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Research papers

Human-driven long-term disconnect of nutrient inputs to the Yellow River basin and river export to the Bohai Sea

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ABSTRACT

Combining long-term measurements of nitrogen (N) and phosphorus (P) concentrations in the Yellow River (YR) (1981–2018) and river discharge (1981–2018) to the Bohai Sea (BS) with model-based estimates of nutrient delivery shows a disconnect between nutrient delivery and export fluxes to the BS. Basin-wide nutrient delivery to surface water, primarily from agriculture and sewage, exhibited an increasing trend over the whole period. However the nutrient export fluxes to BS declined from 1981 to 2002, followed by a period of large inter-annual variability. Nutrient export is strongly controlled by water discharge, the Water and Sediment Regulation Scheme (WSRS), and the large volume of water containing nutrients that is extracted for primarily irrigation. Dissolved organic N (average 15% in the period 2001–2018) and dissolved organic P (68%) accounted for an important share of total dissolved N and P, which is due to agricultural inputs and preferential retention of inorganic forms in the YR basin. River nutrient export fluxes alone cannot explain the changes in the frequency and area of red tides in the BS due to impacts of the WSRS, the long water residence time in the BS and climate change. This calls for the application of mechanistic models for describing the impact of combined long-term changes in the BS ecosystem.

1. Introduction

River export of nutrients has a large impact on the ecology of coastal seas, especially in estuaries (Dai et al., 2011; Gao and Wang, 2007; Pan et al., 2013; Tong et al., 2015). Anthropogenic activities, through wastewater from households and industries and leaching and runoff from agricultural fields have caused a rapid increase in river export of the nutrients nitrogen (N) and phosphorus (P) to coastal waters in large parts of the world leading to severe eutrophication (Beusen et al., 2022) and changes in nutrient forms and proportions (Billen et al., 1991). One of the symptoms of eutrophication and nutrient distortions is the increase in the frequency and area of harmful algal blooms (HABs) (Beusen et al., 2022; Glibert, 2017; Gobler, 2020).

HABs are a major environmental problem in China, as they increasingly occur in the Bohai Sea (BS), Yellow Sea, East China Sea and South China Sea since the 1980 s (Wang et al., 2021a). The BS is a special case, as it is a semi-confined sea in which the Yellow River (YR), the second largest river in China, drains water with large loads of sediments and nutrients from extensive agricultural landscapes. The discharge of the YR is strongly regulated by dams and reservoirs, which control the seasonal patterns of the hydrology and nutrient and sediment transport (Wang et al., 2016; Wang et al., 2017a). Water consumption in the YR has a strong impact on the hydrology and nutrient cycling (Tong et al., 2016).

In order to prevent floods, provide irrigation water, and generate hydropower, more than 148 large and medium reservoirs with a total average volume of 31 km^3 were constructed in the Yellow River Basin (YRB) during the period 1997–2018 (Yellow River Conservancy

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Commission (YRCC)). Approximately 19 Gt of sediment was trapped by YR reservoirs until 2010 (Ran et al., 2013), which has strongly reduced the water storage capacity of many reservoirs. The YRCC has implemented the Water-Sediment Regulation Scheme (WSRS) at the beginning of every flood season since 2002 to improve the imbalance between water discharge and sediment loading, and remove accumulated sediments from reservoirs and the YR channel.

The WSRS changed the water discharge seasonal patterns, with an average of 28% of the annual water discharge and 41% of the annual nutrient flux into the BS occurring in less than one month (Liu et al., 2012). This dramatic alteration of the hydrological regime has profound physicochemical, ecological and geomorphological effects on the lower reaches of the river and the estuary (Li et al., 2017; Liu et al., 2012; Wu et al., 2019).

Regarding the impacts of the YR on the BS ecosystem, most studies focused on nutrient changes since the WSRS and did not consider changes in the proportions and forms of nutrients due to damming (Tong et al., 2015; Wu et al., 2019; Wu et al., 2021; Yu et al., 2010). However, distortion of nutrient proportions and a shift in the nutrient composition, particularly towards more organic N and P forms, may lead to the proliferation of specific HAB species (Ou et al., 2018; Wang et al, 2011; Zhang et al., 2020b).

To improve our understanding of the long-term changes in the BS ecosystem, we need to compare the situation before and after the WSRS and consider the changes in the N and P forms in the YRB due to damming. Therefore, the aims of this paper are to (1) quantify long-term (1970–2010) nutrient loading in the YRB and analyze the impact of changes of different sources (agriculture, wastewater, nature) and water extraction, (2) explore the changes of nutrients by river export in terms of total loading and inorganic and organic nutrient species to the BS under the influence of the WSRS, reservoirs and water usage, and (3) assess the impact of nutrient inputs, proportions and chemical species of the YR on the ecology of the BS, particularly the occurrences of red-tides.

2. Materials and methods

2.1. Data and methods

The nutrient, hydrological and supporting data used in this paper are listed in Table 1. Nitrate (NO_3-N), nitrite (NO_2-N), ammonia (NH_4-N) and phosphate (PO_4-P) were measured in samples collected monthly from 2001 to 2018 at the Lijin (LJ) station. LJ is the nearest hydrological station to the river mouth (about 100 km upstream), which can represent the riverine materials flowing into the BS (Fig. 1). After filtering water samples through a 0.45 μm cellulose acetate membrane, NO_3-N, NO_2-N, NH_4-N and PO_4-P were measured using respectively the cadmium-copper reduction method, the standard pink azo dye method, the indophenol blue method and phosphate-molybdenum method. The detection limits for NO_3-N, NO_2-N, NH_4-N and PO_4-P were 0.02, 0.02, 0.03 and 0.01 μmol/L, respectively. Total nitrogen (TN), total phosphorus (TP), total dissolved nitrogen (TDN) and total dissolved phosphorus (TDP) were measured by wet oxidation in acid persulfate with an analytical precision of 0.68 μmol/L for TN and TDN and 0.02 μmol/L for TP and TDP (Grasshoff et al., 1999). The dissolved inorganic nitrogen (DIN) concentration was calculated as the sum of NO_3-N, NO_2-N and NH_4-N. Dissolved organic nitrogen (DON) and dissolved organic phosphorus (DOP) concentrations were calculated as the difference between TDN and DIN, and between TDP and PO_4-P, respectively. Monthly DIN and PO_4-P data from 1981 to 1989 at Luokou Station were taken from the Institute of Environmental Health Monitoring (1990). Annual DIN data during 1991–1998 at the Lijin station were obtained from Lü and Zhang (2000) (PO_4-P concentrations are lacking). The monthly nutrient fluxes were calculated as the product of monthly nutrient concentrations and water discharge. The annual nutrient concentrations and fluxes were calculated by the mean monthly nutrient concentrations and the sum of monthly nutrient fluxes, respectively.

Table 1
Datasets used for nutrient concentrations, nutrient delivery to the river from various sources, discharge, suspended sediments, reservoirs, population and agriculture and occurrence of red tides of the Bohai Sea.

Parameter	Year, and source
Monthly NO_3-N, NO_2-N, NH_4-N, PO_4-P, DON, DOP, TDN, TDP, TN, TP at Lijin station	2001–2018, this paper
Monthly DON, DOP, TDN, TDP at Lijin station	2001–2002 (Tan, 2002); 2002–2004 (Wang, 2007)
Monthly DIN and PO_4-P at Luokou	1981–1989 (IEHM, 1990)
Annual DIN at Lijin	1991–1998 (Lü and Zhang, 2000)
Nutrient delivery to the river from various sources	1970–2010 (IMAGE-GNM modeled results)
Water discharge and suspended sediment discharge at Lijin	1981–2018 (YRCC)
Number and capacity of large and medium-sized reservoirs, water consumption and wastewater	1998–2018 (YRCC)
Annual population and chemical fertilizer use	1985–2018 (NBS)
Frequency and area of red tide in the Bohai Sea	2000–2018, Chinese Marine Disaster Bulletins and Xin et al. (2019)

2.2. IMAGE-GNM model

Based on land cover and climate data from the integrated assessment model IMAGE (Stehfest et al., 2014) and hydrological data from the model PCR-GLOBWB (Van Beek et al., 2011), the grid-based IMAGE-GNM (0.5 × 0.5°) (Beusen et al., 2015) was used to simulate river N and P loading from various sources for the period 1970–2010. A brief description of IMAGE-GNM is given below, details can be found elsewhere (Beusen et al., 2015, Beusen et al., 2022). Nutrient sources include diffuse sources in natural and agricultural land, point sources (sewage water), aquaculture, allochthonous organic material from vegetation in floodplains, and P from rock weathering. Diffuse nutrients are delivered to surface water by surface runoff, leaching from soils via shallow and deep groundwater, and riparian zones. For the diffuse sources, IMAGE-GNM uses the soil budget approach. N inputs for the soil budget are synthetic N fertilizer, animal manure, biological N fixation, and atmospheric deposition, and outputs are crop and grass withdrawal, and ammonia volatilization. Leaching and denitrification are calculated on the basis of soil properties and climate. Input terms in the soil P budget are fertilizer use and animal manure, and outputs include crop and grass withdrawal. Nutrients are delivered to inland surface water by different sources and transport pathways within a grid cell, and via streams and rivers flowing from upstream to downstream grid cells.

3. Results

3.1. Changes in water discharge and nutrient fluxes

The water discharge of the YR at Lijin showed strong inter-annual variability, with a distinct decreasing trend in the period 1981–2002, and a sharp increase to a higher average level of 19 km^3/yr after 2002 (Fig. 2a). Annual DIN and PO_4-P fluxes and water discharge exhibited similar inter-annual variability (Fig. 2b, c). Both DIN and PO_4-P fluxes showed a decline in the years 1981–2002, followed by a period of strong inter-annual variability lacking a clear trend. The patterns of annual TN and TP fluxes were similar to those of DIN and PO_4-P, respectively (Fig. S1). Annual DON and DOP fluxes during 2001–2018 vary strongly and lack a clear trend (Fig. 2b, c).

The molar DIN/PO_4-P ratios are much higher than the Redfield ratio (16) and showed small changes before 2002, and continuously increased between 2002 and 2007, followed by large year-to-year variability

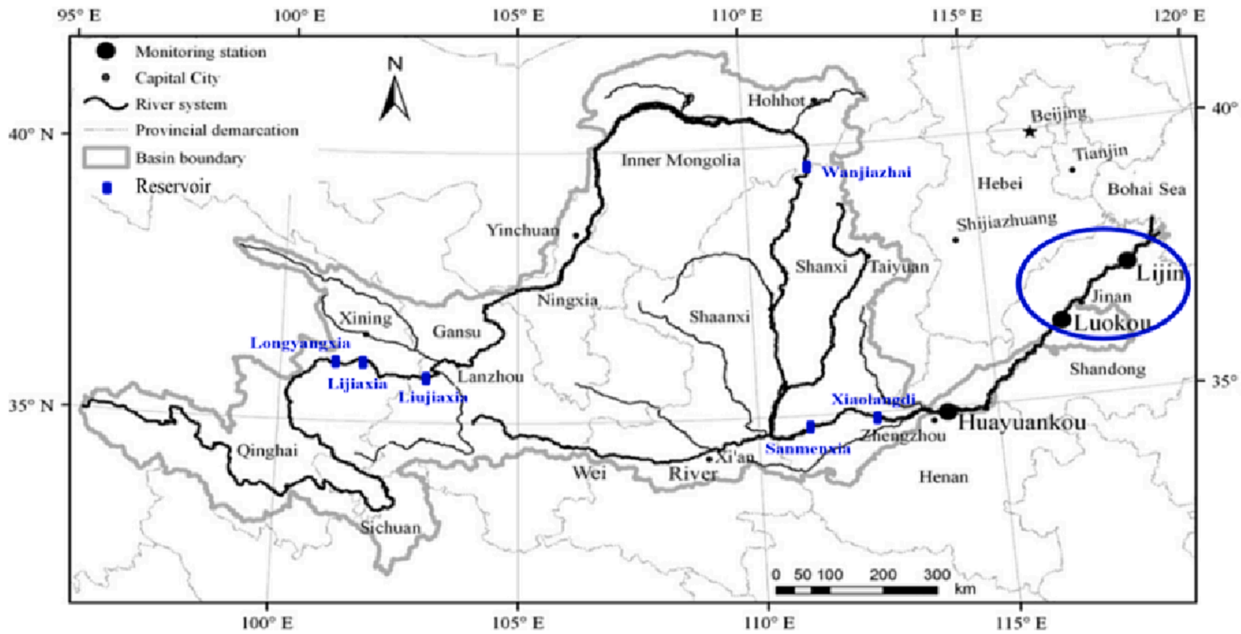

Fig. 1. Map of the Yellow River Basin, location of the six largest reservoirs and the sampling stations Luokou and Lijin (marked by the blue circle). The Liujiaxia, Longyangxia and Xiaolangdi reservoirs were built in 1968, 1986 and 1999, respectively. The WSRS was implemented in 2002–2015 and 2018. Due to insufficient water storage in the Xiaolangdi reservoir, the WSRS was interrupted in 2016–2017. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

(Fig. 2d). Molar TDN/TDP ratios are lower than DIN/PO_4-P ratios and exhibited large inter-annual variability with values ranging between 300 and 900 (Fig. 2d, e).

Monthly DIN ($R = 0.840$, $n = 326$, $p < 0.0001$) and PO_4-P ($R = 0.813$, $n = 290$, $p < 0.0001$) fluxes both show strong positive relationships with water discharge (Fig. S2). Monthly patterns of water discharge prior to 2002 showed a clear peak during the flood season from July to October (Fig. 3a). WSRS changed the seasonal variations of the water discharge, with lower peaks compared to the period before 2002, but earlier in the year (July, Fig. 3a). Similar to the monthly water discharge, monthly DIN and PO_4-P fluxes were consistently high from July to October before 2002, and this peak advanced to July after the implementation of WSRS (Fig. 3b, c).

3.2. *Organic nutrient composition*

Calculated average annual DON/TDN, DON/TN, DOP/TDP and DOP/TP ratios in the YR from 2001 to 2018 were 0.15, 0.13, 0.68 and 0.16, respectively, similar to other studies (Table 2). With the strong variability, in some years the share of the dissolved organic nutrients may be even higher (Fig. 2b, c).

3.3. *Temporal variation of nutrient sources*

The IMAGE-GNM soil TN budget in the YRB increased 5-fold from 642 Gg N/yr in 1970 to 3307 Gg N/yr in 2010 (Fig. 4a). The soil TP budget rose over 50 times from 11 Gg P/yr to 603 Gg P/yr during the period 1970–2010 (Fig. 4b). Parallel to the soil nutrient budgets, the N and P delivery to surface water of the YR simulated by IMAGE-GNM continuously increased from 303 to 1204 Gg N/yr and 56 to 167 Gg P/yr, especially after 1980.

Delivery of N through agricultural surface runoff and groundwater discharge and from sewage increased almost 3-fold (134 to 367 Gg N/yr), 9-fold (42 to 362 Gg N/yr) and 4-fold (102 to 387 Gg N/yr) between 1970 and 2010, respectively (Fig. 4c). P loading through surface runoff from agriculture and sewage increased more than 2-fold from 42 to 104 Gg P/yr and 5-fold from 10 to 52 Gg P/yr (Fig. 4d).

The N and P delivery from natural sources remained stable at about 20 Gg N/yr and 3 Gg P/yr, with a declining percentage of total delivery (Fig. 4c, d). P runoff from natural ecosystems in China declined from 0.35 Tg P/yr in 1600 to 0.25 Tg P/yr in 2012 (Liu et al., 2016). Nutrients from aquaculture increased from 1 to 63 Gg N/yr and 0.08 to 7 Gg P/yr with a contribution to total delivery growing from 0.4% to 5% for N and from 0.1% to 4% for P (Fig. 4c, d).

4. Discussion

4.1. *Agriculture and sewage sources dominate the nutrient sources*

The average proportion of agricultural sources and sewage in TN delivery was 63% (55–70%) and 32% (24–38%), which is consistent with independent estimates of 59% and 23%, respectively (Yu et al., 2019). Agriculture was the main source for both N and P delivery (Fig. 4c, d), similar to the situation in the Yangtze River (Liu et al., 2018). The changes in the agricultural crop sown area during the years 2000–2018 were not large, but fertilizer application in the YRB increased rapidly during 1985–2018 (Fig. S3a, b).

The share of TP delivery from agricultural surface runoff decreased from 75% to 63% with an average of 72% and that of sewage increased from 18% to 31%, which is comparable with agricultural contribution of 64% of the TP load in the YRB (Wang et al., 2021c). The total population and domestic wastewater in the YRB demonstrated an increasing trend (Fig. S3c, d). Both the urban population and proportion of urban residents in the YRB gradually increased (Fig. S3e, f). Increasing urban populations with connection to sewage systems resulted in large amounts of untreated human waste due to absence of any treatment (in the initial years) or the dominance of secondary treatment systems (in more recent years). The contribution of nutrients from aquaculture to total delivery increased from negligible amounts to 4–5% for N and P (Fig. 4c, d), which is similar to findings for the Yangtze River basin (Cui et al., 2021; Liu et al., 2018).

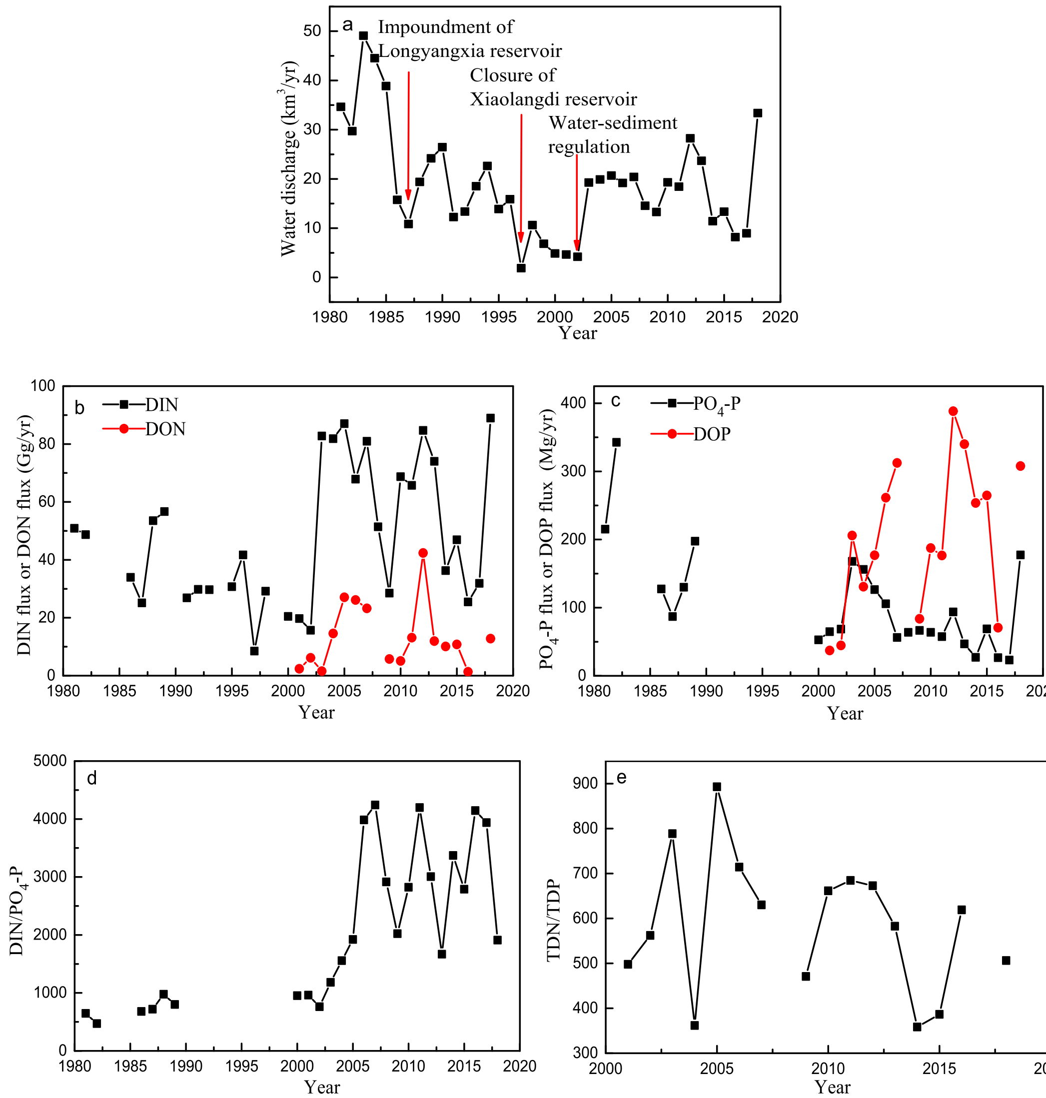

Fig. 2. (a) Annual water discharge at Lijin, (b) annual DIN and DON fluxes, and (c) PO_4-P and DOP fluxes to the BS (see Table 1 for stations and years); (d) Annual molar DIN/PO_4-P and (e) molar TDN/TDP ratios in the YR export to the BS.

4.2. Long-term disconnect of terrestrial inputs and river nutrient export

In contrast to the increasing delivery to surface waters in the YRB during the period 1970–2010, the N and P export fluxes do not show such a gradual increase, as they are strongly controlled by water discharge. These changes in export by the YR to the BS are the net result of changes in the soil nutrient budgets in agriculture (section 4.2.1), retention through temporary storage of nutrients in soils, groundwater and sediments (section 4.2.2), and water consumption (section 4.2.3).

4.2.1. Soil nutrient budgets

In most years, the soil TN and TP budget surpluses were much larger than the output fluxes from surface runoff and groundwater (Fig. 4). Budget P deficits prior to 1975 may be due to neglecting human manure, which was playing an important role in that period (FAO, 1977). The difference between soil budgets and delivery to surface water is due to soil and groundwater denitrification, and accumulation of N and P in soils and groundwater, as was also reported in other river basins like the Mississippi River (Van Meter et al., 2016). High inorganic nitrogen (mainly NO_3-N) accumulation in soil profiles was observed in many regions of China, especially in dryland soils of north China (Cui et al., 2008; Ju et al., 2004). Soil NO_3-N accumulated in winter wheat/summer maize rotation system in the North China Plain was above 172 kg/ha in 90 cm soil depth (Cui et al., 2008).

4.2.2. Temporary storage of nutrients

The surplus N and P applications in soils (Bouwman et al., 2017) accumulate in soils, and another part of the N and P is lost by surface runoff and erosion or leaching and subsequent groundwater transport, and may be trapped in river and reservoir sediments, groundwater, wetlands, riparian floodplains, and lakes (Sharpley et al., 2013). With the increasing water storage in large and medium-sized reservoirs in the YRB, the nutrient retention in the reservoirs is an important filter for nutrients (Fig. 5a). The water storage of large and medium-sized reservoirs in the YRB increased 3-fold from 14 km^3/yr in 1997 to 42 km^3/yr in 2018 (Fig. 5a). The long water residence times in the reservoirs are prone to nutrient retention through uptake, sedimentation, absorption and denitrification (Liu et al., 2020; Maavara et al., 2020; Xu et al., 2018). For example, the XLD Reservoir is generally thermally stratified during warmer summers with hypoxic bottom water, conditions known to favour denitrification (Cheng et al., 2019). The cascade dam in the upper YR decreased the flux of TN 38-fold and that of TP 6-fold (Ouyang et al., 2011). DIN retention flux was 0.9 Gg in the Xiaolangdi reservoir in June 2017 (Xu et al., 2018), which was approximately 3% of the annual DIN flux at Lijin. The retention effect of large reservoirs has led to significant reductions of nutrient concentrations at Lijin station since 2014

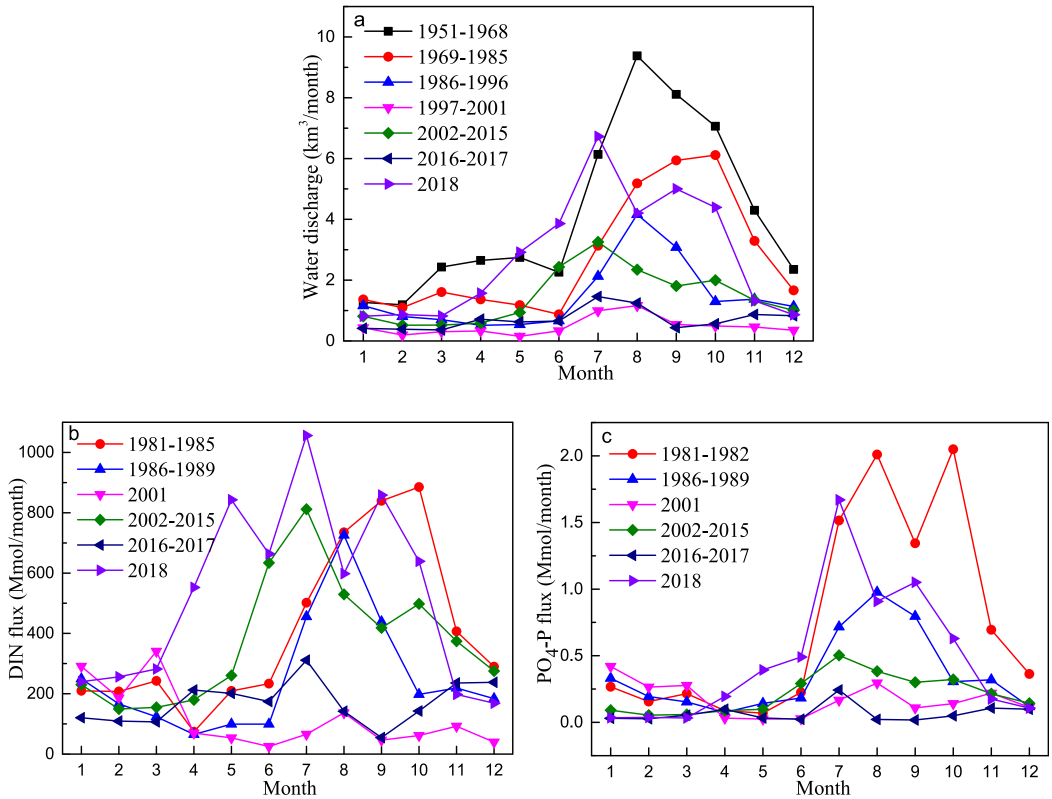

Fig. 3. (a) Monthly average water discharge, (b) DIN flux, and (c) PO_4-P flux for different periods (1981–1985, 1986–1989, 2001 prior to implementation of the WSRS; WSRS-years 2002–2015 and 2018, and no-WSRS-years 2016–2017).

Table 2
Molar nutrient ratios* in the YR and the Yangtze River.

DON/TDN	DON/TN	DOP/TDP	DOP/TP	Period covered (source)
	Yellow River			
(0.10)		(0.41)		2001–2011 (Liu, 2015)
(0.07)		(0.40)		2012–2014 (Wu et al., 2017)
0.04–0.32 (0.15)	0.01–0.31 (0.13)	0.31–0.96 (0.68)	0.04–0.31 (0.16)	2001–2018 (This study)
	Yangtze River			
0.05–0.36 (0.18)	0.05–0.29 (0.16)	0.16–0.70 (0.44)	0.06–0.14 (0.10)	1998–1999 (Duan et al., 2008)
(0.10)		(0.27)	(0.13)	2009–2010 (Ding et al., 2019)
0.06–0.34 (0.20)				2012–2013 (Shi et al., 2015)

*Values in parentheses are average values.

(Wu et al., 2021).

The N retention in reservoirs is partly lost by denitrification (Finlay et al., 2013) or burial in sediments (Vilmin et al., 2020). P retention in reservoirs is due to P burial in sediments. These (temporary) stores for N and P can be released to the water column when concentrations of N (Vilmin et al., 2020) and P decline (Reddy et al., 1999) and thus form a legacy of past nutrient management.

4.2.3. Water consumption

Another factor influencing river nutrient export is water consumption, which removes nutrients from the YR during transport to the BS. Average water consumption through surface water extraction from 1998 to 2018 was 30 km^3/yr, which exceeds the water discharge at the river mouth during the same period by a factor of 2 (Fig. 5a). Agricultural water consumption (excluding the return flow from field drainage) declined from 91% to 66% of total water consumption in the period 1998 to 2018 (Fig. 5b). The average annual total water consumption during 2000–2010 made up approximately 60% of net precipitation (precipitation minus evapotranspiration) in YRB. Based on the N and P delivery loads (Fig. 4) and river export to the BS from 2000 to 2010, and accounting for total water consumption (Fig. 5a), the calculated average annual biogeochemical retention in the YR was approximately 33% (9–56%) for N and 39% (19–64%) for P (Fig. S4). This would be higher if we account for water extraction. However, it is difficult to estimate the net effect because we do not know the composition of the drainage return flow. Hence, the N retention in the YR was similar to that in the Yangtze River (average 34% during 2001–2010) (Liu et al., 2018) but lower than Mississippi River (average 43% during 2001–2010); the P retention fraction in the YR was lower than that in the Yangtze River (47%) (Liu et al., 2018) and Mississippi River (66%). This may be attributed to the relatively small reservoir volume and the large volume of consumption in the YR compared to that in the Yangtze River (Table S1).

4.3. Nutrient composition

Agriculture, as the primary nutrient source in the river basin, may have a strong impact on the nutrient composition in the YR. Excessive fertilizer application resulted in the extremely high DIN in the YR (Fig. 4c and S3b), which was similar to previous studies (Wu et al., 2021; Yu et al., 2010). Although the average proportion of DON in DTN is around 15%, the average DON concentration is approximately 53 μmol/L, and the highest concentration can be up to more than 100 μmol/L.

DOP concentrations in the YR increased after 2009 (making up 0.95 of the TDP), and annual minimum DON concentrations also showed an upward trend (Wu et al., 2021). The DON/TDN and DOP/TDP ratios were approximately 0.13 and 0.41 in the Yellow River Estuary,

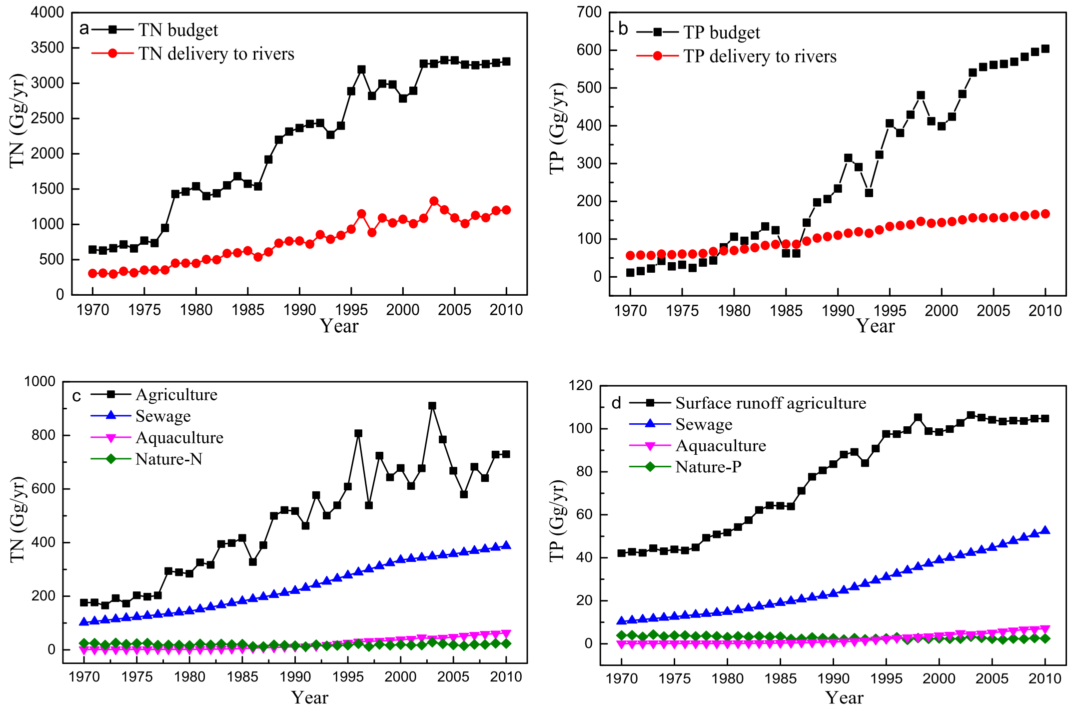

Fig. 4. Simulated (a) TN and (b) TP soil budget and delivery loads to surface water in the YRB from 1970 to 2010; (c) TN and (d) TP delivery to the YR from different sources during 1970–2010. The TN term "Agriculture" is the sum of surface runoff and groundwater in agricultural areas. The TN term "Nature-N" includes surface runoff and groundwater in natural areas, and N delivery from vegetation in floodplains and deposition onto surface water. "Nature-P" is the sum of surface runoff in natural areas, P delivery from vegetation in floodplains and rock weathering.

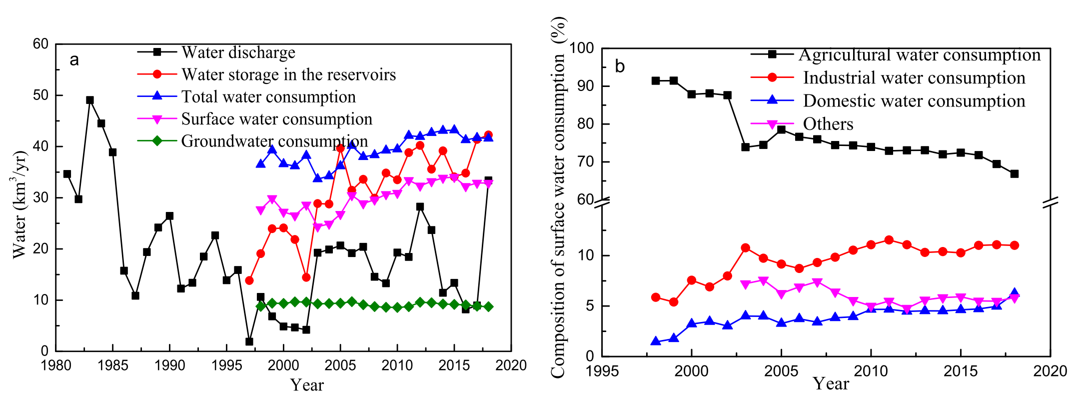

Fig. 5. (a) Variation of water discharge, water storage in large and medium reservoirs and water consumption, and (b) the percent composition of surface water consumption in the YRB. The total water consumption is the sum of surface water consumption and groundwater consumption.

respectively (Liu et al., 2009). The DON/TDN, DON/TN and DOP/TP ratios in the YR were comparable to those in the Yangtze River, while the DOP/TDP ratios in the YR exceed those in the Yangtze River (Table 2).

It is known that urea fertilizers contribute to river DON (Glibert et al., 2006). The use of urea in China was close to 60% of total N fertilizer use in the early 2000 s, and at present it is still around 35% (IFA, 2022). Urea made up 19% of DON at Datong in the Yangtze River during the period 2011–2014 (Zhang, 2015). Similarly, increasing use of P fertilizers and management of animal manure inevitably leads to increasing P losses through surface runoff (Bouwman et al., 2017). Human excreta in sewage water and animal manure also contribute to DON and DOP loading.

The YR has the highest suspended sediment concentration of any major river in the world (Pan et al., 2013). The high suspended particulate matter are accompanied by high DON, which can lead to rapid transformation, including ammonification and subsequent nitrification rates (Xia et al., 2013). Due to high suspended particulate matter concentrations in the YR, PO_4-P is easily absorbed into particulates, thus lowering the PO_4-P concentrations in the water column (Liu et al., 2012; Pan et al., 2013); therefore, DOP concentrations exceed the PO_4-P concentrations in most years (Fig. 2c).

In addition, the reservoirs change the nutrient composition. DON and DOP retention in some reservoirs are lower than those of DIN and PO_4-P. For example, DON/TDN, DOP/TDP and DOP/TP ratios in the outflow of the Three Gorges Reservoir at Yichang were higher than the same ratios in the inflow at Fuling (Ding et al., 2019). Finally, WSRS promoted the release of several dissolved free amino acids Asp, Glu and Gly from particulate amino acids and sediment amino acids to the dissolved phase by 4%-17% (Zhang et al., 2020a).

4.4. Impact of nutrients on the environment of the Bohai Sea

YR contributed 60% of the freshwater discharge, 90% of the sediment and the largest proportion of nutrient loads, including organic N and P in the freshwater flow to BS (Wang et al., 2009). With this important share in the supply of water, materials and nutrients, BS may thus be impacted by nutrient ratios, nutrient flushing in early summer during WSRS, and the high fluxes of organic nutrients in the YR discharge.

The YR discharge into BS is characterized by high DIN/PO_4-P and TDN/TDP ratios (Fig. 2). Nutrient export by the YR could therefore lead to P-limitation for phytoplankton growth in the YR estuary and BS (Zhang et al., 2004). Due to the combined effect of riverine inputs, atmospheric deposition, anthropogenic activities and the relatively long residence time of water in the BS (Liu et al., 2017), DIN concentrations and DIN/PO_4-P ratios have been increasing in the BS (Ning et al., 2010; Wang et al., 2018; Xin et al., 2019), along with high DON and DOP fluxes.

The frequency and area covered by red tides the major HAB in BS, has rapidly increased since 1995 (Fig. S5). Approximately 70% of the annual occurrences of red tides occurred in the months June, July and August (Song et al., 2016). WSRS promoted the diffusion of nutrients to the central BS (Wang et al., 2017b), which may be related to the gradual extension of HABs from the coastal waters to the central part of the BS after 2000 (Song et al., 2016).

The dominant HAB species in observed red tides was *Noctiluca scintillans* before the year 2000, while since then the species became more diverse (major species are *Noctiluca scintillans*, *Skeletonema costatum*, *Karenia mikimotoi*, *Mesodinium rubrum* and *Phaeocystis globosa*) (Song et al., 2016; Wang et al., 2018; Xin et al., 2019). Although our data show no clear correlations between DIN, DON, PO_4-P and DOP fluxes and DIN/PO_4-P ratio with the frequency and area of red tides in the BS during different periods (Figs. S5-S7), the high DON and DOP fluxes may impact the proliferation of certain HAB species. Various studies showed that the red-tide species *Skeletonema costatum* can utilize various forms of DON and DOP to maintain high abundance in DIN and PO_4-P deficient systems (Zhang et al., 2020b). Flagellates (*Alexandrium tamarense*, *Chattonela marina* and *H. akashiwo*) in the BS, grew well in the presence of DOP (Wang et al., 2011). DON is known to drive the shift from diatom to dinoflagellate in the Bohai Sea (Chen et al., 2022).

The major brown tide species *Aureococcus anophagefferens* began to appear after the year 2009 (Wang et al., 2021b). Ou et al. (2018) demonstrated that *Aureococcus anophagefferens*, grows faster in the presence of urea and glutamic acid than with inorganic substrates. Brown tides are normally limited to a small area in the northwestern part of the BS, with a strong impact on mariculture production, primarily shellfish.

5. Conclusions

Our results show that the human interference in the YR hydrology is so strong that the nutrient sources on the one hand and the export and impacts in BS became disconnected. The human interferences (agricultural intensification, wastewater discharge, dam construction, water extraction, and the WSRS) cause changing seasonality of water and nutrient fluxes, changing stoichiometry and high levels of organic nutrient forms, which are known to strongly impact HAB proliferation. However, the BS biogeochemical system is complex, amongst others due to the long residence time of water in the BS, and possibly climate change, that there is no direct relationship between nutrient sources and fluxes into BS, and their impact on HAB proliferation. The impacts of nutrient loading may be delayed or HABs may appear after successions of algal blooms. Apart from changing seasonal patterns, the nutrient composition changed. The high N:P ratios are due to the high DIN concentrations and extremely low PO_4-P concentrations which are easily absorbed to the high suspended sediment. The high organic N and P export fluxes to BS are the result of agricultural organic nutrient sources, preferential retention of inorganic forms, suspended sediments in the water column and reservoir retention processes.

Lacking clear direct relationships between nutrient fluxes, nutrient ratios and forms in the YR export to BS on the one hand and the frequency and area of red tides in the BS on the other hand show that there is a need for employing mechanistic models for analyzing the combined long-term impact of multiple and simultaneous changes on biogeochemistry in the BS.

CRediT authorship contribution statement

Fuxia Yang: Investigation, Conceptualization, Formal analysis, Writing – original draft, Visualization. **Zhigang Yu:** Formal analysis, Writing – review & editing. **Alexander F. Bouwman:** Conceptualization, Formal analysis, Writing – review & editing. **Hongtao Chen:** Methodology, Data curation, Resources. **Huimin Jian:** Methodology, Data curation, Resources. **Arthur Beusen:** Methodology, Resources, Writing – review & editing. **Xiaochen Liu:** Methodology, Resources, Writing – review & editing. **Qingzhen Yao:** Conceptualization, Funding acquisition, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jhydrol.2023.129279.

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