

Long-Term Impacts of Reservoir Operation on the Spatiotemporal Variation in Nitrogen Forms in the Post-Three Gorges Dam Period (2004–2016)

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Abstract

Nitrogen (N) is an essential nutrient limiting life, and its biochemical cycling and distribution in rivers have been markedly affected by river engineering construction and operation. Here, we comprehensively analyzed the spatiotemporal variations and driving environmental factors of N distributions based on the long-term observations (from 2004 to 2016) of seven stations in the Three Gorges Reservoir (TGR). In the study period, the overall water quality status of the river reach improved, whereas N pollution was severe and tended to be aggravated after the TGR impoundment. The anti-seasonal reservoir operation strongly affected the variations in N forms. The total nitrogen (TN) concentration in the mainstream of the Yangtze River continuously increased, although it was still lower than that in the incoming tributaries (Wu and Jialing rivers). Further analysis showed that this increase occurred probably because of external inputs, including the upstream (76%), non-point (22%), and point source pollution inputs (2%). Besides, different N forms showed significant seasonal variations; among them, the TN and nitrate nitrogen concentrations were the lowest in the impoundment season (October–February), and the ammonia nitrogen concentrations were the highest in the sluicing season (March–May). These parameters varied likely because of internal N transformation. Redundancy analysis revealed that the water level regulated by the anti-seasonal operation was the largest contributor. Our findings could provide a basis for managing and predicting the water quality in the Yangtze River.

1. Introduction

Dams play a significant role in addressing the demand for flood control, power generation, and navigation improvement (Chen et al., 2019; Li et al., 2012; Nilsson et al., 2005). Rivers worldwide have been intensively dammed; more than 70,000 large dams have been constructed, and many others have been proposed or are under construction (Maavara et al., 2015; Shi et al., 2020). However, these projects likely disrupt the river continuity and may have adverse consequences on the balance and functional integrity of river systems (Nilsson et al., 2005; Tang et al., 2018; Wang, 2020; Yan et al., 2015). After impoundment, dam-affected river reaches would be converted into lakes, and this modified fluvial regime likely increases the water retention time and changes the seasonality of suspended and dissolved material fluxes (Eiriksdottir et al., 2017; Friedl and Wüest, 2002; Maeck et al., 2013). Moreover, the biota, especially microorganisms, may be affected by anoxia, sedimentation, and nutrient level variations in reservoir systems (Eiriksdottir et al., 2017; Yan et al., 2015).

Nitrogen, an essential component of all living organisms and primary nutrient for biological growth, is strongly related to the water trophic status (Kuypers et al., 2018; Ran et al., 2017; Zheng et al., 2016). The microbial transformation of N is generally described as an orderly cycle that includes six processes, namely, N fixation, nitrification, denitrification, anammox, assimilation, and ammoniation. In the aquatic ecosystem, inorganic N conversion, such as nitrification and denitrification, has been an essential topic for several decades (Boyer et al., 2006; Zhu et al., 2018). Ammonia can be oxidized to nitrate through nitrification and eventually converted back to dinitrogen through denitrification or anaerobic ammonium oxidation. These alterations of the N oxidation state are controlled primarily by microbial reactions, which

can be affected by many factors (Kim et al., 2016; Povilaitis et al., 2012). For instance, nitrification is aerobic, whereas denitrification usually involves anaerobic and heterotrophic bacteria (Kim et al., 2016; Zhu et al., 2018).

With a length of over 6000 km, the Yangtze River has hundreds of large dams (higher than 15 m; Li et al., 2017; Ran et al., 2017). Three Gorges Reservoir (TGR), one of the largest hydropower complex projects in the world, has significantly reversed the seasonal changes in natural hydrology; in its operation, the water level is artificially regulated to a low level for the need of hydropower energy or flood control in summer and a high level for stable water supply or navigation in winter (Han et al., 2018). The dam holds water and sediments, and 1.8×10^{12} kg of sediments (retention rate over 80%) were trapped from 2003 to 2013 along the 700 km-long TGR (Yang et al., 2014); the clear water discharge has caused substantial river bed erosion downstream the dam. The Three Gorges Project also faced severe controversies concerning the environmental and ecological impact of dams; for instance, water eutrophication, along with construction and operation, has become a hot and critical issue (Chai et al., 2009; Gao et al., 2016; Liu et al., 2018; Ran et al., 2017; Zhou et al., 2013). In the TGR basin, NO₃-N and TN are identified as vital pollution indices in an assessment based on the Canadian Council of Ministers of the Environment Water Quality Index (Xia et al., 2018). Although the TGR has accounted for 5% of N retention in the Yangtze River basin from land to sea since 2004, the enhanced signals of dissolved inorganic nitrogen (DIN) concentrations in the lower reach of the Yangtze River have also been observed (Sun et al., 2013). The DIN concentration dramatically increased from an average of $37 \mu\text{mol L}^{-1}$ in the 1980s to $120 \mu\text{mol L}^{-1}$ in the 2000s (Dai et al., 2011). The reservoir operation has implications not only for N transport but also for N transformation in the TGR (Chai et al., 2016; Shi et al., 2020; Wang, 2020). For example, frequent artificial floods created by the reservoir operation can reduce the ability of soil to retain nutrients and promote the release of N in sediments via coupled nitrification-denitrification processes (Ye et al., 2019; Yu et al., 2020). More functional genes involved in N cycling have been observed in the TGR basin, indicating a higher level of bacterial activity in generating more nitrogenous nutrients (Yan et al., 2015). Since the construction of the Three Gorges Dam (TGD), the transport and transfer patterns of N have changed dramatically, and these variations potentially have a sustainable and crucial effect on the N distribution and trophic status in the TGR.

Therefore, the spatiotemporal variations in N and their relationship with environmental factors should be studied to assess the water quality status and impact of TGR operation, especially when the hydrology regime has undergone tremendous changes since TGR impoundment. The influencing mechanisms of the changing hydrological regime on N cycling in the TGR have been revealed through laboratory experiments by artificially increasing hydrostatic pressure, creating an anti-seasonal wet-dry cycle, and prolonging water residence time (Chai et al., 2016; Shi et al., 2020; Yu et al., 2020). However, most studies have preferred short-term investigation because of difficulties in obtaining long-term observed data (Ding et al., 2019; Huang et al., 2014; Luo et al., 2011; Ran et al., 2017). The time variability of N in TGR involves a wide range of scales from days, months, to multi-years because of the coupled effect of natural (precipitation and monsoon) and anthropogenic (regular operation and staged impoundment of TGR)

factors; as such, studies based on massive monitoring data are more valuable for assessing the long-term impact of TGR operation on N distribution. Besides, studies may explore the relationship between environmental factors and different N forms based on long-term monitoring data on water quality and hydrological parameters.

Here, we collected the observed data of 20 parameters, including N concentrations and other hydrology and water quality variables, in seven gaging stations in the TGR basin from 2004 to 2016. We then analyzed them with various analysis methods. Our study aimed to (i) investigate the long-term effects of TGR operation on the hydrology and water quality, (ii) analyze the N distribution in different temporal stages, and (iii) explore the driving environmental factors of dam-induced spatiotemporal variations in nitrogen forms. This study helped enhance the understanding of the relationships between N concentration and damming-induced environmental variations and provide a scientific basis for evaluating nutrient contents and managing the system of damming rivers.

2. Materials And Methods

2.1 Study area

The TGR basin (29°16'–31°25' N, 106°–110°50' E) spans the Jiangjin District of Chongqing to the Yichang City of Hubei and covers more than 20 county-level administrative regions; of these regions, over 70% are in Chongqing (Fig. 1). With a water surface area of 1084 km², the TGR is rich in water resources, and nearly 90% of the inflow water in the upper reach of the TGR comes from the Yangtze River (71%), Jialing River (13%), and Wu River (16%; Wang et al., 2015; Zheng et al., 2016).

The construction of TGD started in 1994, and the water storage and sedimentation began in 2003. As shown in Fig. 2, the water level stepwise raised to a maximum of 175 m after three impoundment periods (Period I, June 2003–September 2006; Period II, October 2006–September 2009; and Period III, October 2009–present), formed a 650 km-long reservoir with a maximum capacity of approximately 3.93×10^{10} m³ (Wang et al., 2015; Wang, 2015). The TGR usually stores clear water in the dry season and discharges muddy water during the flood season to limit sedimentation and create advantages in terms of navigation, flood control, and power generation as much as possible (Ran et al., 2017). Therefore, the operation cycle of the TGR can be divided into three seasons: low-water level season (June–September), impounding season (October–February), and sluicing season (March–May).

Data were collected from seven key hydrological stations (Fig. 1) to study the N variation in the TGR over the entire cycle of the operation schedule. Among these stations, the Zhutuo (ZT), Beibei (BB), and Wulong (WL) sites were chosen as the inflow stations of the TGR located in the Yangtze River, the Jialing River, and the Wu River, respectively. In the TGR mainstream, ZT, Cuntan (CT), Qingxichang (QXC), and Wanxian (WX) sites are 756, 604, 479, and 288 km away from the TGD, respectively. The QXC site and its upstream sites are considered the tail region of TGR, while the WX site is the representative site of the middle region. Besides, the Yichang (YC) site 38 km downstream of TGD represents the outflow control

station for a comparative study. The river reaches from the WX to the YC site were converted to a lake with a decreased water velocity and prolonged retention time.

2.2 Data collection, sampling, and analysis

The water samples were collected and analyzed in accordance with the *Environmental Quality Standards for Surface Water in China* (MEPC, 2002). The observed monthly hydrology and water quality data from 2004 to 2016 were gathered from the Changjiang Water Resources Commission. Twenty parameters were included: water level (Z , m), flow rate (Q , $\text{m}^3 \text{s}^{-1}$), water temperature (WT, $^{\circ}\text{C}$), flow velocity (U , m s^{-1}), pH, electrical conductance (EC, $\mu\text{S cm}^{-1}$), oxidation-reduction potential (ORP, mv), fluoride (F^{-} , mg L^{-1}), suspended sediment (SS, mg L^{-1}), chloride (Cl^{-} , mg L^{-1}), sulfate ($\text{SO}_2 - 4$, mg L^{-1}), water hardness (mg L^{-1}), alkalinity (mg L^{-1}), permanganate index (PI, mg L^{-1}), dissolved oxygen (DO, mg L^{-1}), 5-day biochemical dissolved oxygen demand (BOD_5 , mg L^{-1}), ammonium nitrogen ($\text{NH} + 4\text{-N}$, mg L^{-1}), nitrite-nitrogen ($\text{NO} - 2\text{-N}$, mg L^{-1}), nitrate-nitrogen ($\text{NO} - 3\text{-N}$, mg L^{-1}), and total nitrogen (TN, mg L^{-1}). Here, the sum of $\text{NH} + 4\text{-N}$, $\text{NO} - 2\text{-N}$, and $\text{NO} - 3\text{-N}$ refers to the DIN, and the difference between TN and DIN refers to residue-N, including particulate nitrogen and dissolved organic nitrogen. At the YC site, several parameters, including flow velocity, F^{-} , $\text{SO}_2 - 4$, PI, and BOD_5 , and observations before 2007 (Period I) were unavailable.

One-way ANOVA was performed to explain the significance of variations in N concentrations ($\text{NH} + 4\text{-N}$, $\text{NO} - 2\text{-N}$, $\text{NO} - 3\text{-N}$, and TN) in different temporal stages. The mutation points and trends of these variations were determined via the Mann–Kendall (MK) test. The relationships between various N forms and environmental variables were determined through redundancy analysis (RDA). In RDA, all data were logarithmically transformed to eliminate the influence of extreme values on ordination scores. Pearson correlation analysis was also applied for comparison.

2.3 N input, output, and retention

Rocks are the major components of the riverbed along the main channel, so the direct groundwater discharge into the TGR can be ignored. Therefore, the total N input of the TGR mainly includes upstream, point source, and non-point source pollution inputs. Given the difficulties in obtaining detailed and comprehensive data, the load of total N input (L_{in}) can be estimated based on the mass balance for the TGR as follows:

$$L_{\text{in}} = L_{\text{out}} / (1 - R_{\text{N}}), \quad (1)$$

where R_{N} is the annual N retained by the reservoir (%), which can be calculated on the basis of the theoretical relationship proposed by Howarth et al. (1996):

$$R_{\text{N}} = 88.45(H/T)^{-0.3677}, \quad (2)$$

where H is the mean depth (m), and T is the water residence time (yr) estimated as

$$T = V / Q, \quad (3)$$

Where V is the effective reservoir volume (m^3). L_{out} is the load of outflow (YC site), which can be calculated as

$$L_{out} = C_N \times Q \times t, \quad (4)$$

where C_N is the TN concentration ($mg L^{-1}$), and t is the elapsed time.

3. Results

3.1 General variation trend of hydrological and water quality regimes

Since the operation of the TGR began, the hydrological and water quality regimes have undergone significant temporal and spatial variations. The values of 16 environmental factors (except four N forms) in different impoundment periods and seasons are listed in Table s1. The three impoundments dramatically raised the water level and substantially decreased the suspended sediment concentration (C_{ss}) and flow velocity in the TGR. Among the seven stations, the WX site suffered the most remarkable effect of TGR operation, that is, the water level rose by 23.8 m, whereas flow velocity and C_{ss} respectively dropped by 46.7% and 84% from Period I to Period II (Table s1). One-way ANOVA revealed that the water temperature and flow rate in all stations exhibited no significant trend (Table s2). The periodic mean pH values were greater than 8.0, and water alkalinity also increased over time. This result indicated that the overlying water in the TGR would remain in a weak alkaline state in the long run. An overall rise in ion concentration level was found during the three periods, with a sharp increase in Cl^- and SO_4^{2-} , a slight increase in F^- , EC, and water hardness, especially at the WX site, the closest site to the TGD. The periodic averaged ORP and DO concentrations shared a similar trend; they significantly decreased from Period I to Period II and slightly increased in Period III. During the monitoring period, the F^- , DO, PI, and BOD_5 concentrations were in the ranges of 0.07–1.05, 5.45–10.95, 0.55–32.59, and 0.20–2.41 $mg L^{-1}$, respectively. Although the inter-annual variations in these four parameters showed different trends, the F^- , DO, and BOD_5 concentrations in all the stations reached the requirement of Class II standard (MEPC, 2002). The PI concentration was lower than the Class II standard (6 $mg L^{-1}$), but it met the Class III standard (4 $mg L^{-1}$), indicating an overall water quality improvement after the TGR impoundment.

Through the anti-seasonal reservoir operation, the water level in the dry season was higher than that in the rainy season (low-water level season or June–September). The flow rate, flow velocity, water temperature, and C_{ss} were the highest in the low-water level season because of frequent flooding in

summer (Table s1). In addition to pH, ORP, Cl^- , and BOD_5 , other water quality parameters exhibited significant seasonal changes; among them, water hardness, water alkalinity, EC, F^- , $\text{SO}_2 - 4$, and DO were the highest in the impounding season and the lowest in the low-water level season (Table s2).

3.2 Spatial-temporal variation in N in the TGR basin

Although the overall water quality has been improved, N pollution in the TGR is severe and aggravated. As shown in Fig. 3, the TN concentration in almost all stations reached or was even worse than the Class Ⅲ standard ($> 2 \text{ mg L}^{-1}$; MEPC, 2002). The TN concentrations significantly increased toward the TGD in the mainstream from 1.57 mg L^{-1} at the ZT site to 1.86 mg L^{-1} at the WX site. The DIN is the existing primary form of TN, which mainly consisted of $\text{NO} - 3\text{-N}$ (80–91%) and some $\text{NH} + 4\text{-N}$ (2–10%) and $\text{NO} - 2\text{-N}$ ($< 2\%$). The percentage of $\text{NO} - 3\text{-N}$ increased gradually from upstream (80.2% at ZT) to downstream (85.5% at WX) in the mainstream. On a multi-year average, the TN concentration was 2.43 mg L^{-1} in the Wu River (WL station) and 2.01 mg L^{-1} in the Jialing River (BB station), indicating relatively higher TN concentrations in tributaries than in the mainstream. Similar to the mainstream of Yangtze River, tributaries dominantly had $\text{NO} - 3\text{-N}$, which accounted for 91.0% of TN at the WL station and 81.5% at the BB station. Besides, the multi-year averaged TN concentration decreased slightly in the outlet (1.83 mg L^{-1} at the YC station), whereas $\text{NO} - 3\text{-N}$ took more part of TN form (87.5%) than that in the middle region of TGR (85.5% at the WX station).

In the long term, the TN concentration in the tail region was less affected by the construction and operation of the TGR without an evident trend in the staged TN concentrations, but it notably increased in the middle region (WX site) and outlet (YC site; Table s2). By contrast, the concentrations of N fractions significantly changed because of the considerable variations in the hydrologic regime in the three impoundments (Period Ⅰ to Period Ⅲ). In Fig. 4, the $\text{NO} - 3\text{-N}$ concentration in the mainstream of the TGR continuously increased, whereas the $\text{NH} + 4\text{-N}$ concentration decreased. These similar trends were observed at the WL site; however, the $\text{NH} + 4\text{-N}$ increased in Period Ⅲ at the BB site compared with that in Period Ⅰ. The staged averaged $\text{NO}_3\text{-N}$ and $\text{NH} + 4\text{-N}$ concentrations at the YC station in Period Ⅲ were higher than those in Period Ⅰ. Besides, the concentrations of $\text{NO} - 2\text{-N}$ in the seven stations were relatively lower than those of the other N forms, and the varying trend was not evident.

The temporal variations in N concentrations displayed dramatic seasonality patterns (Fig. 5). The highest concentration of $\text{NH} + 4\text{-N}$ was observed in the sluicing season (March–May), and the maximum ratio of 3.8 in the two other seasons occurred at the WX site in 2014. Although extreme differences were found in several years, no clear trend in the seasonal concentrations of $\text{NO} - 2\text{-N}$, especially in the tail region ($p > 0.05$), was detected. The concentrations of $\text{NO} - 3\text{-N}$ and TN were one order magnitude higher than those of $\text{NH} + 4\text{-N}$ and $\text{NO} - 2\text{-N}$, and ANOVA revealed that their seasonal variations were significant. The concentrations of the $\text{NO} - 3\text{-N}$ and TN were the lowest in the impounding season and relatively high in the two other seasons. This event reoccurred in each year at the WX site within the TGR, especially in 2008 when the TN concentration in the low-water level season (2.12 mg L^{-1}) was approximately 1.42 times that in the impounding season (1.50 mg L^{-1}).

3.3 Influence of environmental variables on N distribution

The correlation structures between the N forms ($\text{NH}_4\text{-N}$, $\text{NO}_2\text{-N}$, $\text{NO}_3\text{-N}$, and TN) and other environmental variables in the mainstream sites (ZT, CT, QXC, and WX sites) of the TGR from 2004 to 2016 were achieved through RDA. As presented in the ordination biplot (Fig. 6), the water level was the greatest contributor to the variations in N concentrations. This result indicated that the operation of the TGR might significantly affect the N distribution. The flow velocity (U) also strongly correlated with the N forms, whereas the flow rate (Q) with insignificant periodic changes slightly contributed to the N variations. High C_{SS} might correspond to an increase in $\text{NH}_4\text{-N}$ concentrations and a decrease in $\text{NO}_3\text{-N}$ concentrations. As critical environmental factors in the N cycle, pH and DO could alter the existing N forms, but the observed N concentrations were affected by the aggregate of environmental conditions over time. Although PI, BOD_5 , and F^- had no direct effect on N transformation, these three water quality parameters had significant positive correlations with the N forms in the mainstream of the TGR. Similar results were also supported by Pearson correlation tests. The corresponding coefficients of N forms and environmental factors are listed in Table s3.

4 Discussion

4.1 N input in the TGR basin

After the impoundment of the TGR, the water quality in the tail and the middle region demonstrated an overall improvement. For instance, the periodic concentrations of PI and BOD_5 gradually decreased, but N pollution may be a severe problem in the future. Although no significant annual and periodic variations in the tail region of the TGR were observed, the MK test results (Fig. s1) revealed that the TN concentration in the WX site increased after the impoundment, especially after the 175 m impoundment operation in 2010. Among the seven observation sites, the WX station closest to the dam showed the highest TN concentration and was most severely affected by the reservoir operation. As one of the important economic centers in the TGR, the water quality status of the WX site is also closely related to the industrial, agricultural and population development of the TGR. Hence, the variations in TN concentrations at the WX station proved that the whole TGR faces a severe risk of N pollution.

The increasing trend of TN concentrations may be related to external N input from the TGR basin. The total N input listed in Table 1 was obtained based on the export load and retention rate (Eq. (1)). The detailed information can be found in Table s4. Among the N inputs, upstream, point, and non-point source pollution inputs accounted for about 76%, 2%, and 22% from 2007 to 2016, respectively. Similar to the N output calculation, the upstream input that included three incoming rivers was related to the flow rate and TN concentrations (Eq. (4)). Despite the observable seasonal and annual variations, the TN concentrations in the three incoming rivers were insignificant in the periodic variations (Table s2). Conversely, the improvement of the industrial wastewater treatment technology caused an evident reduction of sewage discharge, but domestic sewage discharge showed a sustained increase because of the high urbanization rate (Table s5). The detailed information on annual sewage discharge is presented

in the TGR Bulletin (MEPC, 2017). According to the emission standard of the sewage treatment plant ($TN \leq 15 \text{ mg L}^{-1}$), the point source N input, including N in the domestic and industrial wastewater, was estimated to range from $1.34 \times 10^7 \text{ kg}$ to $2.02 \times 10^7 \text{ kg}$ from 2004 to 2016, with an average of $1.55 \times 10^7 \text{ kg}$ (Table 1).

Table 1
N input, output, and retention rate in the TGR.

Year	Input				Output (10^7 kg)	Retention rate (%)
	Total (10^7 kg)	Upstream (10^7 kg)	Point source pollution (10^7 kg)	Non-point source pollution (10^7 kg)		
2004	NA	68.66	1.59	NA	NA	NA
2005	NA	NA	1.47	NA	NA	NA
2006	NA	NA	1.55	NA	NA	NA
2007	77.59	62.08	14.3	14.08	71.28	8.13
2008	76.34	67.64	1.73	6.97	70.01	8.29
2009	73.08	70.90	1.66	0.52	66.77	8.63
2010	74.60	60.65	1.40	12.55	67.94	8.92
2011	58.70	47.33	1.35	10.02	53.14	9.48
2012	NA	76.69	1.36	NA	NA	NA
2013	77.18	58.81	1.47	16.90	70.10	9.17
2014	97.64	43.02	1.51	53.11	89.36	8.48
2015	83.07	63.43	1.54	18.11	75.49	9.13
2016	94.74	54.16	2.02	38.56	86.85	8.33
mean	79.22	61.22	1.55	18.98	72.33	8.73
NA means data are not available.						

Another important cause of N increase in TGR might be non-point source pollution, which was obtained by subtracting the upstream and point source pollution input from the total TN input in this study. Therefore, the non-point source pollution may be overestimated because of the incomplete statistics of point source pollution and upstream input; for example, some micro-enterprise discharges may not be included in monitoring and sewage treatment. However, non-point source pollution that has attracted

more attention plays a vital role in the cumulative increase in TN (Alexander et al., 2002; Ma et al., 2011). In addition to natural N fixation through natural vegetation and atmospheric lightning, drastically increased human activities have strongly influenced N loads in the TGR basin (Boyer et al., 2006; Chen et al., 2016; Galloway et al., 2008; Xv et al., 2020). With expanding population and agricultural activity, chemical fertilizers have been excessively utilized in China, and approximately 53.2% were N fertilizers in the TGR basin from 2004 to 2016 (NBSCC, 2017). In the entire TGR, the incremental net N fertilizer ranges from 294.0×10^6 kg to 332.2×10^6 kg, with an average of 320.5×10^6 kg N (Table s5). The massive use of N fertilizer has become a crucial N source, accounting for more than 50% of the net anthropogenic regional N input (NANI), followed by atmospheric N deposition, feed nitrogen input, and crop fixation (Ding et al., 2020; Xv et al., 2020). According to data from hundreds of observational sites, the average N wet deposition over China increased by nearly 25% from the 1990s to the 2000s (Jia et al., 2015). A similar increasing trend also occurred in the TGR basin, where atmospheric N deposition increased by 22% from 2006 to 2016 (Table s5). This variation could be attributed to the exponential increase in energy consumption and industrial waste gas (Table s6), identified as potential sources of atmospheric N deposition (Wang et al., 2018). Moreover, crops have been increasing since the 2006 drought, and the use of feed N has increased with the exponentially growing population and economy in Chongqing (Table s6). Under rainfall and irrigation actions, considerable non-point source N likely enters the water column through surface runoff, subsurface flow, farmland drainage, seepage (Gao et al., 2016b), and frequent flooding caused by reservoir operation aggravates the loss of N.

4.2 Impact of the water level variations in the TGR on N transformation

Internal biogeochemical transformations, including 14 discrete redox reactions that can convert N redox states from -3 to $+5$, are more complex and unpredictable based on our current understanding than the determined external N inputs in the TGR (Kuypers et al., 2018). These reactions (Fig. 7) were susceptible to environmental factors and likely to be altered by large water level fluctuations (145–175 m) and the corresponding dramatic environmental changes, resulting in the variations in N forms. The strong impact of environmental variables was also demonstrated by the result of RDA (Fig. 6).

Although the water dilution effect caused by the impoundment can alleviate pollution (Jiang et al., 2018), the periodic mean NO_3^- -N and TN concentrations continuously increased during three impoundments, indicating the greater effect of the ever-increasing external input from a long run. However, the change in the proportion of N forms caused by reservoir storage could not be ignored. For example, the NH_4^+ -N concentration decreased significantly when the concentrations of other N forms increased (Fig. 4). During the three impoundments, the water area of the TGR basin, which was about 2.53 times at 175 m (1084 km^2) than at 135 m (428 km^2), increased as the water level rose, leading to a sharp increase in the water-sediment interface (Wang et al., 2020). This increased interface area would provide larger places for N cycling and facilitate the entry of N to the waterbody. An anti-seasonal hydrological regime may bring about more marked differences in N distribution in a year than the long-term effects of the three impoundment periods.

In a low-water level season, a water level fluctuation zone (WLFZ) of about 350 km² along the reservoir becomes exposed; in the WLFZ, carbon and N contents in soil are high because of the continuous accumulation of organic matter (Wang et al., 2020; Ye et al., 2011). In this season, high temperature is favorable to the growth of plants in the WLFZ, where more than 80 species of vascular plants were recovered in 2015 (MEPC, 2017); thus, the absorption and utilization of bioavailable N forms are promoted (Ye et al., 2015). The corresponding water temperature is suitable for nitrification; at this temperature, the involved microorganisms generally have greater abundance and diversity (Kuypers et al., 2018). The low-water level season of the TGR is consistent with the rainy season of the Yangtze River (May–October). Thus, the increased rainfall and frequent flood in the upper reaches of the Yangtze River and the TGR basin could lead to an increase in the flow velocity in this season by one order of magnitude compared with those in the impounding season (Fig. 7). The increased water velocity strengthens the disturbance to the bottom of the river and promotes the ammonification of organic N with oxygen replenishment in the water-sediment surface (Yu et al., 2019); this phenomenon may partially explain the higher NH₄-N concentration in the sluicing and low-water level seasons. On the other hand, the strong hydrodynamic disturbance facilitates the suspension of sediments, while the nitrification rate enhances as C_{SS} increases (Wang et al., 2010). The SS is possibly an anoxic/low-oxygen microsite, so coupled nitrification-denitrification may occur in the water column (Xia et al., 2017), and nitrate produced through nitrification at SS can be converted into dinitrogen gas (N₂) through denitrification. This N loss enhancement is approximate 25–120% caused by 1 g L⁻¹ SS in the Yangtze River (Xia et al., 2017). Although the release amount is relatively small, nitrous oxide (N₂O) is the primary ozone-depleting agent and potent greenhouse gas that profoundly affects the ecological environment (Kuypers et al., 2018; Shi et al., 2020).

When the water level remains high (impoundment season), the short-term vegetation in the fluctuating zone becomes submerged, decomposes, and releases N, thereby increasing the risk of eutrophication of the TGR during the impoundment season. For example, 81.1 kg N ha⁻¹ was released from nine dominant plant species after 200 days of soaking in the WLFZ (Xiao et al., 2017). The high hydrostatic pressure caused by the large water depth significantly increased the release and ammonification of N but slightly affected nitrate reductase activity (denitrification); consequently, NH₄-N and NO₃-N accumulate (Chai et al., 2009). However, the concentrations of NO₃-N and TN were the lowest in the impounding season. This phenomenon may be caused by many factors; among them, the dilution effect might make the greatest contribution to reducing N concentrations because of the dramatically increased storage capacity from 1.71 × 10⁹ m³ to 3.93 × 10⁹ m³, comparing to the relatively insignificant variations in N input in the short term. Besides, the reduced water velocity could weaken the entry of N into the water body and prolong the residence time of water in the TGR (Shi et al., 2020). In the reach from the ZT to the WX site, the water residence time increased from 2.69 days in the low-water level season to 30.27 days in the impoundment season in 2016. The observably extended water residence time accelerates N removal from a waterbody (Keys et al., 2019; Saunders and Kalff, 2001; Tong et al., 2019). However, a decrease in C_{SS} provides fewer places for coupled nitrification-denitrification processes, and these processes are also inhibited by low temperature in the impoundment season (Palacin-Lizarbe et al., 2018). The nitrification

rate decreases rapidly when the temperature is lower than 15°C and nearly stops below 5°C. Similarly, the denitrification rates immediately decrease with both cooling and lower reactive nitrogen load (Palacin-Lizarbe et al., 2018).

The reservoir operation has regulated the water level and resulted in dramatic environmental variations. Further developments about the relationship between N cycling and other environmental factors are still needed to help explain the N variation caused by the reservoir operation and eventually improve the predictions and management of the water quality in the Yangtze River.

5. Conclusion

In this study, data on 20 hydrological and water quality parameters of seven gaging stations in the TGR basin were collected from 2004 to 2016. The operation of the TGR significantly changed the hydrological regime of natural rivers, improving the water level while decreasing the C_{SS} and water velocity. The impoundment alleviated the water pollution and reduced the PI and BOD₅ concentrations, but the TN concentration still met or was even worse than the Class Ⅲ standard of China. The multi-year averaged TN concentration increased along the mainstream of the Yangtze River, but it was still lower than that in the incoming Wu River (2.43 mg/L) and Jialing River (2.01 mg/L). The DIN was the most abundant N form, which consisted of NO₃-N (80%–91%) and some NH₄⁺-N (2%–10%), and NO₂-N (<2%). The N distribution at different temporal levels was subjected to synthesis analysis. No evident trend was found in the periodic TN concentrations except at the WX and YC sites, whereas other DIN forms markedly changed. The anti-seasonal reservoir operation significantly caused the seasonal variations in different N forms. Among them, the NO₃-N and TN concentrations were the lowest in the impoundment season, whereas the NH₄⁺-N concentrations were the highest in the sluicing season.

External input and internal transformation contribute to variations in N distribution. The continuous long-term increase in the TN concentrations of the TGR was the integrated result of the upstream, non-point, and point source pollution inputs, which accounted for 76%, 22%, and 2%, respectively. In terms of internal transformation, the RDA results revealed that the water level regulated by the anti-seasonal reservoir operation had the highest correlation with the variations in N forms. In the low-water level season, high water temperature, flow velocity, and C_{SS} would enhance the N release from the water-sediment interface and promote the coupled nitrification-denitrification process. In the impoundment season, the dilution effect and low N reaction rate might jointly result in the lowest NO₃-N and TN concentrations.

Further studies on the impact of reservoir operation based on long-term observation and analysis will promote an accurate and comprehensive understanding of N distribution and improve the assessment and prediction of the water quality of the TGR and the Yangtze River.

Declarations

Ethics approval and consent to participate

All the authors have read and approved the manuscript and consented to participation.

Consent for publication

All the authors have consented to publication.

Availability of data and materials

All the data and materials in the manuscript are available upon request.

Competing interests

The authors declare no competing interests.

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Authors' contributions

Bei Nie: Conceptualization, Formal analysis, Visualization, Writing- Original draft preparation

Yuhong Zeng: Supervision, Writing- Reviewing and Editing, Funding acquisition

Lanhua Niu: Resources

Xiaofeng Zhang: Project administration, Funding acquisition

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Figures

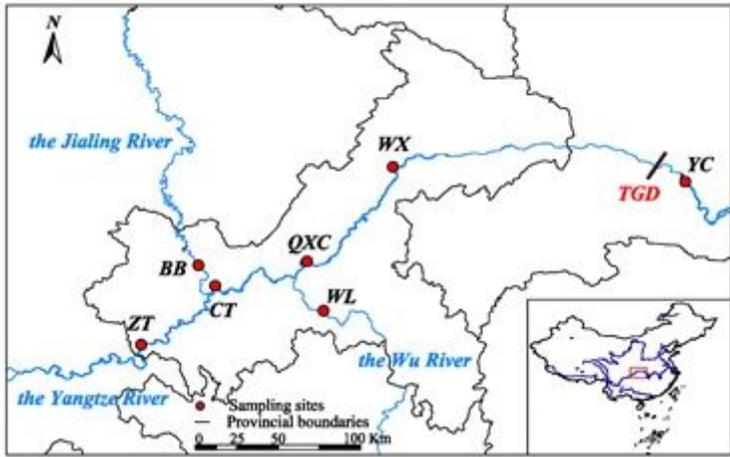


Figure 1

Maps of the study area and sampling sites. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

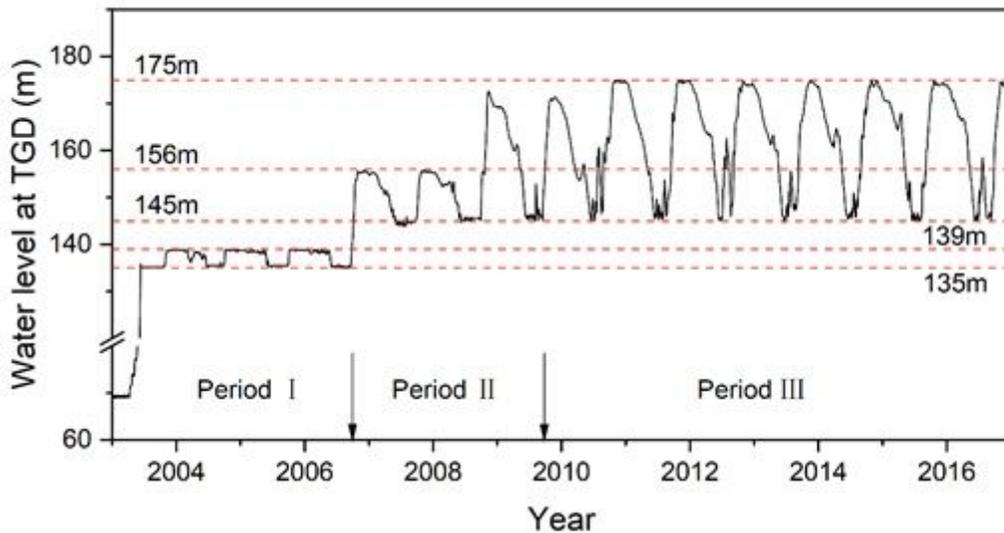


Figure 2

Operation strategy of the TGR from 2003 to 2016. The daily water level at the TGD was sourced from www.ctg.com.cn.

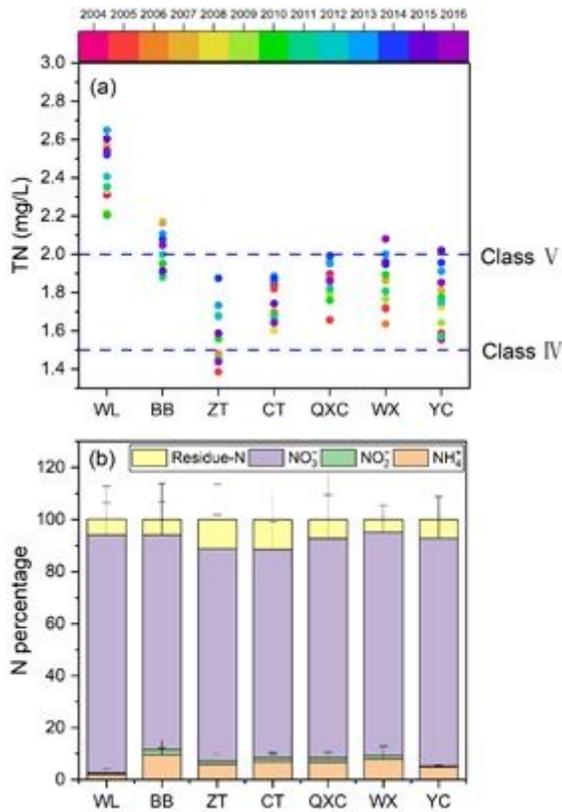


Figure 3

Spatial distribution of annual TN concentrations and multi-year averaged N percentage in the TGR basin.

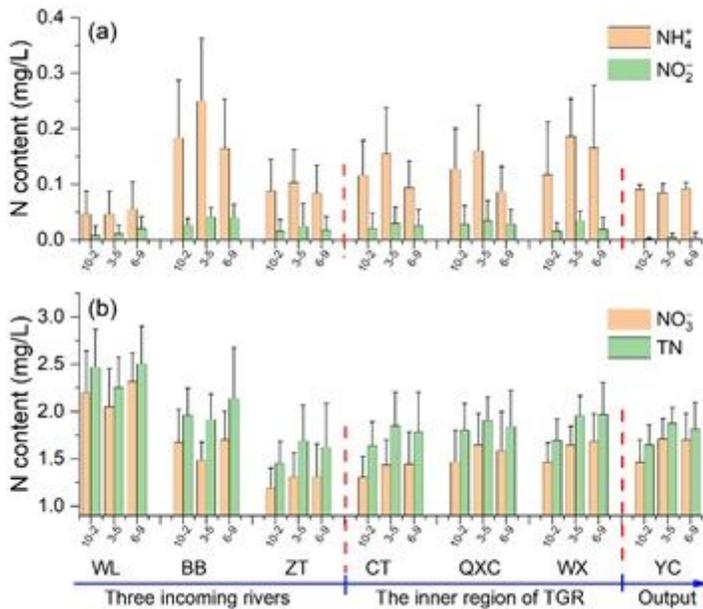


Figure 4

Variations in different N fractions in the three periods in the TGR.

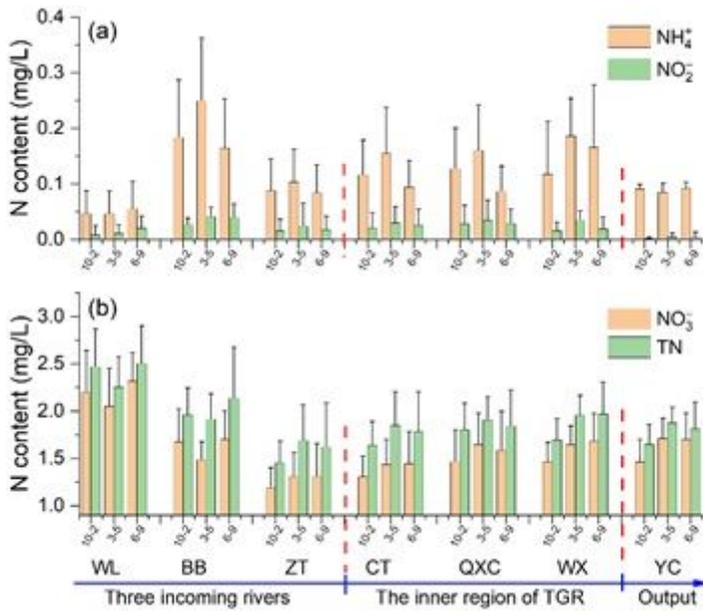


Figure 5

Seasonal variations in different N forms in the TGR basin.

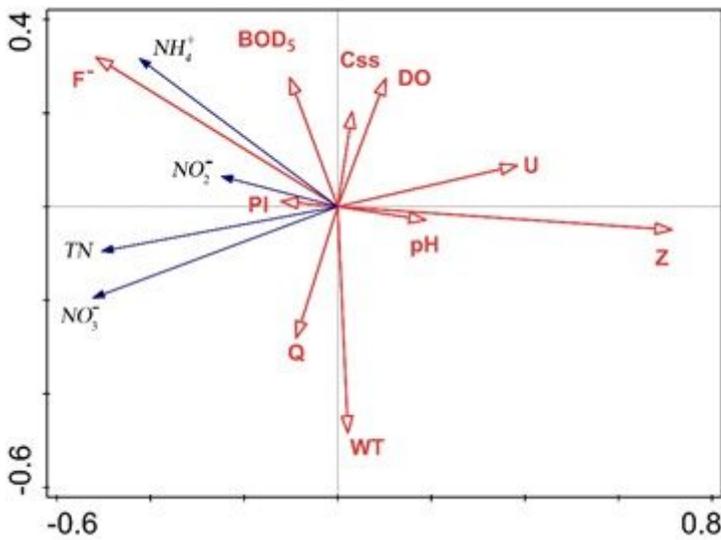


Figure 6

RDA on the effects of environmental variables on N forms at the mainstream sites. The mainstream sites were the ZT, CT, QXC, and WX stations. Of the 16 environmental variables, 10 were selected, that is, four hydrological parameters (Z, U, Q, and Ccss) and six water quality parameters (DO, PI, pH, BOD5, WT, and F-) mentioned in MEPC (2002).

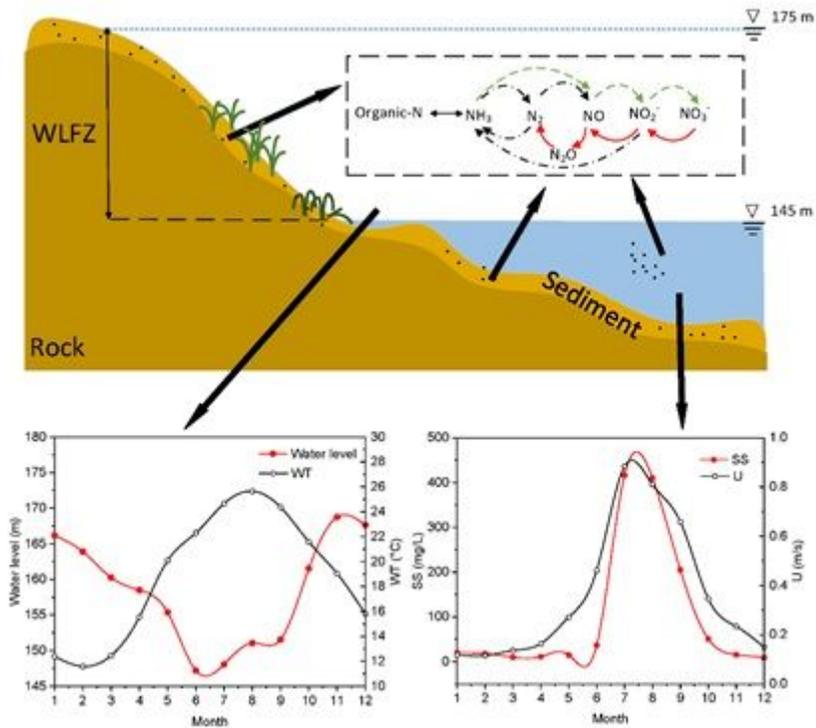


Figure 7

Schematic of the TGR incorporating the N reaction processes and monthly variations in environmental factors at the WX site caused by the reservoir operation in 2004–2016. Among the six N reactions, the solid red lines represent nitrification, the dashed green lines correspond to denitrification, and the dot-dash black lines refer to other reactions. WT stands for water temperature.

Supplementary Files

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