Kroeze, C.J., 2016. Excessive nitrogen and phosphorus in european rivers: 2000–2050. Ecol. Indic. 67, 328–337.

Bouraoui, F., Grizzetti, B., 2011. Long term change of nutrient concentrations of rivers discharging in European seas. Sci. Total Environ. 409, 4899–4916.

Brzezinski, M.A., 2004. The si:C: N ratio of marine diatoms: interspecific variability and the effect of some environmental variables1. J. Phycol. 21, 347–357.

Changjiang River Water Resource Committee, 1955-1999. Hydrological Yearbooks of the People's Republic of China (in Chinese), Beijing.

Chen, Y., Liu, R., Sun, C., Zhang, P., Feng, C., Shen, Z., 2012. Spatial and temporal variations in nitrogen and phosphorous nutrients in the Yangtze River estuary. Mar. Pollut. Bull. 64, 2083–2089.

Chen, X., Strokal, M., Kroeze, C., Ma, L., Shen, Z., Wu, J., Chen, X., Shi, X., 2019. Seasonality in river export of nitrogen: a modelling approach for the Yangtze River. Sci. Total Environ. 671, 1282–1292.

Chen, Q., Shi, W., Huisman, J., Maberly, S.C., Zhang, J., Yu, J., Chen, Y., Tonina, D., Yi, Q., 2020a. Hydropower reservoirs on the upper Mekong River modify nutrient bioavailability downstream. Natl. Sci. Rev. 7, 1449–1457.

Chen, X., Strokal, M., Kroeze, C., Supit, I., Wang, M., Ma, L., Chen, X., Shi, X., 2020b. Modeling the contribution of crops to nitrogen pollution in the Yangtze River. Environ. Sci. Technol. 54, 11929–11939.

Council, EEC., 1991a. Directive 91/676/EEC concerning the protection of waters against pollution caused by nitrates from agricultural sources, L 375. Off J Eur Communities.

Council, EEC., 1991b. Directive 91/271/EEC concerning urban waste water treatment, L 135. Off J Eur Communities.

Dai, Z., Du, J., Zhang, X., Su, N., Li, J., 2011. Variation of riverine material loads and environmental consequences on the Changjiang (Yangtze) estuary in recent decades (1955–2008). Environ. Sci. Technol. 45, 223–227.

Ding, S., Chen, P., Liu, S., Zhang, G., Zhang, J., Dan, S.F., 2019. Nutrient dynamics in the Changjiang and retention effect in the three gorges reservoir. J. Hydrol. 574, 96–109.

Duan, S., Xu, F., Wang, L.J.J.B., 2007. Long-term changes in nutrient concentrations of the Changjiang River and principal tributaries. 85, pp. 215–234.

Feng, X., Fu, B., Piao, S., Wang, S., Ciais, P., Zeng, Z., Lü, Y., Zeng, Y., Li, Y., Jiang, X., 2016. Revegetation in China's loess plateau is approaching sustainable water resource limits. Nat. Clim. Chang. 6, 1019–1022.

Fixen, P., Brentrup, F., Bruulsema, T., Garcia, F., Norton, R., Zingore, S., 2014. Nutrient/fertilizer use efficiency: measurement, current situation and trends. In: Drechsel, P., Heffer, P., Magen, H., Mikkelsen, R., Wichelns, D. (Eds.), Managing Water and Fertilizer for Sustainable Agricultural Intensification. IFA, IWMI, IPNI and IPI, Paris, France.

Gan, J., Lu, Z., Cheung, A., Dai, M., Liang, L., Harrison, P.J., Zhao, X., 2014. Assessing ecosystem response to phosphorus and nitrogen limitation in the Pearl River plume using the Regional Ocean modeling system (ROMS). J. Geophys. Res. Oceans 9, 8858–8877.

Gao, S., Wang, Y.P., 2008. Changes in material fluxes from the Changjiang river and their implications on the adjoining continental shelf ecosystem. Cont. Shelf Res. 28 (12), 1490–1500.

Ge, J., Shi, S., Liu, J., Xu, Y., Chen, C., Bellerby, R., Ding, P., 2020. Interannual variabilities of nutrients and phytoplankton off the changjiang estuary in response to changing river inputs. J. Geophys. Res. Oceans 125, e2019JC015595.

Glibert, P.M., Burford, M.A.J.O., 2017. Globally changing nutrient loads and harmful algal blooms: recent advances, new paradigms, and continuing challenges. Oceanography 30, 58–69.

Grimvall, A., Stålnacke, P., Tonderski, A., 2000. Time scales of nutrient losses from land to sea – a European perspective. Ecological Engineering 14, 363–371.

Gu, B.J., Ge, Y., Chang, S.X., Luo, W.D., Chang, J., 2013. Nitrate in groundwater of China: sources and driving forces. Glob. Environ. Chang. 23, 1112–1121.

Guo, C., Bai, Z., Shi, X., Chen, X., Chadwick, D., Strokal, M., Zhang, F., Ma, L., Chen, X., 2021. Challenges and strategies for agricultural green development in the Yangtze River basin. J. Integr. Environ. Sci. 18, 37–54.

He, Y., Qi, Y.B., Tang, J.Q., 2019. Regional difference and ConvergenceAnalysis on fertilizer application efficiency of grain production in China (in Chinese). Jiangsu J. Agric. Sci. 35, 729–735 is10.3969/j.issn.1000-4440.2019.03.032.

Heffer, P., Prud'homme, M., 2016. Global nitrogen fertilizer demand and supply: trend, current level and outlook. International Nitrogen Initiative Conference. Melbourne, Australia.

Howarth, R.W., Boyer, E.W., Pabich, W.J., Galloway, J.N., 2002. Nitrogen use in the United States from 1961–2000 and potential future trends. AMBIO: a journal of the humanEnvironment 31, 88–96.

Jiang, Z., Liu, J., Chen, J., Chen, Q., Yan, X., Xuan, J., Zeng, J., 2014. Responses of summer phytoplankton community to drastic environmental changes in the Changjiang (Yangtze River) estuary during the past 50 years. Water Res. 54, 1–11.

Ju, X.T., Xing, G.X., Chen, X.P., Zhang, S.L., Zhang, L.J., Liu, X.J., Cui, Z.L., Yin, B., Christie, P., Zhu, Z.L., 2009. Reducing environmental risk by improving N management in intensive Chinese agricultural systems. Proceedings of the National Academy of Sciences 106, 3041–3046.

Justić, D., Rabalais, N.N., Turner, R.E., 1995. Stoichiometric nutrient balance and origin of coastal eutrophication. Mar. Pollut. Bull. 30, 41–46.

Kong, L., Zheng, H., Rao, E., Xiao, Y., Ouyang, Z., Li, C., 2018. Evaluating indirect and direct effects of eco-restoration policy on soil conservation service in Yangtze River basin. Sci. Total Environ. 631, 887–894.

Li, S., Bush, R.T., 2015. Rising flux of nutrients (C, N, P and Si) in the lower Mekong River. J. Hydrol. 530, 447–461.

Li, H., Tang, H., Shi, X., Zhang, C., Wang, X., 2014. Increased nutrient loads from the changjiang (Yangtze) river have led to increased harmful algal blooms. Harmful Algae 39, 92–101.

Lin, B., Liu, Z., Eglinton, T.I., Kandasamy, S., Blattmann, T.M., Haghipour, N., de Lange, G.J., 2019. Perspectives on provenance and alteration of suspended and sedimentary organic matter in the subtropical Pearl River system, South China. Geochim. Cosmochim. Acta 259, 270–287.

Liu, S.M., Zhang, J., Chen, H.T., Wu, Y., Xiong, H., Zhang, Z.F., 2003. Nutrients in the Changjiang and its tributaries. Biogeochemistry 62, 1–18.

Liu, X., Beusen, A.H.W., Van Beek, L.P.H., Mogollón, J.M., Ran, X., Bouwman, A.F., 2018. Exploring spatiotemporal changes of the Yangtze River (Changjiang) nitrogen and phosphorus sources, retention and export to the East China Sea and Yellow Sea. Water Res. 142, 246–255.

Liu, Y., Deng, B., Du, J., Zhang, G., Hou, L., 2019. Nutrient burial and environmental changes in the Yangtze Delta in response to recent river basin human activities. Environ. Pollut. 249, 225–235.

Liu, D., Bai, L., Li, X., Zhang, Y., Qiao, Q., Lu, Z., Liu, J., 2022. Spatial characteristics and driving forces of anthropogenic phosphorus emissions in the Yangtze River Economic BeltChina. Resour. Conserv. Recycl. 176, 105937.

Liu, J., Yao, Q., Mi, T., Wei, Q., Chen, H., Yu, Z., 2022. Change of the long-term nitrogen and phosphorus in the Changjiang (Yangtze) river estuary. Front. Mar. Sci. 9.

Loos, S., Middelkoop, H., van der Perk, M., van Beek, R., 2009. Large scale nutrient modelling using globally available datasets: a test for the Rhine basin. J. Hydrol. 369, 403–415.

Ludwig, W., Dumont, E., Meybeck, M., Heussner, S.J.P.I.O., 2009. River discharges of water and nutrients to the Mediterranean and Black Sea: major drivers for ecosystem changes during past and future decades? Prog. Oceanogr. 80, 199–217.

Ludwig, W., Bouwman, A.F., Dumont, E., Lespinas, F., 2010. Water and nutrient fluxes from major Mediterranean and Black Sea rivers: past and future trends and their implications for the basin-scale budgets. Glob. Biogeochem. Cycles 24.

Maavara, T., Akbarzadeh, Z., Van Cappellen, P., 2020a. Global dam-driven changes to riverine N:P:Si ratios delivered to the coastal ocean. Geophys. Res. Lett. 47, e2020GL088288.

Maavara, T., Chen, Q., Van Meter, K., Brown, L.E., Zhang, J., Ni, J., Zarfl, C., 2020b. River dam impacts on biogeochemical cycling. Nat. Rev. Earth Environ. 1, 103–116.

Malone, T.C., Newton, A., 2020. The globalization of cultural eutrophication in the coastal ocean: causes and consequences. Frontiers in marineScience 7.

Mayorga, E., Seitzinger, S.P., Harrison, J.A., Dumont, E., Beusen, A.H., Bouwman, A., Fekete, B.M., Kroeze, C., Van Drecht, G.J.E.M., 2010. Global nutrient export from WaterSheds 2 (NEWS 2): model development and implementation. Software 25, 837–853.

Mei, X., Zhang, M., Dai, Z., Wei, W., Li, W., 2019. Large addition of freshwater to the tidal reaches of the Yangtze (Changjiang) river. Estuar. Coasts 42, 629–640.

Metson, G.S., Lin, J., Harrison, J.A., Compton, J.E., 2020. Where have all the nutrients gone? Long-term decoupling of inputs and outputs in the Willamette River watershed, Oregon, United States. Journal of geophysical researchBiogeosciences 125 (10), e2020JG005792.

Ministry of Agriculture of the People's Republic of China, 2015. Plan of Actions Aiming for Zero Growth in Synthetic Fertilizer Use From 2020 Onwards. Available from http://www.moa.gov.cn/. (Accessed 1 August 2022).

Ministry of Ecology and Environment of the People's Republic of China, 2019. Phosphorus special regulation activities. Available from http://www.mee.gov.cn. (Accessed 1 August 2022).

Moutin, T., Raimbault, P., Golterman, H.L., Coste, B., 1998. The input of nutrients by the Rhône river into the Mediterranean Sea: recent observations and comparison with earlier data. Hydrobiologia 373, 237–246.

National People's Congress of the People's Republic of China, 2011. Increase sewage treatment investment. Available from http://www.npc.gov.cn/. (Accessed 1 August 2022).

National People's Congress of the People's Republic of China, 2016. Ecological rehabilitation and high-quality development of the Changjiang Basin. Available from http://www.npc.gov.cn/. (Accessed 1 August 2022).

Omara, P., Aula, L., Oyebiyi, F., Raun, W.R., 2019. World cereal nitrogen use efficiency trends: review and current knowledge. Agrosyst. Geosci. Environ. 2, 1–8.

Paczkowska, J., Brugel, S., Rowe, O., Lefébure, R., Brutemark, A., Andersson, A., 2020. Response of coastal phytoplankton to high inflows of terrestrial matter. Frontiers in marineScience 7.

Puckett, L.J., Tesoriero, A.J., Dubrovsky, N.M., 2011. Nitrogen contamination of surficial aquifers-a growing legacy. Environ. Sci. Technol. 45 (3), 839e844.

Rabalais, N.N., Turner, R.E., Díaz, R.J., Justić, D., 2009. Global change and eutrophication of coastal waters. ICES J. Mar. Sci. 66, 1528–1537.

Ran, X., Bouwman, L., Yu, Z., Beusen, A., Chen, H., Yao, Q., 2017. Nitrogen transport, transformation, and retention in the three gorges reservoir: a mass balance approach. Limnol. Oceanogr. 62, 2323–2337.

Ran, X., Bouwman, A.F., Yu, Z., Liu, J., 2019. Implications of eutrophication for biogeochemical processes in the three gorges reservoir, China. Reg. Environ. Chang. 19, 55–63.

Ran, X., Wu, W., Song, Z., Wang, H., Chen, H., Yao, Q., Xin, M., Liu, P., Yu, Z., 2022. Decadal change in dissolved silicate concentration and flux in the Changjiang (Yangtze) river. Sci. Total Environ. 839, 156266.

Reder, K., Bärlund, I., Voß, A., Kynast, E., Williams, R., Malve, O., Flörke, M., 2013. European scenario studies on future in-stream nutrient concentrations. Trans. ASABE 56, 1407–1417.

Redfield, A.C., Ketchum, B.H., Richards, F.A., 1963. The influence of organisms on the composition of sea-water. In: Hills, M.N. (Ed.), The Sea. Wiley and Sons, New York, pp. 12–37.

Romero, E., Garnier, J., Lassaletta, L., Garnier, J., Lassaletta, L., Billen, G., 2013. Large-scale patterns of river inputs in southwestern Europe: seasonal and interannual variations and potential eutrophication effects at the coastal zone. Biogeochemistry 113, 481–505.

Runkel, R.L., Crawford, C.G., Cohn, T.A., 2004. Load Estimator (LOADEST): a FORTRAN program for estimating constituent loads in streams and rivers. Techniques and Methods Book 4, Chapter A5.

Shapley, L., 1953. In: Artin, Emil, Morse, Marston (Eds.), Quota Solutions op n-Person Games1. 1953, p. 343.

Shen, Z.L., Liu, Q., 2009. Nutrients in the Changjiang River. Environ. Monit. Assess. 153, 27–44.

Shuler, C.K., El-Kadi, A.I., Dulai, H., Glenn, C.R., Fackrell, J., 2017. Source partitioning of anthropogenic groundwater nitrogen in a mixed-use landscape, Tutuila, American Samoa. Hydrogeol. J. 25 (8), 2419–2434.

Stackpoole, S., Sabo, R., Falcone, J., Sprague, L., 2021. Long-term Mississippi River trends expose shifts in the river load response to watershed nutrient balances between 1975 and 2017. Water Resour. Res. 57, e2021WR030318.

Strokal, M., Kroeze, C., Wang, M., Ma, L., 2017. Reducing future river export of nutrients to coastal waters of China in optimistic scenarios. Sci. Total Environ. 579, 517–528.

Tian, Z., Wang, L., Li, Y., Zheng, B., Chu, Z., 2021. Changes of phosphorus delivery from Yangtze River to dongting Lake under new water and sediment conditions. J. Clean. Prod. 316.

Tian, Q., Xu, K.H., Wang, Y.P., Liang, C.J., 2022. Dam impacts on seasonality of water and sediment transport in intensively managed river basins: a case study of the Yangtze River. J. Geophys. Res. Earth Surf. 127, e2021JF006.

Tong, C.L., Hall, C.A.S., Wang, H.Q., 2003. Land use change in Rice, wheat and maize production in China, (1961–1998). Agric. Ecosyst. Environ. 95, 523–536.

Van Meter, K.J., Basu, N.B., Veenstra, J.J., Burras, C.L., 2016. The nitrogen legacy: emerging evidence of nitrogen accumulation in anthropogenic landscapes. Environ. Res. Lett. 11 (3).

Van Meter, K.J., Van Cappellen, P., Basu, N.B., 2018. Legacy nitrogen may prevent achievement of water quality goals in the Gulf of Mexico. Science 360, 427–430.

Wakida, F.T., Lerner, D.N., 2005. Non-agricultural sources of groundwater nitrate: a review and case study. Water Res. 39, 3–16.

Wang, B.D., 2006. Cultural eutrophication in the Changjiang (Yangtze River) plume: history and perspective. Estuar. Coast. Shelf Sci. 69, 471–477.

Wang, B., Xu, Z., Wang, K., Li, J., Xu, T., Han, Y., Xu, Bin, Hao, X., Gao, S., 2019a. Correlation of GDP per capita and chemical fertilizer consumption of different countries from 1961 to 2015 (in Chinese). J. Agric. Resour. Environ. 36, 718.

Wang, M., Strokal, M., Burek, P., Kroeze, C., Ma, L., Janssen, A.B., 2019b. Excess nutrient loads to Lake taihu: opportunities for nutrient reduction. Sci. Total Environ. 664, 865–873.

Wang, H., Yan, H., Zhou, F., Li, B., Zhuang, W., Yang, Y., 2019c. Dynamics of nutrient export from the Yangtze River to the East China Sea. Estuar. Coast. Shelf Sci. 229, 106415.

Wang, H., Yan, H., Zhou, F., Li, B., Zhuang, W., Shen, Y., 2020. Changes in nutrient transport from the Yangtze River to the East China Sea linked to the three-gorges dam and water transfer project. Environ. Pollut. 256, 113376.

Wang, J., Bouwman, A.F., Liu, X., Beusen, A.H.W., Van Dingenen, R., Dentener, F., Yao, Y., Glibert, P.M., Ran, X., Yao, Q., Xu, B., Yu, R., Middelburg, J.J., Yu, Z., 2021a. Harmful algal blooms in Chinese coastal waters will persist due to perturbed nutrient ratios. Environ. Sci. Technol. Lett. 8, 276–284.

Wang, Y., Xu, H., Li, M., 2021b. Long-term changes in phytoplankton communities in China's Yangtze estuary driven by altered riverine fluxes and rising sea surface temperature. Geomorphology 376, 107566.

Wang, H., Ran, X., Bouwman, A.F., Wang, J., Xu, B., Song, Z., Sun, S., Yao, Q., Yu, Z., 2022. Damming alters the particulate organic carbon sources, burial, export and estuarine biogeochemistry of rivers. J. Hydrol. 607, 127525.

Wang, H., Bouwman, A.F., Van Gils, J., Vilmin, L., Beusen, A.H.W., Wang, J., Liu, X., Yu, Z., Ran, X., 2023. Hindcasting harmful algal bloom risk due to land-based nutrient pollution in the eastern Chinese coastal seas. Water Res. 2023, 119669.

Wu, W., Ran, X., Li, J., Wang, H., Li, M., Liu, J., Zang, J., 2019a. Sources, distributions, and fluxes of major and trace elements in the Yangtze River (in Chinese). Environ. Sci. 40, 4900–4913.

Wu, G., Cao, W., Wang, F., Su, X., Yan, Y., Guan, Q., 2019b. Riverine nutrient fluxes and environmental effects on China's estuaries. Sci. Total Environ. 661, 130–137.

Wu, N., Liu, S.M., Zhang, G.L., Zhang, H.M., 2021. Anthropogenic impacts on nutrient variability in the lower Yellow River. Sci. Total Environ. 755, 142488.

Wu, Z., Li, J., Sun, Y., Peñuelas, J., Huang, J., Sardans, J., Jiang, Q., Finlay, J.C., Britten, G.L., Follows, M.J., Gao, W., Qin, B., Ni, J., Huo, S., Liu, Y., 2022. Imbalance of global nutrient cycles exacerbated by the greater retention of phosphorus over nitrogen in lakes. Nat. Geosci. 15, 464–468.

Xiao, W., Liu, X., Irwin, A.J., Laws, E.A., Wang, L., Chen, B., Zeng, Y., Huang, B., 2018. Warming and eutrophication combine to restructure diatoms and dinoflagellates. Water Res. 128, 206–216.

Xiao, X., Agustí, S., Pan, Y., Yu, Y., Li, K., Wu, J., Duarte, C.M., 2019. Warming amplifies the frequency of harmful algal blooms with eutrophication in chinese coastal waters. Environ. Sci. Technol. 53, 13031–13041.

Xie, Y.X., Zhang, S.L., Zhao, X., Xiong, Z.Q., Xing, G.X., 2008. Seasonal variation pattern of NH4+-N/NO3–N ratio and δ15NH4+ value in rainwater in Yangtze River Delta (in Chinese). Chin. J. Appl. Ecol. 19.

Xin, J., Liu, Y., Chen, F., Duan, Y.J., Wei, G.L., Zheng, X.L., Li, M., 2019. The missing nitrogen pieces: a critical review on the distribution, transformation, and budget of nitrogen in the vadose zone-groundwater system. Water Res. 165, 114977.

Xu, H., Paerl, H.W., Qin, B., Zhu, G., Gaoa, G.J.L., oceanography, 2010. Nitrogen and phosphorus inputs control phytoplankton growth in eutrophic Lake Taihu, China. Limnol. Oceanogr. 55, 420–432.

Xu, H., Chen, Z., Finlayson, B., Webber, M., Wu, X., Li, M., Chen, J., Wei, T., Barnett, J., Wang, M., 2013. Assessing dissolved inorganic nitrogen flux in the Yangtze River, China: sources and scenarios. Glob. Planet. Change 106, 84–89.

Xu, Y., Wu, X., Lu, R., Yang, W., Zhao, Y., 2018. Total phosphorus pollution, countermeasures and suggestions of the Yangtze River Economic Belt (in Chinese). Environ. Conform. Assess. 1, 69–73.

Xu, L., Yang, D., Greenwood, J., Feng, X., Gao, G., Qi, J., Cui, X., Yin, B., 2020. Riverine and oceanic nutrients govern different algal bloom domain near the Changjiang estuary in summer. Journal of geophysical researchBiogeosciences 125, e2020JG005727.

Yan, X., Thieu, V., Garnier, J., 2021. Long-term assessment of nutrient budgets for the four reservoirs of the Seine Basin (France). Sci. Total Environ. 778, 146412.

Yang, H., Yang, S., Xu, K., Milliman, J., Wang, H., Yang, Z., Chen, Z., Zhang, C.J.G., Change, P., 2018. Human impacts on sediment in the Yangtze River: a review and new perspectives. Glob. Planet. Chang. 162, 8–17.

Yang, H., Zhu, Q., Liu, J., Zhang, Z., Yang, S., Shi, B., Zhang, W., Wang, Y., 2023. Historic changes in nutrient fluxes from the Yangtze River to the sea: recent response to catchment regulation and potential linkage to maritime red tides. J. Hydrol. 617, 129024.

Zhang, J., Du, Y.N., Zhang, G.S., Chang, Y., Zhou, Y.C., Zhang, Z.F., Wu, Y., Chen, J.W., Zhang, A.Y., Zhu, Z.Y., Liu, S.M., 2021. Increases in the seaward river flux of nutrients driven by human migration and land-use changes in the tide-influenced delta. Sci. Total Environ. 766, 14501.

Zhang, J., Guo, X., Zhao, L., 2021. Budget of riverine nitrogen over the East China Sea shelf. Environ. Pollut. 289, 117915.

Zheng, L.W., Zhai, W.D., 2021. Excess nitrogen in the bohai and yellow seas, China: distribution, trends, and source apportionment. Sci. Total Environ. 794, 148702.

Zheng, J., Cao, X., Ma, C., Weng, N., Huo, S., 2022. What drives the change of nitrogen and phosphorus loads in the Yellow River Basin during 2006–2017? J. Environ. Sci. 126, 17–28.