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Phosphorus – The main limiting factor in riverine ecosystems in China

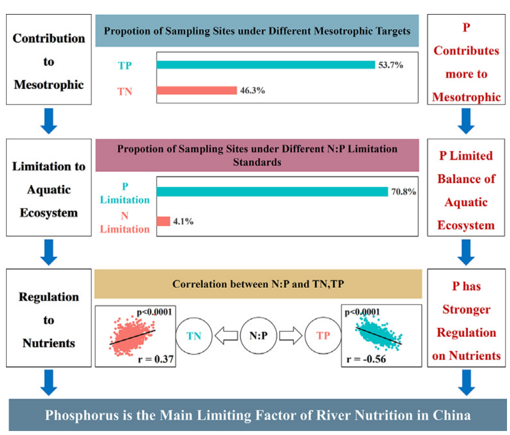
Yan Chen [a,b,1], Jie Chen [e,1], Rui Xia [a,c,d,*], Wenpan Li [i], Yuan Zhang [h], Kai Zhang [a,c,d], Shanlin Tong [e], Ruining Jia [a,f], Qiang Hu [a,c,d], Lu Wang [a,g], Xiaojiao Zhang [a,c,d]

[a] State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China
[b] Chongqing Institute of Green and Intelligent Technology, Chinese Academy of Sciences, Chongqing 400714, China
[c] National Engineering Laboratory for Lake Pollution Control and Ecological Restoration, Chinese Research Academy of Environmental Sciences, Beijing 100012, China
[d] State Environmental Protection Key Laboratory of Estuarine and Coastal Research, Chinese Research Academy of Environmental Sciences, Beijing 100012, China
[e] State Key Laboratory of Water Resource and Hydropower Engineering Science, Wuhan University, Wuhan 430072, China
[f] Northwest University, College of Urban and Environmental Sciences, Northwest University, Xi'an 710127, China
[g] College of Water Science, Beijing Normal University, Beijing 100875, China
[h] School of Ecology, Environment and Resources, Guangdong University of Technology, Guangdong 510006, China
[i] China National Environmental Monitoring Centre, Beijing 100012, China

HIGHLIGHTS

- Riverine nutrition in China showed "high in the north and low in the south".
- The patterns of riverine nutrient limitations were investigated in terms of contribution, limitation and regulation.
- Phosphorus is the main limiting factor in riverine ecosystems in China based on N:P stoichiometry.

GRAPHICAL ABSTRACT



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ABSTRACT

River receive substantial nutrient inputs, and serve as the main channel for nitrogen and phosphorus to enter the lake, their nutrient control is of great significance to the alleviation of lake eutrophication. While nutrient limitation affects the primary productivity of water ecosystems and the biodiversity of aquatic communities, identifying the limiting factors in riverine ecosystems across China remains elusive. Here, we explore which nutrients have a stronger effect on nutritional balance and aquatic ecosystems in China's rivers based on the total nitrogen (TN) and total phosphorus (TP) observations from 1412 sampling sites in 2018. This study supports the following three main conclusions. Though the percentages of the sites with TN or TP exceeding the limits varied as per different mesotrophic targets, and TP (53.7 %) contributed more to nutrient enrichment than TN (46.3 %). In addition, the spatial distribution characteristics of river nutrients were high in the north (arid zone) and low in the south (humid zone) in China. According to four classification criteria of N:P ratio, 70.8 % of the sampling sites were attributed to phosphorus limiting, much higher than the sites with nitrogen limiting (4.1 %). TN and TP have a synergistic effect on river nutrients, while TP has a stronger regulation framework. Our results reveal that the nutrients in China's rivers are mainly phosphorus limiting,

* Corresponding author at: State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China.
E-mail address: xiarui@craes.org.cn (R. Xia).

[1] Co-first author.

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which implies that phosphorus-oriented best management practices are more likely to maintain the nutrient balance of rivers towards healthy aquatic ecosystems.
Synopsis: Phosphorus is the key factor that affecting the stability and nutrient balance of riverine ecosystem.

1. Introduction

Human-induced nutrient flow (e.g., nitrogen (N) and phosphorus (P)) has become an important part of the biogeochemical cycles in terrestrial ecosystems (Elser and Bennett, 2011; Tong et al., 2018; Peñuelas et al., 2012, 2013; Bouwman et al., 2013; Tong et al., 2015). N and P are usually considered as main drivers of phytoplankton growth (Conley et al., 2009). However, the excessive input of N and P from anthropogenic sources causes massive eutrophication and increases harmful algal blooms (HABs) in freshwater systems worldwide (Ho et al., 2019; Vollenweider, 1992; Carpenter et al., 1998), imposing challenge for sustainable nutrient management (European Environment Agency, 2010; Leaf, 2018). The coupling between N and P can affect the trophic webs and biogeochemical cycles of aquatic ecosystems (Yan et al., 2016) and is regulated by environmental factors and aquatic organisms (Sereda and Hudson, 2011). High N/P ratios alter benthic functional feeding patterns by mediating primary productivity. Since N and P are essential nutrients for primary agricultural producers, alterations in their input into the environment would affect the structure and functioning of ecosystems (Smith et al., 1999).

Absolute concentrations of a limiting nutrient play a central role in ecosystem dynamics, but the relative availability of some nutrients, such as N:P, also affect a range of ecological patterns and processes (Sterner and Elser, 2002). Nutrient co-limitation by N and P is pervasive across a range of terrestrial, aquatic, and marine ecosystems (Elser et al., 2007; Harpole et al., 2011; Paerl et al., 2016). As a result, reducing the input of one single nutrient into the water body does not always yield the desired outcome of alleviating eutrophication (Elser et al., 2007; Dodds, 2006). The N:P reveals the potential limiting nutrients and also helps to regulate the structure, function, and processes of ecosystems (Sterner and Elser, 2002; Redfield, 1958), typically, the N:P is used to determine the limiting nutrient for water eutrophication and phytoplankton growth (Conley et al., 2009). The growth of the global demand for P in a world flooded with reactive nitrogen (N) and carbon dioxide (CO_2) emitted by human activities is faster than the growth of supply (Peñuelas et al., 2012; Galloway et al., 2004; Mahowald et al., 2008). Anthropogenic inputs of N and P alter the nutrient limitation balance and affect the functioning of aquatic ecosystems (Loladze and Elser, 2011). For example, nitrogen deposition alleviates nitrogen constraints in most terrestrial ecosystems, stimulating biomass production and carbon sequestration (Schulte-Uebbing et al., 2021). However, the increase of the N:P caused by excess N input aggravates eutrophication of water bodies, change ecological structures and functions, and even deteriorate aquatic ecosystems (Lewis et al., 2011). The N:P has been found to be significantly related to the plant community composition and diversity (Sasaki et al., 2010). Moreover, the relationship between the N:P and species diversity[28] is not necessarily linear. Several important ecosystem processes, such as energy and element transfers through trophic levels, are also affected by these change in N:P (Moe et al., 2005). Exceedances of the optimal N:P can reduce the growth rates of primary producer (Sterner and Elser, 2002) and change the functional feeding types of benthic animals, thereby directly affecting the eutrophication status of a water body. Maintaining appropriate ratios of N:P is thus key to regulating the nutrient balance of aquatic ecosystem.

Investigations on whether P and/or N limit the productivity and growth of algae in lakes over a long time period have concluded that phosphorus dominates primary productivity (Sterner and Elser, 2002). The P-reduction paradigm has been effective in many lake restoration initiatives (Schindler et al., 2008; Schindler and Hecky, 2009). However, there is no consensus on whether the same effect exists in rivers. Understanding N:P and algal nutrient limitation provides insights into the nutrient management strategies applied to reduce eutrophication and prevent HABs (Andersen et al., 2020). While many studies have investigated nitrogen and phosphorus as nutrient limits in lakes, very little work has been done on nitrogen and phosphorus nutrient limits in rivers. Currently, there are three methods for determining nutrient limits in water bodies: Nottle test, nutrient-diffusing substrates (NDSs) and the N:P. Nottle tests of algal productivity, sometimes called biostimulation assays or bioassays have long been used for measuring the nutrient limitation of water bodies (JosephC et al., 1975; Lee et al., 2012). Using the level of NDSs as a means to identify nutrient limitation in stream periphyton communities (Hauer and Lamberti, 2017), to determine nutrient limitation in lakes, streams, and large rivers has been the practice for approximately the past years (Hauer and Lamberti, 2017; Capps et al., 2011; Francoeur et al., 1999). One of the oldest methods, the N:P has been widely used to evaluate the nutrient limitation of water bodies, including Redfield's N:P (Redfield, 1958) and the global thresholds of algal nutrient limitation (Guildford and Hecky, 2000).

Compared to lake eutrophication, rivers and estuaries eutrophication is less well understood (Ibáñez and Peñuelas, 2019). While several studies have shown that static water bodies such as lakes are primarily phosphorus-limited (Schindler et al., 2008; Schindler and Hecky, 2009; Qin et al., 2020; Schindler et al., 2016), it is not yet clear if this finding can be confirmed in flowing water bodies (Dodds, 2017). Concerns about stream eutrophication have increased as the United States and other countries have started to adopt nutrient control in stream management (Dodds and Welch, 2000). Rivers and streams receive substantial nutrient inputs, and they serve as the main channel for nitrogen and phosphorus to enter the lakes. Controlling their nutrient level is therefore crucial to alleviate lake eutrophication. River eutrophication has also led to algal blooms. The inflow of a large quantity of domestic and industrial wastewater, as well as agricultural non-point source pollution, combine to increase the amount of rich nutrients (nitrogen, phosphorus, potassium, silicon, etc.), which leads to a large increase of algae (diatom, cyanobacteria, green algae, etc.) and lakes. These blooms form a thick algal film on the surface of rivers and lakes; films that have become an extreme representation of a water body's eutrophication state. As the hydrological regimes of rivers change more dramatically than those of lake and reservoirs, they are not generally prone to algal blooms, especially since rivers have normally stronger self-purification abilities. However, once algal bloom occurs in a large river, the eutrophication may have a larger scale and more severe consequences (Xia et al., 2020, 2019; Cheng et al., 2019). For instance, in 2008, >400 km of algal bloom broke out in the middle and lower reaches of the Han River in China (Xia et al., 2020), which led to a drinking water shortage for >200,000 people. Several public water plants around the Han River stopped using it as their source due to the blockage of water intake, resulting in major social impacts.

Accordingly, in this study, we explore the indication and regulation of TN and TP on the nutrient excess and nutritional balance of rivers in China based on the paired N and P monitoring data from 1412 sites in 2018. More importantly, we identify the local nutrient limitation levels according to the widely used classification standards of nitrogen and phosphorus limitation. In general, we expect to clarify which nutrients can help aquatic ecosystems develop in a more balanced direction, so as to alleviate river eutrophication. The aim of this study is to discuss the nutrient limitation status of rivers and to provide a basis for the eutrophication control of static water bodies such as lakes, reservoirs.

2. Methods and materials

2.1. Data collection

Monthly paired TN and TP concentrations from 1412 sampling sites across China's rivers (excluding lakes, reservoirs, etc.) in the year of 2018 were obtained from China National Environmental Monitoring Centre (https://szzdjc.cnemc.cn:8070/GJZ/Business/Publish/Main.html) (Fig. 1). The data for analysis in this study were mainly annual averages using monthly data. Consistent sampling sites selection and procedure were adopted as per the standard of "Technical Specifications Requirements for Monitoring of Surface Water and Waste Water in China (HJ/T 91-2002)".

2.2. Statistical analysis

The main methods used in this paper are data statistics and linear regression. We used three complementary approaches to identify the potential nutritional limitations of river. Firstly, based on national-scale river TN and TP concentrations, the proportion of sampling sites with TN or TP exceeding the mesotrophic target was categorized to initially determine which nutrient contributed more to river nutrition. Then, we chose three benchmark values for TN and TP that met the mesotrophic target (MT) in these rivers, which are the Dodd's mesotrophic target (MT1: $0.7\ mg{\cdot}L^{-1} < TN < 1.5\ mg{\cdot}L^{-1}$; $0.025\ mg{\cdot}L^{-1} < TP < 0.075\ mg{\cdot}L^{-1}$) (Dodds et al., 1998), UK's mesotrophic target (MT2: $TN > 1.5\ mg{\cdot}L^{-1}$; $TP > 0.02\ mg{\cdot}L^{-1}$), and Norway's mesotrophic target (MT3: $TN > 0.6\ mg{\cdot}L^{-1}$; $TP > 0.02\ mg{\cdot}L^{-1}$) (Tong et al., 2018). According to these nitrogen and phosphorus limitation criteria, we then determined whether the riverine nutrients were nitrogen- or phosphorus-limiting. Lastly, a Pearson's correlation and reduced major axis (RMA) regression were used to characterize the relationships among TN, TP and the N:P. Therefore, the interaction and feedback effects of TN and TP were determined by a linear regression analysis for TN and TP. The response relationship between TN and the N:P, and between TP and the N:P could then be established to determine which nutrient has a stronger effect on nitrogen-phosphorus limitation. Log-transformed data were employed for the above analyses were performed.

2.3. Definition of nutrient limitation

Freshwater phytoplankton assemblages are sensitive to changes in the N:P. The variation of N inputs is likely to shift the composition of phytoplankton communities (Peñuelas et al., 2013). The classification of nutrient limitation has led to complex results. According to the thresholds derived from global patterns of phytoplankton stoichiometry, potential N-only limitation is indicated when the N:P < 9.0, N—P co-limitation occurs when $9 \leq$ N:P < 22.6, and P-only limitation occurs when the N:P ≥ 22.6 (Guildford and Hecky, 2000). In freshwater phytoplankton, an N:P > 20 is considered to be P-limited, while a ratio < 10 is considered to be N-limited (Schanz and Juon, 1983). Wu et al. (2015) proposed that N:P with N + P co-limited ranged from 12 to 30, while N:P for N-only limitation was <13, and P-only limitation occurs when the N:P ≥ 30. The 'Redfield (N:P = 16)' theory has also been widely used. When the N:P is higher

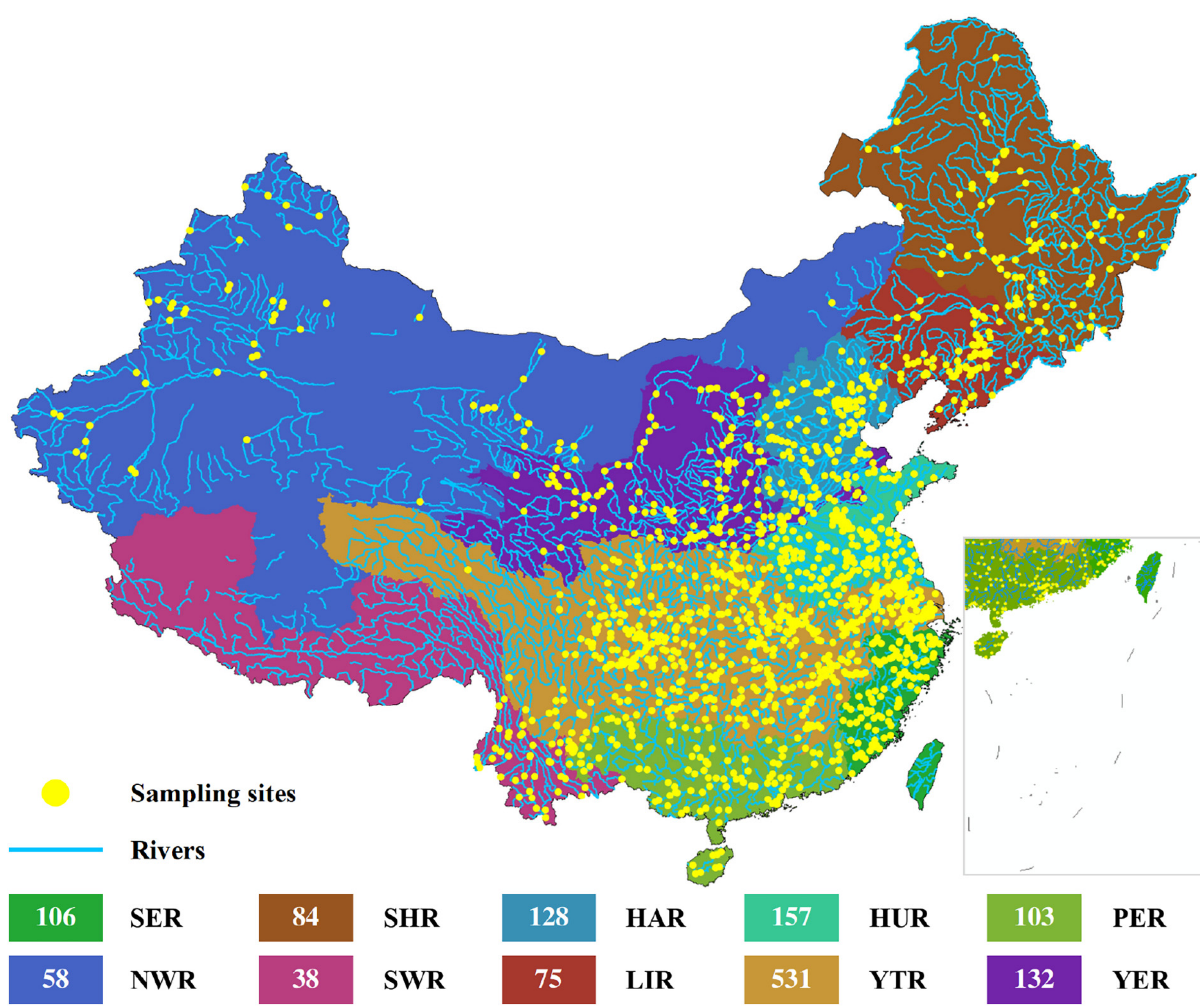

Fig. 1. The 1412 sampling sites in 10 river basins of China. The numbers in each colored box in the legend indicate the total number of sampling sites within a watershed (SER: South East River; SHR: Songhua River; HAR: Hai River; HUR: Huai River; PER; the Pearl River; NWR: North West River; SWR: South West River; LIR: Liao River; YTR: Yangtze River; YER: Yellow River). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

than the 'Redfield' ratio, algae are considered to be limited by phosphorus; and when it is lower than the 'Redfield' ratio, algae are considered to be N-limited (Redfield, 1958). In this study, we used the follow four standard thresholds to determine the nutritional limitation for China's rivers (Table 1).

3. Results

3.1. River nutrients and stoichiometry

Fig. 2 presents the distribution of TN concentrations (Fig. 2a), TP concentrations (Fig. 2b), and N:P (Fig. 2c) for the selected river sampling sites in 2018. The statistics and frequency distributions of the above three variables are shown in Table S1. From a national perspective, the average concentrations of TN and TP are 3.12 and 0.106 mg·L^{-1} respectively (Table S1). From a watershed perspective, the highest TN concentration was observed in the LIR with an average of 5.45 mg·L^{-1}, while the lowest TN concentrations occurred in that of the SWR (mean = 1.50 mg·L^{-1}). The highest TP concentrations was observed in the LIR (mean = 0.205 mg·L^{-1}), while the lowest TP concentrations occurred in the NWR (mean = 5.45 mg·L^{-1}). In terms of the spatial distributions, higher TN and TP concentrations were most often located in the HAR, HUR, LIR, and YER watersheds, while the lower TN and TP concentrations generally occurred in the SWR, NWR, and PER watersheds (Fig. 2a, b). The lowest N:P is in the SWR with an average of 27.2 (50 % CI = 16.9–35.5), which is much lower than the other river basins (HAR: mean = 133.7, LIR: mean = 106.7, YER: mean = 97.2) (Fig. 2c, Table S1). Consistent with the spatial distribution characteristics of TN and TP concentrations, high values of N:P were found in Northern China, including the YER, HUR, LIR and HAR watersheds, which are mainly characterized by low rainfall, dry climate and relatively scarce water resources.

Based on three different mesotrophic targets (MT1 to MT3), the proportion of sampling sites exceeding mesotrophic targets in rivers judged by TP was always higher than that judged by TN (Fig. 3). According to MT1, 23.4 % of TN and 38.5 % of TP sites exceed the mesotrophic target in all sampling sites. Among the 10 river basins, only the LIR, SWR and YER showed higher proportions judged by TN than by TP. With the MT2, the TN in 73.3 % and TP in 94.2 %, respectively, of the sampling sites exceed the mesotrophic target. The proportion of sampling sites in the 10 river basins that exceeded the mesotrophic target according to the TP judgment criteria was 90 %, while only 10 % of the sites exceeded their MT2 targets when judged by TN. Compared to MT1 and MT2, 98.1 % of TN and 94.2 % of TP sites exceeded the mesotrophic target under MT3. Although the proportion of sampling sites exceeding the mesotrophic target judged by TN is slightly higher than that judged by TP, the difference is not significant. Overall, according to the average proportions of sampling sites where TN and TP exceed the three mesotrophic targets, about 46.3 % of the river sampling sites consider TN as the main indicator of nutrient contribution, and about 53.7 % of the sites consider TP as the main indicator of nutrient contribution. Therefore, in China, TP pollution is found to be more serious than TN in river environments.

3.2. Nutrient limitation

Fig. 4 is the scatter plot of TN and TP concentration from all the samples collected across China in 2018, with the X-axis indicating TN concentration and the Y-axis showing the TP concentration. Both the X- and Y-axes are in logarithmic scale. The scatter plot is also divided by multiple diagonal lines according to the four different standards (Table 1). The scatter points within the upper-left region above the diagonal lines indicate P is the limiting nutrient at certain sampling sites, whereas the scatter points in the lower-right region below the diagonal lines show that N is the limiting nutrient at certain sampling sites. The points located in-between different diagonal lines indicate the sampling sites that are N + P co-limited.

According to Fig. 4, the TN and TP concentrations are distributed in a wide range across all sampling sites. Fig. 4 also shows that more sampling sites are P-limited rather than N-limited or N—P co-limited. If adopting the S I standard, 89.9 % of the sampling sites are potentially P-limited, and 10.1 % are potentially N-limited. If adopting the S II standard, 79 % of the sampling sites are potentially P-limited, and 21 % are potentially N-limited or N + P co-limited. Under the S III standard, 71.3 % of all sampling sites showed potential P limitation, and 28.7 % indicated potential N and N + P co-limited. In contrast, with the S IV standard, 55.5 % of sampling sites are potentially P-limited and 3.4 % of sampling sites are N-limited. Overall, under the four standards, the average proportions of sampling sites in the P-limited and N-limited sites are 70.8 % and 4.1 %, respectively, indicating that P limitation predominated across a diverse range of rivers over different basins in China.

Table 1
Different criteria to determine the nutritional limitation.

Standards	P Limited	N + P co-Limited	N Limited	Reference
S I	N:P ≥ 16		N:P ≤ 16	Redfield, 1958
S II	N:P > 20	10 ≤ N:P < 20	N:P < 10	Schanz and Juon, 1983
S III	N:P ≥ 22.6	9 ≤ N:P < 22.6	N:P < 9.0	Guildford and Hecky, 2000
S IV	N:P > 30	12 ≤ N:P < 30	N:P < 20	Wu et al., 2015

3.3. Relationship among TN, TP and N:P

Fig. 5 shows the correlation between TN and TP concentrations (log10 transformed) in ten different river basins, revealing that for all river basins, TN concentrations and TP concentrations are strongly and positively correlated nationally (R = 0.65, $p < 0.0001$, n = 1412). Such trends are consistent at the sample sites within different watersheds, where the correlation between TN and TP concentrations ranges from 0.63 to 0.79, except for three watersheds: NWR, HAR, and LIR. The correlation between TN and TP concentrations at those three watersheds are relatively lower compared to the rest, at 0.4, 0.46, and 0.56, respectively.

Fig. 6 shows the correlations between TN concentrations and N:P, and TP concentrations and N:P in China. For detailed information about the correlations s in each river basins please see Fig. S1 in the supporting materials. According to Fig. 6, in China overall, the N:P is positively correlated with TN concentration (R = 0.366, $p < 0.0001$), and negatively correlated with TP concentration (R = −0.56, $p < 0.0001$). This behavior is also consistent when examining different watersheds. The only exception is in the SWR, where the N:P was negatively correlated with both TN (R = −0.19, p = 0.25) and TP (R = −0.63, $p < 0.05$) concentrations. There is a positive correlation of TN with TN:TP and a negative correlation of TP with TN:TP based on simple mathematical rules. However, our aim is not to discover this inevitable phenomenon. It is more important to compare the correlation between TN & N:P and TP & N:P. According to Fig. 6 and Fig. S1, the correlation between N:P and TP (R^2 = 0.32) is always stronger than that between N:P and TN (R^2 = 0.13). In short, our analysis shows that TN and TP have a strong positive correlation, and that the correlation between N:P and TP is stronger than that between N:P and TN.

4. Discussion

4.1. Spatial distribution of nutrients in China

According to our results, the concentrations of TN and TP, as well as the N:P are generally higher in northern China, especially in the HAR, HUR, YER and LIR, than in southern China, which corresponds to the nutrient concentrations being higher in dry areas than that in wet areas. We suspect the difference between concentrations and N:P in northern and southern China are likely due to multiple factors, including different hydrological regimes (Strokal et al., 2016; Yi et al., 2017; Vilmin et al., 2018), watershed landscape features and anthropogenic activities, such as land use (Ding et al., 2011; Xiao et al., 2016) or wastewater treatment plants (WWTPs) (Singh et al., 2019). Natural factors such as elevation, slope and temperature have been found to contribute more to nutrient concentrations in

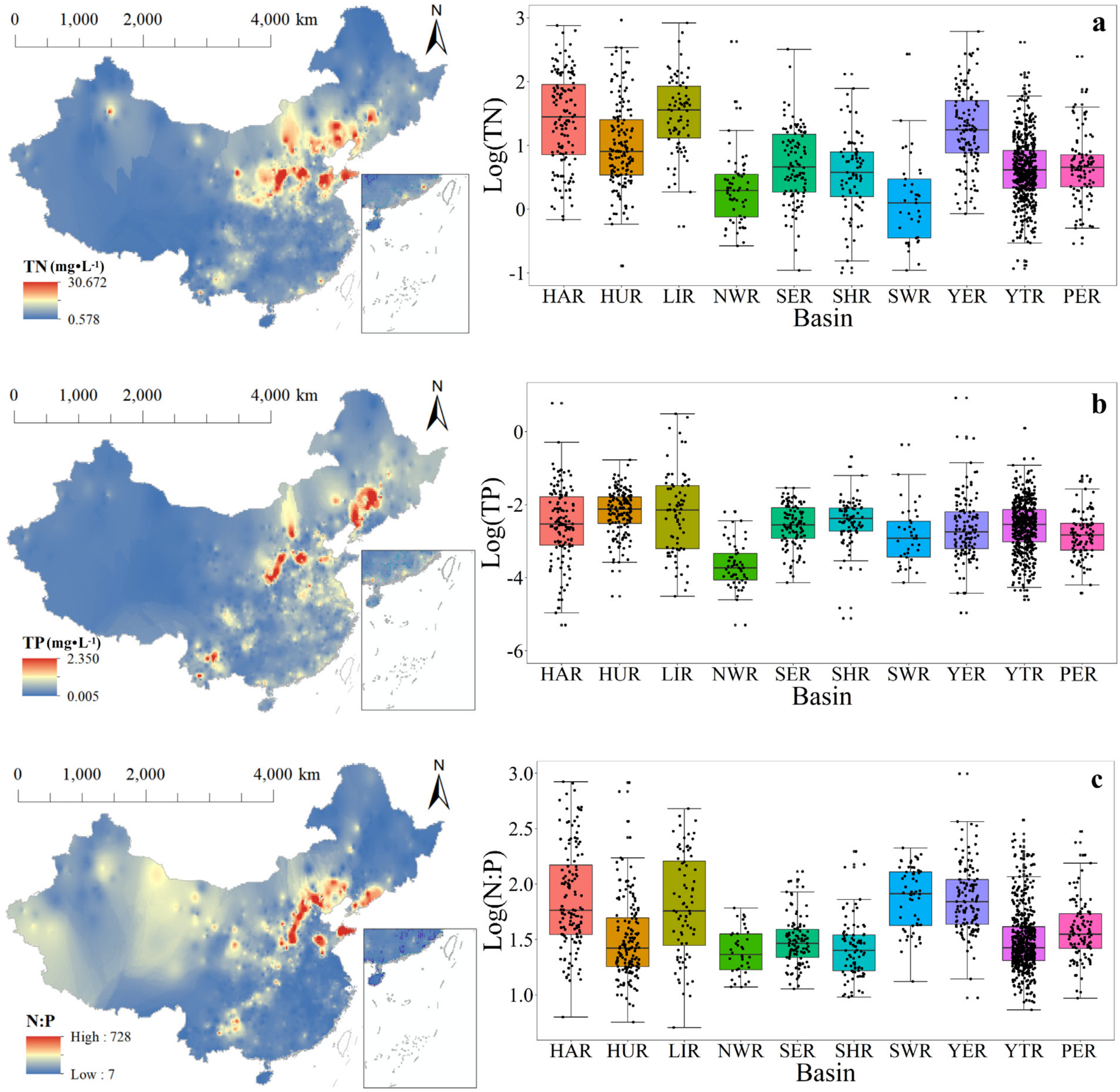

Fig. 2. TN concentrations (a), TP concentrations (b) and N:P (c) from the sampling sites (the left three figures are the distribution of TN, TP and N:P at national scale; the right three figures are the distribution TN, TP and N:P at watershed scale.)

China's rivers (Huang et al., 2021). Human activities, such as agricultural cultivation, locks, channeling and dam construction also play an important role in water environments. For example, YER is one of the most important agricultural regions in China and accounts for 8 % of the country's total grain yield (Chen et al., 2005). Intensive agricultural irrigation in watersheds where recharged water re-enters the river and circulates between the river and tilled land could significantly increase the major ion concentrations and contribute to elevated nutrient levels (Chen et al., 2006; Cai et al., 2008; Fan et al., 2014). However, one limitation of our study is its lack of input nutrient volume data for the different watersheds. Thus, further attribution analysis will be performed in future studies.

4.2. Contribution of phosphorus to mesotrophic rivers

China's rivers have been subjected to profound water quality impairments induced by the rapid and energy-intensive economic development over the past several decades (Chen et al., 2019). Rivers are less prone to nutrient enrichment than lakes, due to their short hydraulic residence time, high flow velocity and shear force, while large rivers have different degrees of water bloom outbreaks, such as the Rhine (Germany), the Darling (Australia), the Nakdong (Korea), and the Han, Pearl, Xiangxi, and Daning Rivers in China (Bowes et al., 2012; Kaspersen et al., 2016; Yang et al., 2017). Among China's 10 river basins, the HAR and HUR had the poorest water quality in 2016–2018, with TP and NH_4^+-N are collectively responsible for >85 % of the identified incidences of impaired conditions (Huang et al., 2021). TP became the main pollution factor in the YTR since 2016 (Qin et al., 2018). Moreover, not all rivers are eutrophic, but many rivers are at risk of eutrophication, and so different mesotrophic targets are used to reflect the trend of nutrient enrichment in rivers. Our results show that the main contributor to river eutrophication in China is TP rather than TN. Given that P impacts the primary productivity of many aquatic ecosystems, increased riverine P fluxes have been identified as a main cause of eutrophication over surface water bodies, including lakes and coastal marine environments (Conley et al., 2009; Correll, 1998; Schindler, 1977), especially for the proliferation of nuisance phytoplankton, epiphytic and benthic algae (Mainstone and Parr, 2002; Jarvie et al., 2004). Excessive P input is considered to be one of the greatest threats to river ecology worldwide (Sinha et al., 2017); it can cause serious

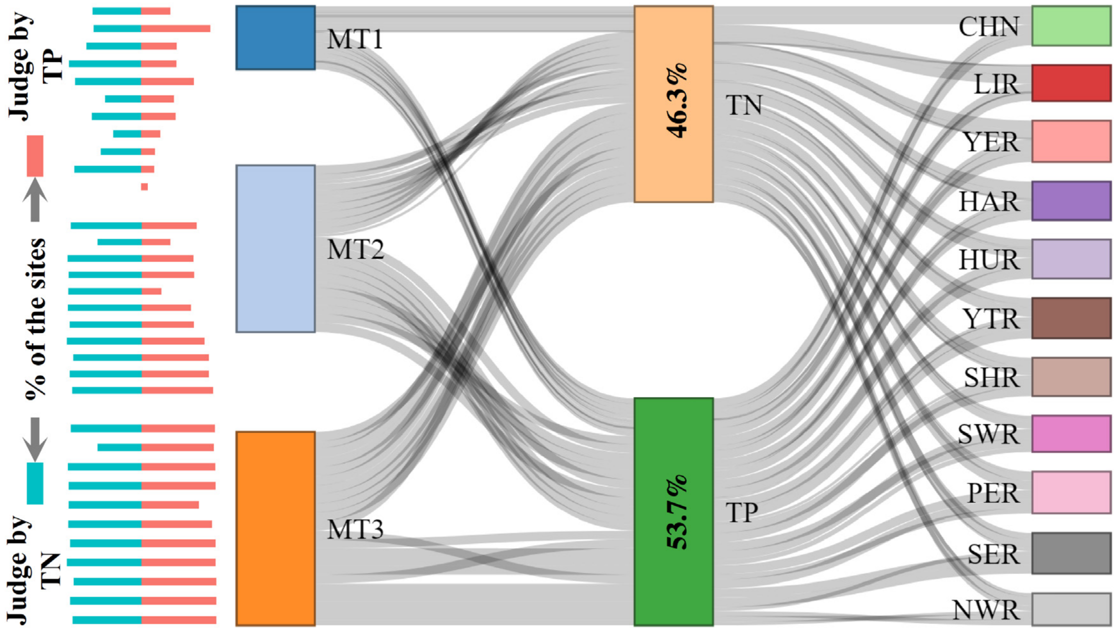

Fig. 3. The proportion of sampling sites with TN or TP exceeding mesotrophic targets in ten different river basins and in China overall (MT: mesotrophic targets; The bars show the percentage of sampling sites where TN or TP reached different mesotrophic targets, Light blue represents total nitrogen, Pink represents TP;The Sankey diagram represents the proportional distribution of TN and TP under different mesotrophic targets). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

problems for different water uses (Boeykens et al., 2017), and lead to eutrophication to the river system and endanger human health. Therefore, mitigating P inputs from wastewater (point) and agricultural (non-point) sources has been adopted as the main watershed management method to control freshwater eutrophication (Daniel et al., 1994; Sharpley et al., 1994).

4.3. Phosphorus limitation to aquatic ecosystems

Studies have shown that, whether at the scale of the ten major river basins or at the national scale, river N:P is generally dominated by phosphorus limitation, which is consistent with the finding that phosphorus is the most common limiting factor in freshwater systems (Tong et al., 2015). The nutrient status in aquatic ecosystems exhibits less nitrogen limitation; the relevant explanations are based on three aspects. Firstly, from the order of magnitude of the two elements, TN levels are several orders of magnitude higher than TP, objectively leading to a generally high N:P (Chen et al., 2018), thereby meeting the phosphorus limitation criteria. Secondly, atmospheric deposition has been revealed as an important contributor to ecosystem nutrient demand, which imposes a considerable burden on the environment (Wen et al., 2022). Recent studies have shown that annual deposition of TN and TP is 27.5 kg/ha and 0.92 kg/ha, respectively, so that TN deposition is about 30 times than that of TP deposition. Nitrogen deposition may therefore alleviate nitrogen limitation in most terrestrial or aquatic ecosystems, thereby stimulating biomass production and carbon sequestration (Schulte-Uebbing et al., 2021). Moreover, excessive nitrogen input is thought to exacerbate eutrophication, alter ecological structure and function, and deteriorate water ecosystems (Lewis et al., 2011). Conversely, trophic status reverts from phosphorus deficiency to nitrogen limitation due to increased atmospheric phosphorus loading (Camarero and Catalan, 2012). This last explanation revolves around predation relationships, where planktonic algae are selectively preyed upon by zooplankton as primary

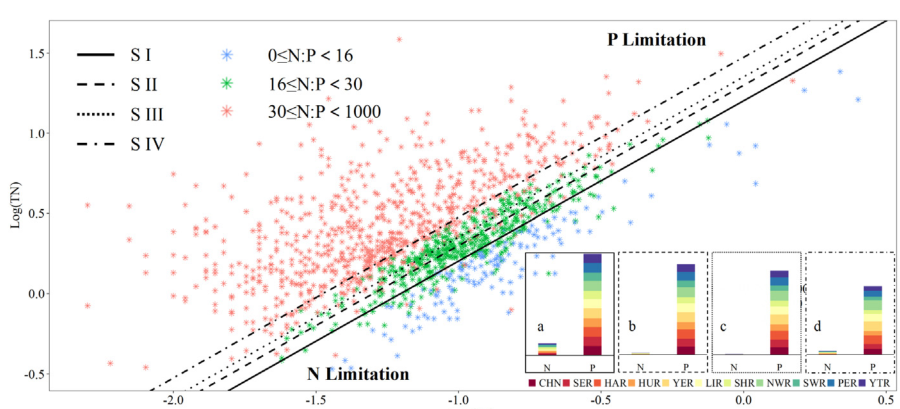

Fig. 4. TN and TP concentrations ($\log_{10}$ transformed) from 1412 sampling sites (S I ~ S IV represents different criteria of nutritional limitation; the scatter shows the distribution of N:P under different criteria, the scatter on the top left of the line means P limitation, the scatter on the bottom right of the line means N limitation; a ~ d indicates the propotion of N limitation and P limitation under different criteria at each basin.)

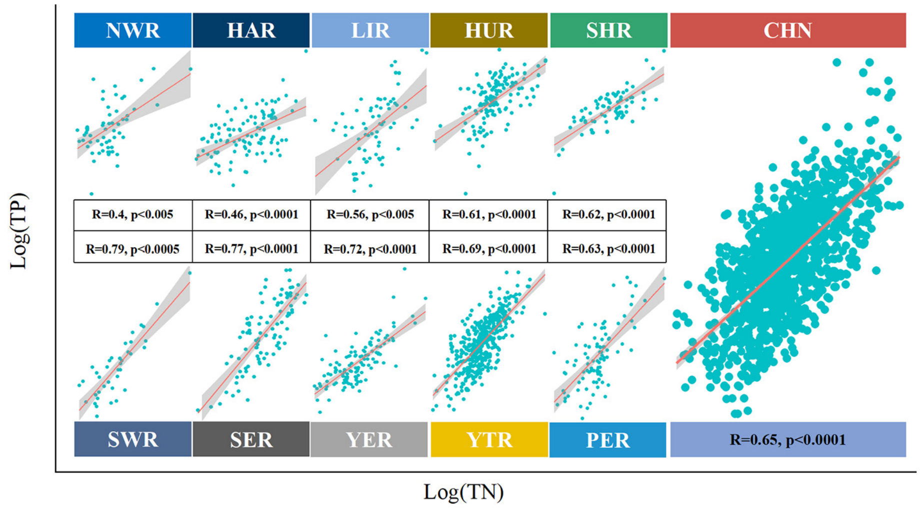

Fig. 5. Correlation analysis between TN and TP concentrations in different river basins.

producers. However, related studies have shown that phosphorus-limited algae are low-quality food for zooplankton due to their unsuitable biochemical composition or low phosphorus content, leading to a high consumption of phosphorus in the aquatic ecosystem by phosphorus-based nutrients, resulting in an elevated N:P and enhanced phosphorus limitation (Peñuelas et al., 2013). In terms of nitrogen and phosphorus inputs at the national scale, about 28 and 3 Tg of TN and TP, respectively, were input to rivers each year in China; TN inputs thus being as much as 5 times higher than those of TP (Chen et al., 2019), leading to an increase in river N:P. Human activities therefore, provide an important contribution to the global N:P of freshwater ecosystems (Beusen et al., 2016; Seitzinger et al., 2010).

4.4. Regulation mechanism of TP to nutrients

The results of our correlation analysis for nitrogen and phosphorus nutrients in Chinese rivers show a strong positive correlation between TN and TP, indicating similar input pathways for nitrogen and phosphorus. This is likely to the degree of nitrogen and phosphorus limitation is strongly influenced by the change of natural factors, because the degree of nitrogen or phosphorus limitation in rivers usually varies with anthropogenic inputs in the absence of proportional increases in nitrogen and phosphorus (Dodds and Smith, 2017). Correlations between factors such as chlorophyll or algal density and TN and TP are usually used to reveal the regulatory role of nitrogen and phosphorus on nutrient balance (Smith, 1983). By establishing the response relationship between chlorophyll *a*nd TN (and TP), related studies have revealed that both TP and TN show positive correlations with chlorophyll, but TP is more strongly correlated with chlorophyll (Lianga et al., 2020). Some other studies found that algal biomass (chlorophyll concentration) and algal growth rate (nutrient status index) were significantly controlled by phosphorus in freshwater systems, and even proposed a phosphorus control threshold (Guildford and Hecky, 2000). The N:P largely characterizes the proportional relationship between the water body receiving nitrogen and phosphorus input loads, and also reflects the impact of nutrient inputs on a water body's nutrient balance and the structure of its aquatic ecosystem (McCauley et al., 1998). Therefore, in our study, linear regressions of N:P with TN and TP were performed separately in our study. Interestingly, the TP and N:P showed a strong negative correlation nationally and for all ten major watersheds. This result directly demonstrates that in Chinese rivers, both TN and TP have regulatory effects on nutrient balance and aquatic ecosystem structure, but TP has a stronger

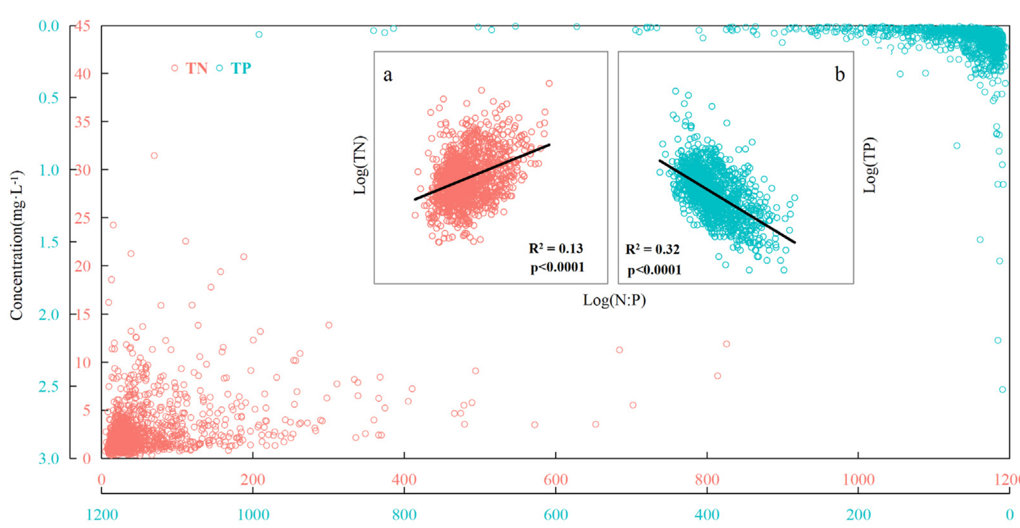

Fig. 6. Correlation analysis between TN and N:P, TP and N:P (the scatter shows the distribution of TN and TP with variation of N:P; a is the correlation between TN and N:P with log-transformed, b is the correlation between TP and N:P with log-transformed).

regulatory effect. That is, fluctuations in TP are more likely to cause changes in river nutrient balance, alter nutrient limitation, and perturb the structure of the aquatic ecosystem (Stackpoole et al., 2019; Jarvie et al., 2006).

5. Conclusions

While nutrient limitation affects the primary productivity of water ecosystems and the biodiversity of aquatic communities, identifying the limiting factors in riverine ecosystems across China remains elusive. Therefore, We explored whether river nutrition and aquatic ecosystem balance were mainly limited by phosphorus or nitrogen based on N:P stoichiometry in China. Our results revealed higher nutrient levels in northern (arid zone) rivers than in southern (humid zone) rivers. In addition, TP contributes more to river mesotrophic levels, as TP (53.7 %) caused higher nutrient enrichment in China's rivers than did TN (46.3 %). A more remarkable phenomenon is that most rivers are distributed in phosphorus-limited areas (70.8 %), with the least distribution in nitrogen-limited areas (4.1 %). More obviously, TN and TP have synergistic effects in rivers, but the correlation between N:P and TP concentrations is always stronger than that between N:P and TN. Therefore, we suggest that regulating phosphorus is more effective than regulating nitrogen to maintain the stability of river nutrient balance and aquatic ecosystem structure.

CRediT authorship contribution statement

Yan Chen: Resources, Data curation, Formal analysis, Methodology, Writing – original draft. **Jie Chen:** Resources, Methodology, Formal analysis, Writing – original draft. **Rui Xia:** Resources, Methodology, Formal analysis, Writing – original draft. **Wenpan Li:** Resources, Data curation, Formal analysis, Methodology. **Yuan Zhang:** Data curation, Formal analysis. **Kai Zhang:** Data curation, Formal analysis. **Shanlin Tong:** Data curation, Formal analysis. **Ruining Jia:** Data curation, Formal analysis. **Qiang Hu:** Data curation, Formal analysis. **Lu Wang:** Data curation, Formal analysis. **Xiaojiao Zhang:** Data curation, Formal analysis.

Data availability

The data that has been used is confidential.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.161613.

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