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Anthropogenic impacts on nutrient variability in the lower Yellow River

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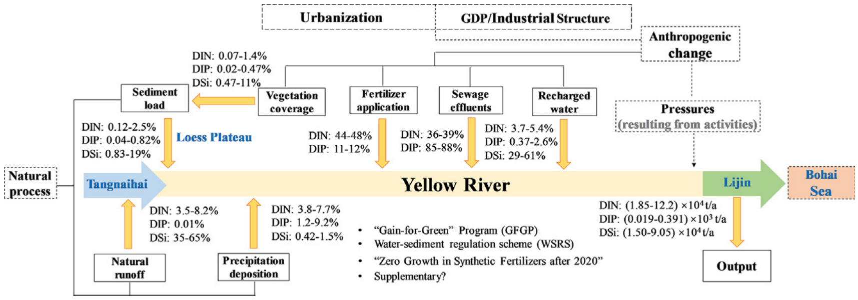
HIGHLIGHTS

- Dissolved inorganic nutrients decreased during 2001-2018 in the lower Yellow River.
- Dam construction and phytoplankton uptake significantly lowered nutrient concentrations.
- The nutrient loading to the Yellow River derived from environmental and societal impacts were quantified.
- Total nutrient influxes for DIN, DIP and DSi were mainly from fertilizer loss, sewage effluents and runoff, respectively.

GRAPHICAL ABSTRACT



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ABSTRACT

Excessive nutrient discharges and changes in nutrient ratios caused by global change and anthropogenic activities have been reported in global rivers; however, the actual alterations occurring in the Yellow River environment is too fast to catch up with. From 2001 to 2018, dissolved inorganic nitrogen (DIN), dissolved inorganic phosphorus (DIP) and dissolved silicon (DSi) concentrations showed decreasing trends in the lower Yellow River throughout the study period. Dissolved organic phosphorus (DOP) concentrations increased since 2009, reaching up to 95% of the total dissolved phosphorus. Annual minimum dissolved organic nitrogen concentrations increased with time. We observed extremely low nutrient concentration events since 2014 in response to the retention effect of large reservoirs; this significantly reduced the downstream water discharge and sediment load and increased phytoplankton uptake. To further analyze the variability of nutrient fluxes, we quantified the fluxes to the Yellow River from natural (runoff, precipitation deposition, and sediment load from the Loess Plateau), anthropogenic (recharged water, fertilizer application, and vegetation coverage), social and industrial (population urbanization, GDP, and sewage effluents) sources. The highest contributions of total nutrient fluxes emptied into the Yellow River was fertilizer losing (44–48%) for DIN, sewage effluents (85–88%) for DIP, and runoff (35–65%) for DSi, respectively. Strictly controlling the amount of fertilizer and improving the application methods, improving sewage treatment technology, and vigorously promoting "green travel" might reduce nutrients emptied into the Yellow River based on the main sources of nutrients. Our study may help policy makers formulate strategies and it is possible to own a better water quality in the Yellow River.

1. Introduction

Rivers transport large amounts of sediment, organic matter, nutrients, and other materials from land to sea and significantly influence the biogeochemical cycling of elements in estuaries (Turner et al.,

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2003; Seitzinger et al., 2010; Goes et al., 2014; Li and Bush, 2015; Liu, 2015; Tong et al., 2015; Bi et al., 2019). Nutrient concentrations in rivers have increased in response to socio–economic development and intensified anthropogenic activity in river basins, resulting in eutrophication and the subsequent degradation of estuarine ecosystems (Xia et al., 2001; Dagg et al., 2004; Seitzinger et al., 2010; Yu et al., 2010). However, there are great differences in the watershed environments of global rivers; and the contribution of diverse sources to the total nutrient loading to aquatic systems can be highly variable over time and space depending on their drivers (e.g., population density, sewage treatment technology, hydrology, land–use, and climate change) (Vilmin et al., 2018). For instance, agricultural activities and sewage effluents were major impacts on the variations of nutrients in the river basin of the Bay of Bengal (Sattar et al., 2014) and the Gulf of Gaeta in Central Italy (Calizza et al., 2020); while precipitation and agricultural sources dominated the dissolved inorganic nitrogen (DIN) in the Mekong basin (Li and Bush, 2015). Besides, damming caused a 90% decrease in flow of the Nile to the Mediterranean and dramatically reduced the fluxes of inorganic nitrogen, biologically available phosphorus and silica to coastal waters (Nixon, 2003; Maavara et al., 2020). And the Danube River also experienced a >60% decrease in dissolved silicon (DSi) at the mouth of the river after damming (Humborg et al., 1997; Maavara et al., 2020). Therefore, the impact factors of each river should be discussed according to the specific characteristics and practices within its basin.

The Yellow River was once the second largest river in the world in terms of sediment load (Milliman and Meade, 1983; Milliman and Syvitski, 1992; Wang et al., 2007; Bi et al., 2019). Like other global rivers, the Yellow River is undergoing many stresses from dam construction, rapid population growth and urbanization, and great industrial structure changes, especially in the last two decades (China Statistical Yearbook (CSY), 2002–2018; Wang et al., 2007, 2017). The key issues of the policies implemented in the Yellow River have greatly altered from "soil and water conservation" (Standing Committee of the National People's Congress, 1991) and "preventing floods and reducing the Yellow River bed silt" (Yellow River Conservancy Commission (YRCC), 2002) to "ecological conservation and high–quality development of the Yellow River Basin" (YRCC, 2013; MOEE, 2020) and "Zero Growth in Synthetic Fertilizer after 2020" (MOA, 2015).

There are long–standing concerns about the variation and transfer of water, sediment, and nutrients in the Yellow River. Since 1950, 3147 reservoirs and dams have been built in the Yellow River basin (Wang et al., 2006; Liu, 2015). As a result, the Yellow River has experienced a rapid decline in water discharge and sediment load since 1980 (Wang et al., 2006, 2007, 2011, 2017; Wang et al., 2016). To balance the relationship between water and sediment loads to reduce deposition and increase the flood carrying capacity of the downstream channel, the YRCC implemented the water–sediment regulation scheme (WSRS) during the flood season (June–September) in 2002. The scheme has been conducted a total of 17 times (excluding 2016 and 2017) and contributed 14–56% of the total annual water discharge to the sea (Liu et al., 2012; Liu, 2015; Wang et al., 2017; Wu et al., 2017). The regulation also resulted in the large fluxes of nutrients from the Yellow River to the Bohai Sea within a short period, accounting for 23–68% of the annual nutrient fluxes (Chen et al., 2013; Liu et al., 2012; Liu, 2015; Wu et al., 2017; Wang et al., 2017). After the expiration of the "The recent key development plan of the Yellow River: 2002–2012" (YRCC, 2002), the water discharge transported into the Bohai Sea during the WSRS has decreased, and even the WSRS was not implemented in 2016–2017. The influence of this action on both nutrient concentrations and fluxes of the lower Yellow River cannot be ignored.

Relative to global average values, dissolved inorganic phosphorus (DIP) concentrations (0.03–0.95 μM) are relatively low in the Yellow River, nitrate (NO_3^-) concentrations (157–501 μM) are high and DSi concentrations are moderate (72–167 μM) (Dagg et al., 2004; Yao et al., 2009; Zhang et al., 2010; Liu et al., 2012; Chen et al., 2013; Gong et al., 2015; Liu, 2015; Wu et al., 2017). Before 2000, the nitrogen load in the Yellow River was mainly impacted by population growth and nitrogen fertilizer application; and phosphorus transportation was dominantly controlled by soil erosion from the Loess Plateau (Yu et al., 2010). While during 2002–2004, the most significant source of DIN and DIP was wastewater (Gong et al., 2015). Soil erosion was the primary influence on DSi (Ran et al., 2015). Previous studies on Yellow River nutrients mainly focused on concentrations and sources, and the main considerations were WSRS, agriculture, and sewage. However, the Yellow River has experienced rapid ecological environment changes that are difficult to catch up with particularly after 2000. And the environmental awareness in China has been increasing (Strokal et al., 2017). Therefore, it is necessary for us to address the variations of nutrients in the Yellow River from the perspective of the human society–ecological environment.

We therefore analyzed the causes for nutrient concentrations as well as their fluxes variability from driver changes of the society and environment based on the monthly water discharge, sediment load, nutrient concentrations, and nutrient fluxes in the lower Yellow River from 2001 to 2018. We also quantitatively estimated the proportional contributions of various factors affecting the nutrient fluxes comprehensively, including natural runoff, precipitation, the Loess Plateau sediment, vegetation coverage, recharged water from irrigation, agricultural fertilizer, sewage effluents, population, urbanization, and industrial structure.

2. Materials and methods

2.1. Study area

The Yellow River originates from the eastern Qinghai–Tibet Plateau and flows through nine provinces (regions) or municipalities—Qinghai (QH), Sichuan (SC), Gansu (GS), Ningxia (NX), Inner Mongolia (NM), Shaanxi (SX1), Shanxi (SX2), Henan (HN) and Shandong (SD) provinces—before emptying into the Bohai Sea (Fig. 1). The Yellow River basin covers an area of 79.5 × 10^4 km^2 and has a total length of 5464 km (YRCC, 1998; Wang et al., 2006; Wang et al., 2007). The Yellow River is split into three reaches based on hydrology and geography: 1) the upper reaches (3472 km) from the source region to Hekou Town in Inner Mongolia, 2) the middle reaches (1206 km) from Hekou Town to Taohuayu (Henan), and 3) the lower reaches (786 km) from Taohuyu to the Bohai Sea (YRCC Yellow River Conservancy Commission, 2007; Ding et al., 2011). Owing to sediment deposition (Bi et al., 2019), the lower reaches eventually form a "hanging river", and there are no tributary inputs to the downstream region (Ding et al., 2011). In this study, nutrient samples were collected at station LJ; water discharge was measured at stations Tangnaihai (TNH), Lanzhou (LZ), Toudaoguai (TDG), Tongguan (TG), Sanmenxia (SMX), Huyaunkou (HYK), Gaocun (GC), and LJ; and sediment load was measured at stations TDG, TG, Xiaolangdi (XLD), and LJ. Station LZ is located upstream; stations TDG, TG, SMX, XLD, and HYK are located midstream; and stations GC and LJ are located downstream (Fig. 1). TNH is the first gauging station in the upper reaches of the Yellow River and thus represents the natural input to the Yellow River from its source region. The LJ is the final gauging station in the downstream region of the Yellow River and therefore represents the output of the Yellow River into the Bohai Sea.

The arid to semi–arid climate of the Yellow River Basin results in high evaporation and low precipitation, leading to the accumulation of weathering products in the soil and weathering crust (Zhang, 1996). The main source of sediment to the Yellow River are middle reaches and which account for 55.7% of the total sediment load (Ding et al., 2011). The middle reaches consist of unconsolidated loess, which is more prone to weathering compared to well–weathered bedrock (Cai et al., 2008). The Yellow River basin is the first agricultural region in China (Wang et al., 2015) and accounts for 8.7% of the country's total population (Wang et al., 2006; Wang et al., 2007). The basin is the main wheat and corn producing region in China (Wang et al., 2015)

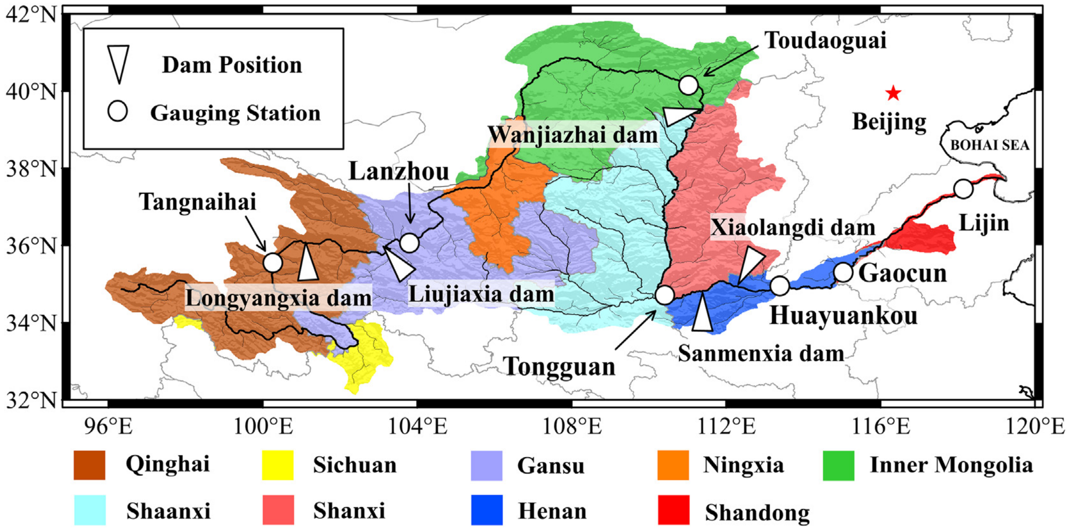

Fig. 1. Map of the Yellow River basin (YRB). The nine provinces are indicated by the colored areas. Nutrient samples were collected at station Lijin (LJ), water discharge was measured at stations Tangnaihai, Longyangxia, Liujiaxia, Lanzhou, Toudaoguai (TDG), Wanjiazhai, Tongguan (TG), Sanmenxia, Huyaunkou, Gaocun, and Lijin, and sediment load was measured at stations TDG, TG, Xiaolangdi, and LJ. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

and has a cultivated area of 1.6×10^7 km^2—approximately 1.4 times the national per capita cultivated land. Cultivation in the Yellow River basin requires high fertilization and water for irrigation.

2.2. Data sources and methods

The monthly and annual water discharge at stations LZ, TDG, TG, SMX, HYK, GC, and LJ were sourced from the Yellow River Water Resources Bulletin (YRWRB, 2001–2017). Annual precipitation, water withdrawal, water consumption, and sewage effluents in the Yellow River basin were also sourced from the YRWRB (2001–2017). The monthly and annual sediment load at stations TDG, TG, and LJ were sourced from the Yellow River Sediment Bulletin (YRSB, 2006–2017) and Wang et al. (2016). The water storage level of the XLD Reservoir and the daily water discharge at stations XLD and LJ during the observation period were obtained from the YRCC (http://www.yrcc.gov.cn/). Forest and vegetation coverage data for each province in the Yellow River basin from 2003 to 2014 were collected from the China Forestry Database (http://www.forestry.gov.cn/data.html). The original provincial data of the Yellow River basin in 2001–2017 were sourced from the National Bureau of Statistics of China (NBSC, http://data.stats.gov.cn/) including the fertilizer application (nitrogen and phosphorus), GDP, primary industry, secondary industry, tertiary industry, rural population, urban population, and total population.

River water samples were collected monthly at LJ in SD province from March 2001 to December 2004 and from November 2008 to May 2018. We used the 2001–2004 nutrient concentrations and fluxes data from Yao et al. (2009), Zhang et al. (2010), and Gong et al. (2015) to supplement our laboratory observational data from November 2008 to May 2018. Our observational data from November 2008 to December 2011 is published in Liu et al. (2012) and Chen et al. (2013), data from February 2012 to March 2014 is published in Wu et al. (2017), and data from March 2014 to May 2018 were newly measured. We collected surface water samples (0–0.5 m) at three to five sites across the river from a bridge using a polyethylene bucket (Liu, 2015). Before sample collection, sample buckets and polyethylene bottles were pretreated with 1:100 HCl and filters were pretreated with 1:1000 HCl; these were then neutralized by rinsing with Milli–Q water. After sample collection, water samples were immediately transported to the laboratory and filtered through 0.40 μm polycarbonate filters. The filtrates were cryopreserved at −20 °C, and the filters were dried at 45 °C and weighed to obtain the SPM.

Nutrients in the filtrates were analyzed using a QuAAtro Continuous–Flow Automatic Analyzer (SEAL Analytical GmbH, Norderstedt, Germany), and the detection limits were 0.01, 0.01, 0.02, 0.01, and 0.04 μM for NO_3^-, nitrite (NO_2^-), ammonium (NH_4^+), DIP, and DSi, respectively. Total dissolved nitrogen (TDN) and phosphorus (TDP) were measured via the boric acid–persulfate oxidation method (Grasshoff et al., 1999). DIN was calculated as the sum of NO_3^-, NO_2^-, and NH_4^+. Dissolved organic nitrogen (DON) was calculated as the difference between TDN and DIN, and dissolved organic phosphorus (DOP) was calculated as the difference between TDP and DIP (Liu et al., 2012).

2.3. Data processing and statistics

The original data of nine provinces in the Yellow River basin were processed using the following equation:

$$Q = \sum Q_{ai} \times A_i, \tag{1}$$

where Q is the data of the Yellow River basin; Q_{ai} is the original data in each province; A_i is the area proportion of each province in the Yellow River basin of the total province area. Other data processing such as precipitation, recharged water, sediment load, fertilizer application, sewage effluents, and industrial structure of nutrient fluxes in the Yellow River were shown in the supplementary material (Methodology in Appendix).

A trend was defined as the monotonic variation (either abruptly or gradually) in the concentration or factors with time. $P < 0.05$ was considered statistically significant. A one–way analysis of variance (ANOVA) test was performed for seasonal nutrient concentrations with a significance of $p < 0.05$. The correlation between the variables was determined by Pearson correlation. The statistical analysis was performed by Sigmaplot 12.5.

3. Results

3.1. Water discharge and sediment load

The water discharge and sediment load at station LJ from 2001 to 2017 were 0.77–68.3 × 10^8 m^3/month and 3.39–16,150 × 10^4 t/

month, respectively (Fig. 2). We observed a strong correlation between the water discharge and sediment load ($R^2 = 0.72$, $P < 0.00001$), with the highest values occurring from June to October (maximum in July at 80%; Fig. 2). The monthly water discharge and sediment load were highest between 2003 and 2013 relative to 2001–2002 and 2014–2017. We compared the annual water discharge and sediment load at station LJ to stations TG and HYK (Fig. B.1 in Appendix B). The observed correlations indicate that the water discharge and sediment load at station LJ are controlled by both the transport fluxes from the upper and middle reaches and by the regulation of the SMX and XLD reservoirs due to the implementation of the WSRS.

3.2. Nutrient concentrations at station Lijin

The concentrations of nitrogen compounds were 105–524 μM for DIN, 0.0–268 μM for DON, and 122–551 μM for TDN. DON accounted for less than 56% of the TDN. DIN and TDN concentrations were highest in winter and spring, and DON concentrations were highest in summer and fall (Fig. 3). In long–term observation of previous studies from 1980 to 2012, there was an increasing trend in annual DIN concentration (Yu et al., 2010; Chen et al., 2013; Ma et al., 2015); yet during this study period, the concentrations of DIN and TDN decreased linearly at a rate of approximately 0.30 μM/month ($P < 0.005$, $R^2 = 0.07$) and 0.50 μM/month ($P < 0.00001$, $R^2 = 0.18$) from 2001 to 2018, respectively. This decrease was particularly pronounced from 2014 onwards; DIN reached as low as 105 μM (Fig. 3). In contrast, the minimum DON concentration (DON_{min}) and $(DON/TDN)_{min}$ increased from 2001 to 2018 (Fig. B.2 in Appendix B).

The concentrations of phosphorus compounds were 0.03–0.95 μM for DIP, 0.05–0.59 μM for DOP, and 0.24–1.28 μM for TDP. The DIP and TDP concentrations were highest in spring and decreased linearly with time at equal rates of 0.001 μM/month ($P < 0.00001$, $R^2 = 0.16$ and $R^2 = 0.12$, respectively). The variation in DIP was consistent with results reported by Yu et al. (2010) and Ma et al. (2015). While there are fewer reports about DOP concentrations (Chen et al., 2013; Liu, 2015), especially in long time series. In this study, DOP concentrations increased linearly from 2009 to 2018 at a rate of 0.002 μM/month ($P < 0.00001$, $R^2 = 0.22$) (Fig. 3), coinciding with an increase in both DOP_{min} and $(DOP/TDP)_{min}$ (Fig. B.2 in Appendix B). The DOP represented 9–58% of the TDP in 2001–2014 and 90–95% of the TDP from 2015 onwards.

DSi concentrations were 0.5–167 μM and declined linearly with time at a rate of 0.31 μM/month ($P < 0.00001$, $R^2 = 0.37$). The long–term

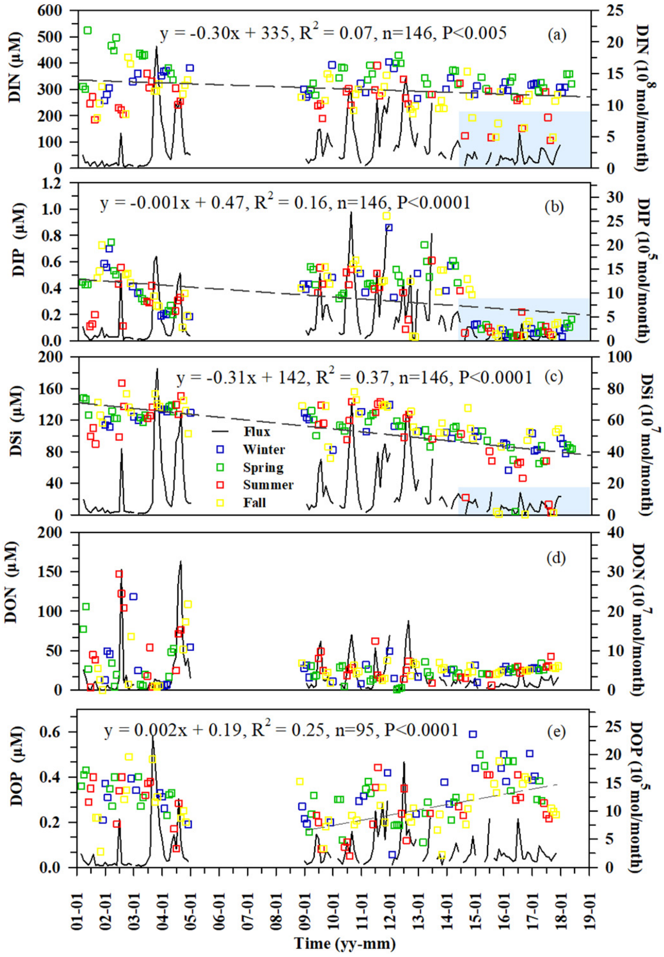

Fig. 3. Monthly concentrations and fluxes of dissolved nutrients in the lower Yellow River from March 2001 to May 2018. Data for 2001–2004 are from Yao et al. (2009), Zhang et al. (2010), and Gong et al. (2015) and the remaining data are from our observations (Chen et al., 2013; Liu et al., 2012; Wu et al., 2017; and this study). DIN (a), DIP (b) and DSi (c) concentrations decreased from 2001 to 2018 at rates of 0.27 μM/month ($P < 0.005$, $R^2 = 0.07$), 0.001 μM/month ($P < 0.00001$, $R^2 = 0.16$), and 0.29 μM/month ($P < 0.00001$, $R^2 = 0.36$), respectively. DOP (e) concentrations increased at a rate of 0.002 μM/month ($P < 0.00001$, $R^2 = 0.22$) from 2009 to 2017.

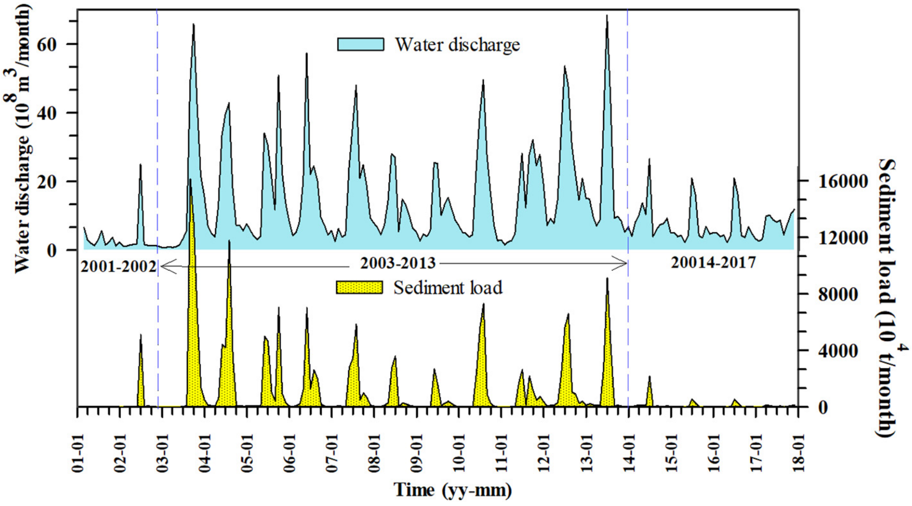

Fig. 2. Monthly water discharge and sediment load at station Lijin from 2001 to 2017. The red and green lines indicate the monthly average water discharge and sediment load, respectively. Data were sourced from the Yellow River Water Resources Bulletin (YRWRB, 2001–2017), the Yellow River Sediment Bulletin (YRSB, 2006–2017), and Wang et al. (2016). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1
Relationships (r) between water discharge (m^3/month), sediment load (t/month), nutrient concentrations (μM), and nutrient fluxes (FDIN, FDIP, FDSi, FDON, FDOP: mol/month) from 2001 to 2018.

	Sediment load	CDIN	CDIP	CDSi	CDON	CDOP	FDIN	FDIP	FDSi	FDON	FDOP
Water discharge	0.847**	−0.165	0.087	0.277*	−0.076	−0.239*	0.972**	0.807**	0.977**	0.628**	0.847**
Sediment load		−0.114	0.029	0.318*	0.008	−0.091	0.802**	0.602**	0.867**	0.551**	0.783**
CDIN			0.314*	0.414**	−0.201*	0.115	−0.005	−0.023	−0.098	−0.212*	−0.112
CDIP				0.550**	0.044	−0.394**	0.150	0.493**	0.151	0.106	−0.018
CDSi					0.191*	−0.133	0.327**	0.372**	0.412**	0.289*	0.229*
CDON						0.041	−0.109	−0.041	−0.036	0.356**	−0.096
CDOP							−0.215*	−0.317**	−0.218*	−0.24*	0.144
FDIN								0.825**	0.960**	0.556**	0.853**
FDIP									0.820**	0.559**	0.622**
FDSi										0.650**	0.840**
FDON											0.429**

Note: $N = 128–140$.
* Represents $P < 0.05$.
** Represents $P < 0.0001$.

variation of the DSi corresponded that of with Ma et al. (2015) and Ran et al. (2015). Nevertheless, the decline in DSi was more obvious. Extremely low concentrations were observed for the first time in this study, reached as low as 0.52 μM since 2014. We did not observe significant seasonal variability in DSi concentrations throughout the study period (Fig. 3).

3.3. Nutrient transport fluxes to the adjacent Bohai Sea

We estimated the monthly nutrient fluxes using monthly water discharge data and nutrient concentrations (Parcom, 1988). The monthly fluxes of DIN, DIP, DSi, DON, and DOP were $0.28–19.3 \times 10^8$, $0.07–26.9 \times 10^5$, $0.02–92.5 \times 10^7$, $0.04–32.7 \times 10^7$, and $0.22–23.3 \times 10^5$ mol/month at station LJ in the lower Yellow River from 2001 to 2017, respectively (Fig. 3). High nutrient fluxes predominantly occurred between June and September, coinciding with peaks in the water and sediment loads due to their significant correlations (Table 1). The monthly nutrient fluxes were highest between 2003 and 2013 and decreased in 2014–2017 (Fig. 3). Both nutrient concentrations and fluxes at station LJ decreased since 2014. The causes for this shift will be discussed in Section 4.

4. Discussion

4.1. Causes of low nutrient concentrations

In this study, we identified the occurrence of notably low nutrient concentrations in the lower Yellow River. Concentrations of DIN, DIP, and DSi reached as low as 105 ± 3.2 μM, 0.03 ± 0.01 μM, and 0.52 ± 0.23 μM, and accounted for 34%, 0.5%, and 9.1% of the 2001–2018 average concentrations, respectively. It was observed for the first time in the Yellow River. ANOVA inferred that the extremely low DSi concentrations were not controlled by seasonal variability ($P = 0.37$, $n = 39–42$). However, we observed significant seasonal changes in the DIN ($P < 0.05$, $n = 39–42$) and DIP concentrations with most values below 0.2 μM from 2015 to 2017. We considered DSi values of <45% of the 2001–2018 mean concentration to be extremely low nutrient events. Nine extremely low nutrient concentration events have occurred since 2014 (Table 2), with DIN and DIP concentrations accounting for 35–89% and 9–67% of the 2001–2018 mean concentrations, respectively.

All of the nine low nutrient concentration events—excluding those in May 2016—occurred during the impoundment of the Xiaolangdi Reservoir as water levels began to increase (Fig. 4). The retention effect of large reservoirs in response to increased water storage substantially reduced both the downstream water discharge and sediment load (Wang et al., 2007, 2016; Wang et al., 2017; Bi et al., 2019). During this period, stations XLD and LJ had relatively low discharge with rates of 195–600 m^3/s and 83–440 m^3/s, respectively; and the SPM at LJ was 40–200 mg/L or only 1–6% of the 2001–2017 average SPM (3342 mg/L). Furthermore, low concentrations of both DSi and DIP were positively correlated with runoff and SPM (Fig. B.3 in Appendix B). However, DIN showed no significant correlation with water discharge or SPM, despite relatively low concentrations during the nine events.

It has been reported that in low water discharge and SPM periods, phytoplankton reproduction may be enhanced in the Yellow River (Wang et al., 2012; Ran et al., 2015). The dominant phytoplankton species in the mainstream of the Yellow River is Bacillariophyta (Wang et al., 2010), and the proportion of diatoms in downstream Lijin can reach 53% (Wang et al., 2012). Diatoms have an absolute silicon requirement and other nutrients for growth (Nelson and Brzezinski, 1990). In this study, the SPM and runoff at station LJ were relatively low during all nine low nutrient concentration events, which created favorable conditions for phytoplankton proliferation and resulted in the synchronous decrease of DIN, DSi, and DIP.

Table 2
The occurrence (year and month) of relatively low nutrient concentration (μM) events and their corresponding water discharge (m^3/s) and SPM (mg/L). The long-term (2001–2018) averages are provided for comparison.

Time	Water discharge	SPM	DSi	DIP	DIN
2014/08	154	140 ± 30	22.1 ± 0.4	0.06 ± 0.01	123 ± 0.3
2015/09	131	40 ± 10	4.13 ± 0.03	0.05 ± 0.00	118 ± 1.2
2015/10	102	90 ± 00	1.04 ± 0.05	0.03 ± 0.00	169 ± 0.1
2016/05	83	100 ± 30	4.26 ± 0.91	0.03 ± 0.01	269 ± 4.1
2016/08	440	200 ± 70	46.5 ± 1.7	0.22 ± 0.03	152 ± 4.2
2016/09	121	60 ± 10	0.52 ± 0.23	0.04 ± 0.00	152 ± 2.8
2017/07	231	150 ± 20	13.1 ± 0.70	0.07 ± 0.02	194 ± 1.4
2017/08	239	90 ± 20	3.0 ± 0.10	0.05 ± 0.01	105 ± 3.2
2017/09	189	100 ± 30	2.9 ± 0.10	0.04 ± 0.00	118 ± 0.6
2001–2018	519 ± 653	3342 ± 6595*	107 ± 33	0.33 ± 0.21	302 ± 75

* Represents that average SPM is from 2001 to 2017.

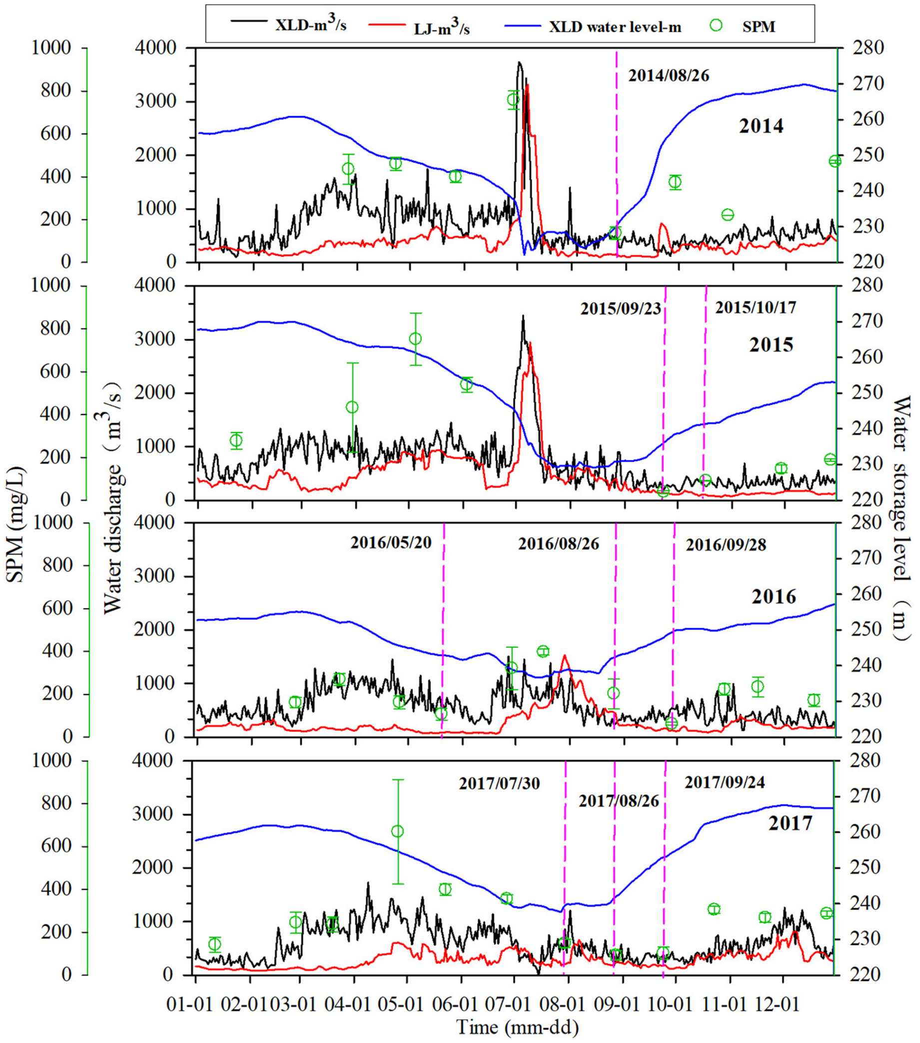

Fig. 4. Daily water discharge (m^3/s) at stations Xiaolangdi and Lijin (LJ), daily water storage level (m) of the Xiaolangdi Reservoir (XLD), and SPM on the day of sampling at station LJ from 2014 to 2017. The pink line represents the nine low nutrient concentration events. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

DOP concentrations increased from 2009 to 2018 (Fig. 3), and the proportion of DOP in TDP exceeded 80%. In contrast, DIP concentrations in the lower reaches of the Yellow River continuously decreased from 2001 to 2018, and more than a half of concentrations were less than 0.10 μM from 2015 onwards. It has been reported that there is a limitation for phytoplankton growth when DIP lower than 0.10 μM (Nelson and Brzezinski, 1990), and DOP is an important source of phosphorus for phytoplankton growth in DIP–deficient waters (Björkman and David, 2003; Dyhrman et al., 2006; Mather et al., 2008). Statistically, species of Pyrroptata that readily uptake DOP are significantly more abundant than other algae (Jin and Liu, 2013). Therefore, changes in nutrient structures may significantly impact the phytoplankton community structure in the ecosystems of the lower Yellow River and adjacent Bohai Sea.

4.2. Factors for long–term nutrient flux variability

4.2.1. Natural factors influencing the nutrient characteristics

Natural factors affecting nutrient concentrations and fluxes mainly include natural runoff, precipitation, sediment transport. Station TNH is the first gauging station in the upper reaches of the Yellow River and is therefore less affected by anthropogenic activities and industrial development. We therefore considered water discharge at station TNH to reflect the natural runoff from the Yellow River's source. The influence of natural runoff on nutrient fluxes in the Yellow River was estimated under the condition that nutrient concentrations at station TNH remained unchanged while water discharge varied. From 2001 to 2017, the annual water discharge at station Tangnaihai was 11–28 km^3/a (YRWRB, 2001–2017) and decreased after 2013. DIN, DIP, and DSi concentrations at station TNH were 47 μM, 0.002 μM, and 88 μM, respectively (Ma et al., 2015). Hence, 0.696–1.87 × 10^4 t/a of DIN, 0.001–0.002 × 10^3 t/a of DIP, and 2.61–7.00 × 10^4 t/a of DSi were influenced by annual water discharge in the Yellow River during 2001–2017.

Precipitation largely influences the hydrology and water resources of river water (Wang et al., 2006); it is also a significant pathway of nutrients from the atmosphere to rivers and oceans (Migon and Sandroni, 1999; Zhang, 1994; Galloway, 2005; Ding et al., 2011; Han et al., 2013). We calculated the annual precipitation load into the Yellow River according to the total precipitation in the basin and the proportion of surface area of the river course in the total basin area (Appendix A1). The annual precipitation loads were 5.74–7.89 km^3/a during 2001–2017 and accounted for 1.79% of the total annual precipitation in the Yellow River basin (321–442 km^3/a). Combining the precipitation and nutrient concentrations in rainwater, the wet deposition fluxes to the Yellow River were 0.80–1.64 × 10^4 t/a for DIN, 0.023–0.159 × 10^3 t/a for DIP,

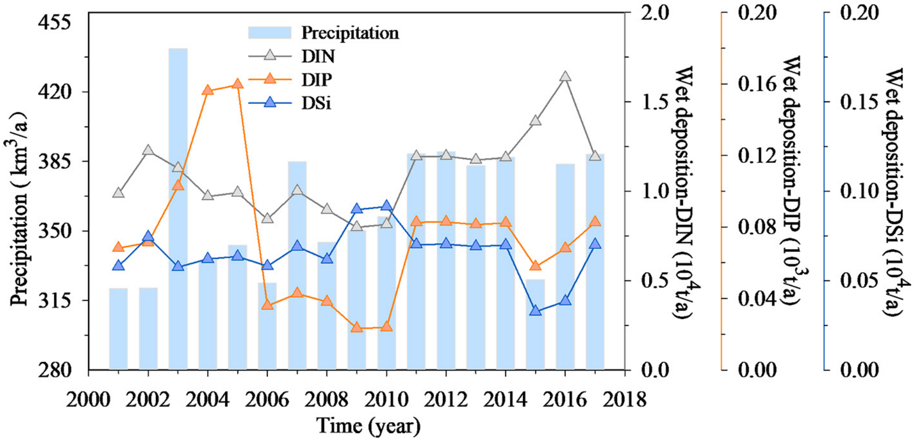

Fig. 5. Precipitation and wet deposition fluxes of DIN, DIP, and DSi to the Yellow River from 2001 to 2017. Yellow River basin precipitation data were accessed from the YRWRB, 2001–2017. The wet deposition of nutrients was estimated by the precipitation and rainwater concentrations. DIN, DIP, and DSi concentrations were 152 μM, 0.40 μM, and 4.63 μM in 2002; 102 μM, 0.42 μM, and 2.60 μM in 2003 (Song, 2006); 116 μM 0.84 μM, and 3.70 μM in 2004–2005 (Bi, 2006); 104 μM, 0.20 μM, and 3.58 μM in 2006–2008 (Jiang, 2009); 91 μM, 0.12 μM, and 5.12 μM in 2009–2010 (Zhu, 2011); and 171 μM, 0.32 μM, and 2.00 μM in 2015–2016 (Xing, 2017), respectively. DIN, DIP, and DSi concentrations in 2001, 2011–2014 and 2017 are average concentrations of above years (2002–2010 and 2015–2016), 123 μM, 0.38 μM and 3.61 μM, respectively.

and 0.033–0.091 × 10^4 t/a for DSi (Fig. 5, Appendix A1). In overall trends from 2001 to 2018, the DIN wet deposition flux increased, while the DSi and DIP wet deposition fluxes declined (Fig. 5). However, it is important to note that the estimations of the wet deposition flux may have large uncertainties due to the limited availability of data on rainwater nutrient concentrations.

Nutrients such as DIN, DIP, and DSi can also be leached from soil (Ding et al., 2011; Dong et al., 2019; Lu et al., 2019; Liu et al., 2019). The sediment load was correlated with each nutrient flux in the lower Yellow River ($R^2 = 0.36$–0.75, $p < 0.0001$, $n = 140$). The sediment load from the Loess Plateau to the Yellow River was estimated for 2001–2017 by calculating the annual sediment load difference between stations TG and TDG (Wang et al., 2016); the values ranged from 0.291 × 10^8 t/a to 5.84 × 10^8 t/a and showed a downward trend, with the lowest value in 2014 (Fig. 6). The average background soil nutrient contents of the Loess Plateau were replaced by the Fenglingdu soil at 8.70 μg/g for DIN, 0.162 μg/g for DIP, and 22.1 μg/g for DSi (Ma et al., 2015). Therefore, the DIN, DIP, and DSi fluxes from the Loess Plateau to the Yellow River were estimated to be 0.025–0.508 × 10^4 t/a, 0.005–0.095 × 10^3 t/a, and 0.064–1.29 × 10^4 t/a in 2001–2017, respectively (Fig. 7, Appendix A2); all the values decreased over time (Fig. 7).

4.2.2. Anthropogenic factors influencing the nutrient characteristics

The Yellow River basin 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). Agricultural activities include irrigation and fertilization. Intensive agricultural irrigation in the drainage basin—where recharged water re–enters the river and circulates between the river and soil—could significantly increase the major ion concentrations and elevate nutrient levels (Chen et al., 2006; Cai et al., 2008; Fan et al., 2014). This is caused by the increased evaporation of water (Ding et al., 2011) and the loss of agricultural fertilizer by leaching and irrigation (Zhu and Chen, 2002; Liu et al., 2013).

The variations of water diverted from and recharged back to the Yellow River remained almost the same throughout 2001–2011. However, the recharged water declined and the diverted water remained stable throughout 2012–2017 (Fig. 8). The influence of recharged water on the DIP and DSi concentrations were calculated (Appendix A1) based

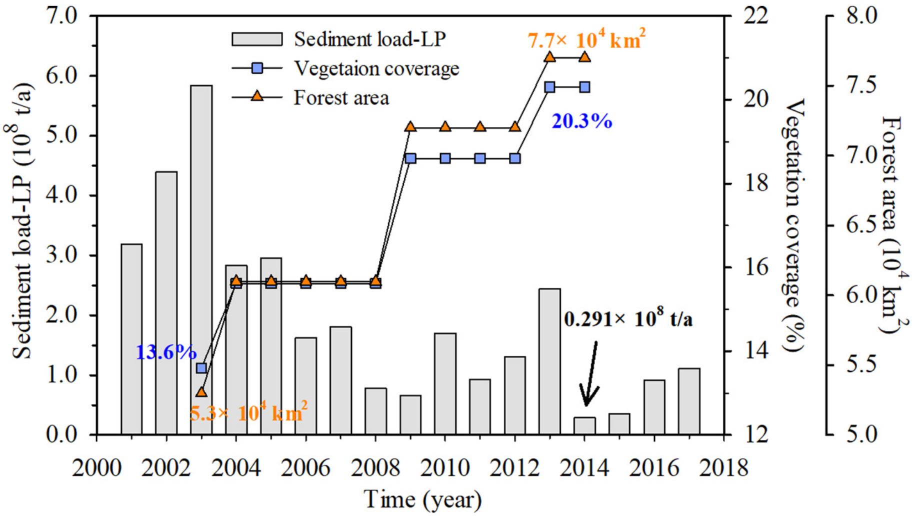

Fig. 6. Sediment load from the Loess Plateau (LP) to the Yellow River from 2001 to 2017 calculated as the difference between stations Toudaoguai and Tongguan. The data were accessed from the Yellow River Sediment Bulletin ((YRSB) Yellow River Water Resources Conservation Commission. Yellow River Sediment Bulletin, 2006–2017) and Wang et al. (2016). The average vegetation coverage and forested area of the nine provinces in the Yellow River basin from 2003 to 2014 were obtained from the China Forestry Database (http://www.forestry.gov.cn/data.html).

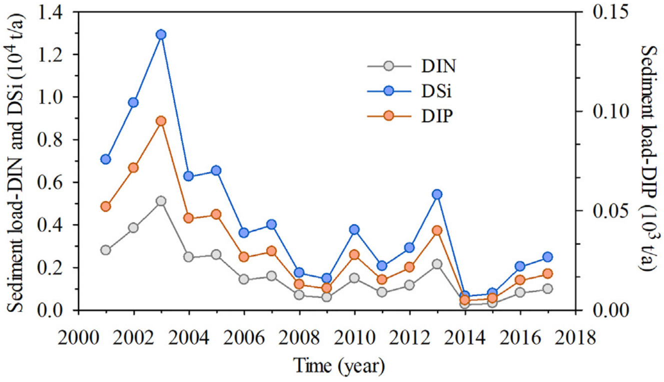

Fig. 7. DIN, DIP, and DSi fluxes to the Yellow River from the Loess Plateau sediment during 2001–2017.

on the relationships between the recharged water and the concentrations of DIP and DSi in the lower reaches of the Yellow River (average relative deviation of 16% and 0.2%, respectively). Combining the calculated concentrations with the water discharge at station LJ (Appendix A1), the DIP and DSi fluxes from recharged water to the Yellow River were estimated to be 0.044–0.326 × 10^3 t/a and 1.48–9.23 × 10^4 t/a during 2001–2017, respectively. Using the same method as the river flux calculations, we estimated the 2001–2017 DIN fluxes to the Yellow River to be 0.80–1.07 × 10^4 t/a based on the recharged water value and the average DIN concentrations of shallow groundwater within the irrigation region of the basin (Appendix A1). We observed a decrease in the DIN flux after 2011. The average DIN concentration was represented by the shallow groundwater NO_3^- concentrations (6–282 μM) within the irrigation area of the basin (Wang et al., 2014); this would lead to a 0.2–40% underestimation in DIN flux due to the influence of NH_4^+ and NO_2^- (Wang et al., 2014).

Fertilizer application from intense agriculture significantly influences the nutrient concentrations and fluxes (Smith et al., 2003; Liu et al., 2009, 2012; Yu et al., 2010; Liu et al., 2013; Gong et al., 2015; Liu, 2015) to the Yellow River basin. The total annual nitrogen and phosphorus fertilizer application in the nine provinces were 732–816 × 10^4 t/a and 263–317 × 10^4 t/a during 2001–2017, respectively (NBSC, http://data.stats.gov.cn/). Total N and P fertilizer applications were significantly related to the sown area, grain production, and fruit production in all nine provinces (Fig. B.4 in Appendix B). Combining the fertilizer applications of the nine provinces with the proportional area of each province (Appendix A3), the calculated N and P fertilizer application in the Yellow River basin were 167–199 × 10^4 t/a and 60–70 × 10^4 t/a, respectively. The contents of N and P in fertilizers are 0.352 and 0.105 (China Chemical Industry Yearbook (CCIY), 1999; Gong et al., 2015), respectively; and the percentage loss of nitrogen and phosphate during fertilizer application in the Yellow River basin was found to be 15% and 2.0%, respectively (Zhang and Shao, 2000; Gong et al., 2015). Fertilizer leaching was therefore estimated to contribute 8.85–10.5 × 10^4 t/a of DIN and 1.27–1.47 × 10^3 t/a of DIP to the Yellow River during 2001–2017 (Appendix A3). From 2001 to 2013, the DIN and DIP fluxes to the river from fertilizer application increased at a rate of 0.14 × 10^4 t/a ($R^2 = 0.99$, $P < 0.0001$) and 0.02 × 10^3 t/a ($R^2 = 0.92$, $P < 0.0001$) (Fig. 9), respectively. From 2014 onwards, DIN and DIP fluxes to the river from fertilizer application declined at a rate of 0.31 × 10^4 t/a ($R^2 = 0.98$, $P < 0.01$) and 0.04 × 10^3 t/a ($R^2 = 0.90$, $P < 0.05$) (Fig. 9), respectively, with the highest value in 2013. The MOA's policy for conducting soil testing to determine appropriate fertilizer formulas prior to application resulted in a 10% increase in the fertilizer utilization rates and caused a decrease in the application of N and P fertilizers. Moreover, the continuous improvement of agricultural cultivation technology has lowered the demand for chemical fertilizers (Fang and Meng, 2013). Due to the growing environmental impacts of excessive fertilization, the use of organic fertilizers in place of chemical fertilizers has been gradually increasing (Zhang et al., 2008; Wang et al., 2019).

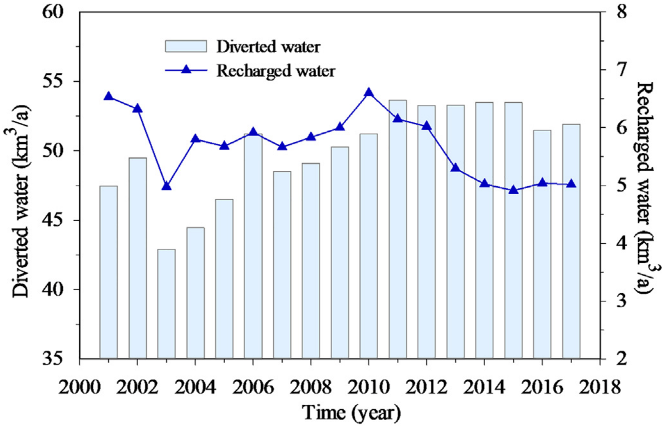

Fig. 8. Diverted water and recharged water in the Yellow River basin from 2001 to 2017. Data were accessed from the YRWRB.

Vegetation coverage increased in response to the initiation of the "Gain–for–Green" Program (GFGP) in 1999–a scheme to restore degraded ecosystems, stabilize soils, and minimize erosion (Feng et al., 2016; Wang et al., 2016). From 2003 to 2014, the average forested area and vegetation coverage in the nine provinces of the Yellow River drainage basin both increased from 5.3 × 10^4 km^2 to 7.7 × 10^4 km^2 and 13.6% to 20.3%, respectively (Fig. 6). The average vegetation coverage in all nine provinces of the Yellow River basin was negatively correlated to sediment load of the Loess Plateau ($R^2 = 0.39$, $p < 0.05$, $n = 12$). We also identified a deceasing trend in the sediment load of the Loess Plateau with increasing vegetation coverage on the Yellow River basin. Increased vegetation coverage resulted in the reduction of soil erosion (Wang et al., 2007), and contributed to approximately 57% of the total reduction in average Loess Plateau sediment transport (Wang et al., 2016). This accounted for a 0.166–3.33 × 10^8 t/a sediment load to the Yellow River from 2001 to 2017. Hence, the fluxes of DIN, DIP, and DSi transported to the Yellow River from 2001 to 2017 as a result of increasing vegetation coverage were 0.014–0.290 × 10^4 t/a, 0.003–0.054 × 10^3 t/a, and 0.037–0.735 × 10^4 t/a, respectively.

4.2.3. Impacts of social and industrial development on nutrient characteristics

Rapid population growth and social development in China has led to an increase in fertilizer application and sewage effluents, which has deteriorated the water quality and impacted the biogeochemical cycling of nutrients in Chinese rivers (Jin and Guo, 1996; Zhang et al., 1999; Xia et al., 2001; Liu et al., 2003; Gong et al., 2015; Liu, 2015). Nevertheless,

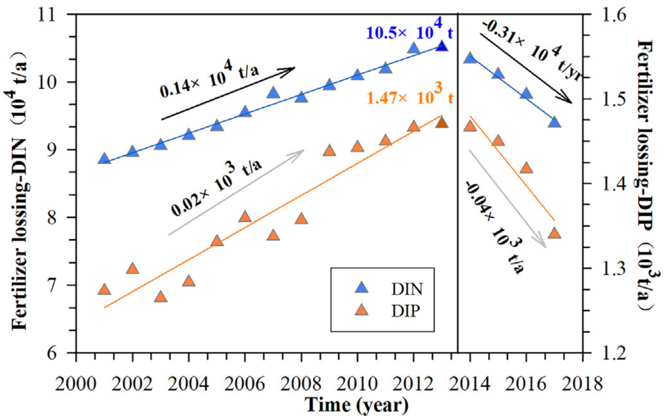

Fig. 9. DIN and DIP fluxes to the Yellow River from N and P fertilizer loss in 2001–2017.

nutrient concentrations and fluxes in the lower Yellow River have decreased in recent years—particularly since 2014 (Fig. 3)—which may be related to variations in natural runoff, precipitation (wet deposition), sediment load, vegetation coverage, recharged water, and fertilizer application as well as sewage effluents, urbanization, and industrial structure.

The amount of sewage effluents discharged to the Yellow River was in the range of 4.13–4.53 × 10^9 t/a (YRWRB, 2001–2017). Sewage effluents consist of municipal, secondary industry, and tertiary industry sewage, accounting for 20–39%, 50–74%, and 6–11% of the total sewage effluents, respectively (Fig. 10). Municipal and tertiary industry sewage increased and secondary industry sewage decreased with time. This led to an increase in total sewage effluents during 2001–2011, followed by a slight decrease from 2012 to 2017 (Fig. 10). The nutrient concentrations of untreated wastewater sources were 4286 μM and 290 μM for DIN and DIP, respectively, and the concentrations of treated wastewater sources were 571 μM and 25 μM for DIN and DIP, respectively (Gong et al., 2015). More than 80% of the total wastewater exceeded the discharging standard (MOEE, 2001–2015). Thus, the annual average DIN and DIP fluxes from sewage effluents to the Yellow River were estimated to be in the range of 7.37–8.33 × 10^4 t/a and 9.65–10.9 × 10^3 t/a during 2001–2017 (Appendix A4), respectively.

From 2005 to 2016, the rural population of the nine provinces in the Yellow River basin decreased by approximately 15–28%, and the urbanization rate increased by 31–64% (Fig. B.5 in Appendix B). The urbanization rate was negatively correlated with DIN, DIP, and DSi fluxes ($R^2 = 0.47–0.67$, $P < 0.05$, $n = 9$) at station LJ based on log10 relationships, which demonstrates a decrease in nutrient fluxes in response to increased urbanization. However, we also identified relationships between urbanization and fertilizer application, sewage effluents, and vegetation coverage (Fig. B.6 in Appendix B), highlighting the potential mechanisms behind the influence of urbanization on Yellow River nutrient characteristics.

We also identified positive correlations between GDP and sewage effluents and vegetation coverage (Fig. B.7 in Appendix B). However, the sustainable growth of GDP led to an initial increase followed by a subsequent decline in fertilizer application (Fig. B.7 in Appendix B); and this variability is also consistent with the observed changes in other countries globally, including France, the United Kingdom, South Korea, and Japan (Wang et al., 2019). The relationships between GDP and fertilizer application, sewage effluents, and vegetation coverage are similar to their relationships with urbanization (Fig. B.6–7 in Appendix B). It is therefore likely that changes in GDP indirectly affect the variations in nutrient concentrations or fluxes to the river.

With China's reform and opening up over the last twenty years, the total GDP of the nine provinces experienced exponential growth during 2001–2017 from 2.65 trillion/a to 22.1 trillion/a (Fig. 11). The industrial structure in the nine provinces of the Yellow River basin has also changed significantly, and the proportion of primary industries—including farming, forestry, animal husbandry, and fisheries—of the total GDP decreased from 18% in 2001 to 8% in 2017 (Fig. 11, Fig. B.8 in Appendix B). The proportion of secondary industries rapidly developed at a rate of 1.0%/a until 2011 and accounted for 55% of the total GDP (Fig. 11). However, the proportion of tertiary industries—including wholesale–retail, financial, and others—developed rapidly (2.3%/a) since 2012, reaching close to or even exceeding the proportion of secondary industries. The proportion of tertiary industries reached a maximum of 47% in 2017 (Fig. 11, Fig. B.8 in Appendix B). The growth of secondary industries in the Yellow River basin was positively correlated with DIN, DIP, and DSi fluxes ($R^2 = 0.62–0.77$, $P < 0.05$, $n = 13$) at station LJ in the lower Yellow River. In contrast, we observed negative relationships between tertiary industry growth and DIN, DIP, and DSi fluxes ($R^2 = 0.29–0.73$, $P < 0.05$, $n = 13$). We therefore estimated the influence of the changing industrial structure on nutrient fluxes through the GDP proportion of the different industries and the relationships between nutrient fluxes and industrial structure (Appendix A5). We found that the changes in industrial structure from 2001 to 2017 contributed 2.20–8.40 × 10^4 t/a of DIN, 0.024–0.322 × 10^3 t/a of DIP, and 1.36–7.47 × 10^4 t/a of DSi; all fluxes decreased after 2012 (Fig. 12).

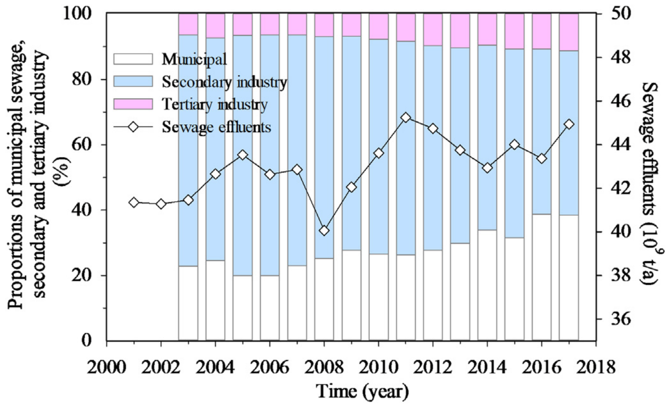

Fig. 10. The discharge of sewage effluents to the Yellow River during 2001–2017, and the proportions of municipal, secondary industry, and tertiary industry sewage of the total sewage effluents. Data were accessed from the YRWRB.

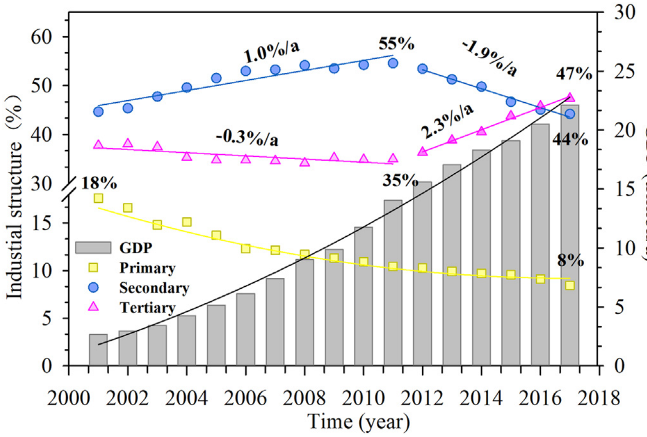

Fig. 11. Total GDP and industrial structure of the nine provinces in the Yellow River basin from 2001 to 2017.
(The National Bureau of Statistics of China, http://data.stats.gov.cn/).

4.2.4. Relative contributions of controlling factors to the total nutrient fluxes to the Yellow River

We identified negative normalized anomalies for DIN, DIP, and DSi fluxes at station LJ during 2001–2002 and 2014–2017 (Fig. 13, Appendix A6), inferring relatively low nutrients fluxes to the Bohai Sea. Negative total normalized anomalies for DIN, DIP, and DSi fluxes to the Yellow

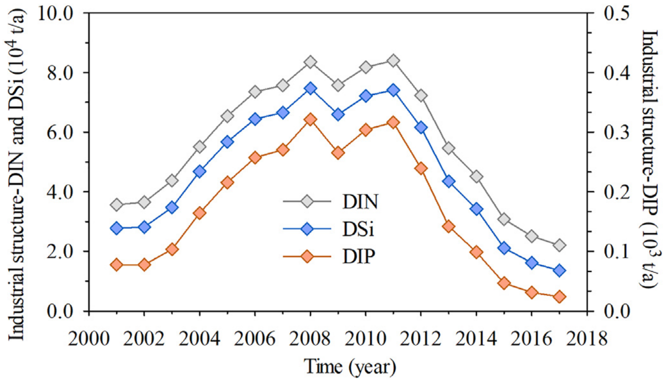

Fig. 12. DIN, DIP, and DSi fluxes to the Yellow River in response to changes in industrial structure during 2001–2017.

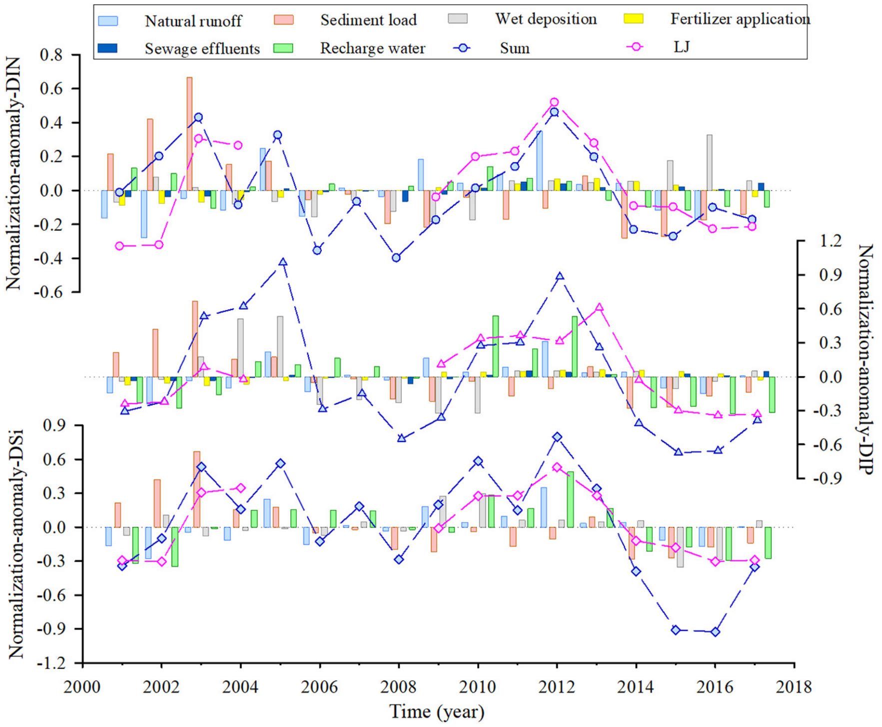

Fig. 13. Color bars represent the normalized anomaly analysis of DIN, DIP, and DSi fluxes to the Yellow River, including natural runoff, sediment load, wet deposition, fertilizer application, sewage effluents, and recharged water. The blue line indicates the sum of all normalized anomalies for each factor, and the pink line represents the normalized anomaly of all fluxes at station Lijin. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

River were also identified from 2001 to 2002 and 2014 to 2017 (Fig. 13). The overall normalized anomaly trends of the different nutrient fluxes at station LJ were consistent and correlated with the external fluxes to the Yellow River ($R^2 = 0.52–0.81$, $P < 0.05$, $n = 13$). This finding confirms that nutrient fluxes to the Bohai Sea at station LJ are controlled by the external input sources of nutrients to the Yellow River. The annual total fluxes of DIN, DIP, and DSi to the Yellow River—including natural runoff, precipitation (wet deposition), sediment load from the Loess Plateau, recharged water, fertilizer loss, and sewage effluents—were 19.7–22.9 × 10^4 t/a, 11.2–12.7 × 10^3 t/a, and 5.13–16.6 × 10^4 t/a, respectively, from 2001 to 2017. The annual fluxes of DIN, DIP, and DSi to the Bohai Sea at station LJ were 1.85–12.2 × 10^4 t/a, 0.019–0.391 × 10^3 t/a, and 1.50–9.05 × 10^4 t/a and account for 9.4–53%, 0.16–3.2%, and 23–70% of the total annual nutrient influx to the Yellow River, respectively (Fig. 14). Water consumptions, retention caused by dam construction, and phytoplankton growth in the Yellow River could all influence the losses of DIN, DIP and DSi (Ding et al., 2011; Maavara et al., 2020; Ran et al., 2015). Additional mechanisms for nutrient losses include denitrification for DIN, and adsorption–desorption in sediment for DIP (Strokal et al., 2017). Variations in

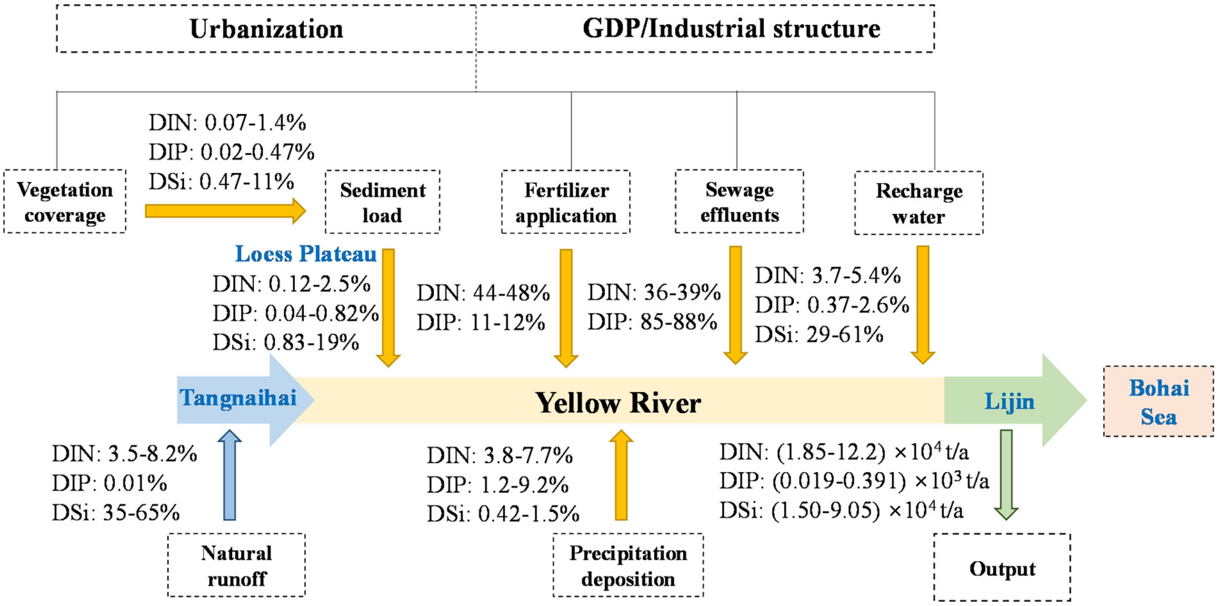

Fig. 14. Relative contributions of various factors toward the total DIN, DIP, and DSi fluxes to the Yellow River.

available nutrient sources could lead to different output rates. Fluxes of DIN, DIP, and DSi from natural runoff, fertilization, and recharged water initially increased from 2001 to 2017, and then decreased from 2013 to 2014. We also observed a continuous decrease in the sediment load nutrient fluxes to the Yellow River. The flux of DIN from wet deposition and the fluxes of DIN and DIP from sewage effluents have increased overall, while wet deposition fluxes of DIP and DSi have declined since 2006 and 2013, respectively. The sources causing the highest impact on total input fluxes of DIN, DIP, and DSi were fertilizer losing (44–48%), sewage effluents (85–88%), runoff (35–65%), respectively (Fig. 14).

In this study we did not estimate all the possible controlling factors on total nutrient influxes to the Yellow River, such as dry deposition, animal manure, groundwater, and water evaporation. Atmospheric dry deposition emptied into the Yellow River was roughly estimated using the dry deposition flux (Zhu, 2011; Xing, 2017) and surface area of the river course (approximately 1.42 km^2); there were approximately 2.36–2.94 $\times 10^4$ t/a for DIN, 0.014–0.054 $\times 10^3$ t/a for DIP, 0.034–0.053 $\times 10^4$ t/a for DSi. However, we did not include these values in the total nutrient fluxes to the Yellow River because the dry deposition flux may have large uncertainties due to the limited availability of data. In addition, animal manure is also one of the important sources for N and P (Strokal et al., 2017). In 2006, 1657 $\times 10^4$ t/a of total N and 1171 $\times 10^4$ t/a of P_2O_5 were produced from animal manure in China (Jia, 2014), however the nutrient losses of animal manure into the Yellow River were not estimated in this study. Finally, it should be noted that water evaporation and groundwater were not included in the total nutrient fluxes into the Yellow River—due to either complex variability or a lack of available data. Although a massive volume of underground water emptied into the Yellow River and could have been as much as 24–45 km^3, according to the river's water discharge balance (Fig. B.9 in Appendix B). Therefore, further research would be required to understand and quantify the effects of complicated factors on nutrient inputs to the Yellow River.

5. Conclusions

This study assessed the dominant controls on the variations in nutrient concentrations and fluxes to the lower Yellow River. We observed decreasing trends in dissolved inorganic nutrient concentrations. DOP concentrations increased after 2009, reaching up to 95% of the TDP. Annual minimum concentrations of DON also increased with time. We identified extremely low concentration events after 2014 due to the retention effect of the Xiaolangdi Reservoir, which significantly decreased the water discharge and sediment load downstream, and then caused an increase in phytoplankton nutrient uptake.

In recent years, the nutrient input to the Yellow River from external sources generally increased first and then decreased. These trends may have been influenced by the continuous promotion of protection policies made by the Chinese government, regulators of the Yellow River basin and an increase in environmental awareness among people. The dominant controlling factors of the total nutrient influxes to the Yellow River for DIN, DIP, and DSi were fertilizer loss, sewage effluents, and runoff, respectively. Therefore, it is necessary to continue to promote policies to decline fertilizer application, such as “soil testing to determine appropriate fertilizer formulas” and “use of organic fertilizers instead of chemical fertilizers”. It is essential to improve sewage treatment technology and reduce the discharge of substandard wastewater. Finally, “green travel” should be utilized to reduce pollutant emission, which could not only improve air quality and reduce the deposition of nutrients, but could also increase life expectancy. Even though the ecological environment of the Yellow River basin is changing rapidly, it is highly possible for the Yellow River to achieve both increased water quality and reduced eutrophication.

CRediT authorship contribution statement

Nian Wu: Conceptualization, Sample determination, Methodology, Software, Writing - Original draft preparation, Manuscript Revision.

Su-Mei Liu: Conceptualization, Writing - Reviewing and Editing.

Gui-Ling Zhang: Investigation.

Hong-Mei Zhang: Sample determination.

Declaration of competing interest

The authors declare no conflict of interest.

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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.2020.142488.

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