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Exploring wastewater nitrogen and phosphorus flows in urban and rural areas in China for the period 1970 to 2015

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HIGHLIGHTS

- The spatiotemporal changes of N and P point source were simulated covering 4436 WWTPs in the whole China for 1970-2015.
- The urban nutrient discharge to surface water increased 22 fold for N and 29 for P.
- The Eastern 10 strongly urbanized provinces contributed 43% of nutrients in China.
- Eliminating the production and sale of P-containing detergents is urgently needed.
- The N:P molar ratio in wastewater discharge decreased from 20 to 15 for 1970-2015.

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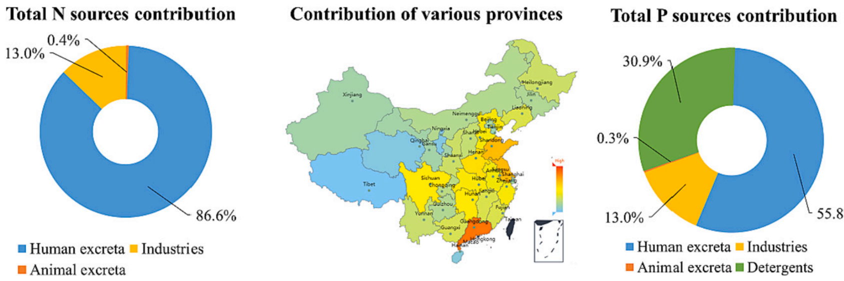
GRAPHICAL ABSTRACT



ABSTRACT

China has experienced rapid population growth and increasing human N and P discharge from point sources. This paper presents a new spatial and temporal model-based, province-scale inventory of N and P in wastewater using detailed information on the location and functioning of 4436 WWTPs covering China for the period 1970–2015. China's nutrient discharge to surface water increased 22-fold from 177 to 3908 Gg N yr^{-1} and 29-fold from 20 to 577 Gg P yr^{-1} in urban areas between 1970 and 2015. The ten strongly urbanized and industrialized provinces along the Eastern coast contributed 43 % of China's total N and P discharge to surface water in 2015. At present, the contribution of rural areas to total wastewater discharge (2082 Gg N yr^{-1} and 434 Gg P yr^{-1}) is 35 % for N and 43 % for P. The model approach and sensitivity analysis of this study indicate that policies aiming at improving water quality need to consider these regional differences, i.e., improvement of the wastewater treatment technology level in Eastern regions and increasing both the sewage connection and wastewater treatment in Central and Western regions.

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1. Introduction

Human-induced intensification of the global cycles of nitrogen (N) and phosphorus (P) has detrimental impacts on aquatic ecosystems through eutrophication; ultimate symptoms of eutrophication include increased frequency and areal expansion of hypoxia and harmful algal blooms (Billen et al., 2001; Cloern, 1996; Dodds, 2002; Gruber and Galloway, 2008). Nutrients are discharged to surface water from diffuse and point sources. Diffuse sources include nutrient flows in surface runoff, erosion, groundwater flows from agricultural fields, natural ecosystems and atmospheric deposition, and direct flows from aquaculture and vegetation in riparian and flooded areas. Point sources of nutrients are urban sewage systems that discharge at a limited number of locations in urban centers.

The impact of nutrient loading depends not only on the amounts of nutrients and the composition in terms of nutrient forms and their proportions but also on discharge location (Kemp et al., 2009; Seitzinger et al., 2010). Therefore, it is essential to know both the diffuse (agricultural and natural) and point (wastewater) sources of nutrients when studying water quality and the impacts of nutrient distortions caused by human activities.

This study focuses on nutrients from point sources. Although urban areas cover only a small fraction (0.2–2.4 %) of the global land area (Potere and Schneider, 2007), N and P discharge to freshwater and coastal areas from point sources are substantial and dominant in surface water in many densely populated parts of the world (Bouwman et al., 2005; Harrison et al., 2005; Liu et al., 2018; Van Drecht et al., 2009) because currently >55 % of the global human population lives in cities (United Nations, 2018). P and N from wastewater became the first and second dominant sources respectively since the 1980s in the downstream of the Yangtze river basin (Liu et al., 2018) (Fig. SI4). The N and P discharged from point sources stem from human households (excreta and P-based detergents) with or without a sewage connection, from industries that discharge wastewater containing nutrients, and from domestic animals (Billen et al., 2012; Li et al., 2012; Morée et al., 2013; Nyenje et al., 2010; Quynh et al., 2005; Van Puijenbroek et al., 2019). Construction of wastewater treatment plants (WWTPs) often lags the sewage construction by several decades in many countries (Van Drecht et al., 2009). For example, in Europe and North America, the N and P loads showed a major increase in the 1960s and 1970s due to the massive connection of households to sewage systems and declined in the late 20th century due to the construction of WWTPs and the use of P-free detergents (Morée et al., 2013).

However, the nutrient discharge from the 45 % of the global population living in rural areas, where next to sewage systems also other forms of sanitation such as septic tanks and pit latrines are found, is poorly known and often ignored. Several studies have explored the N and P fluxes from urban areas both at local, national and global scales based on models and data-based approaches (Chen et al., 2019; Morée et al., 2013; Tong et al., 2017; Van Drecht et al., 2009; Van Puijenbroek et al., 2019; Zhang et al., 2020).

China has experienced a rapidly growing population to a current 1.37*10e^{4} billion and rapid urbanization to 56.1 % until 2015 (Gu et al., 2017), and as a consequence rapidly increasing nutrient discharge from urban point sources (FAO, 2019). The first large scale municipal WWTP in China was constructed in 1984 (Qiu et al., 2010). Since then until 2015, 4436 WWTPs have been constructed along the industrialization and fast urbanization. Chen et al. (2019) developed a model to explore the point sources of N and P on both biophysical and administrative scales for the year 2012. Zhang et al. (2020) studied influent and effluent changes of N and P in municipal wastewater in 52 China's coastal cities during from 2006 to 2015. There is no long-term spatially explicit investigation to simulate the N and P from point source to rivers for China as a whole. The available estimates either cover one single year (Chen et al., 2019), consider urban wastewater discharge only, or ignore changes in the number of inhabitants with a sewage connection (Chen et al., 2019; Qi et al., 2020). In addition, the number of inhabitants served by WWTPs and the removal efficiency of these WWTPs are poorly quantified and the fate of nutrients from households in rural areas is fraught with large uncertainty. Therefore, the long-term nutrient fluxes from China's point sources and performances of all the 4436 WWTPs in all the provinces need to be systematically studied. The model approach that we developed is a coupling of sanitation types, sewer connection rate, detergents use, and WWTPs technologies, and is used to explore ways to reduce the N and P point sources in Chinese inland and coastal waters.

The aim of our study is to assess the actual functioning of wastewater treatment throughout China and improve our knowledge on point source flows. This paper presents a new spatial and temporal model-based, province-scale inventory of N and P in wastewater using detailed information of the location and functioning of 4436 WWTPs covering China for the period 1970–2015. This period was selected on the basis of data availability which allowed for covering this period of >4 decades starting in 1970. Together with long-term estimates of nutrients from aquaculture (Wang et al., 2020) and agriculture (Bouwman et al., 2017), the results of the present work which covers 31 provinces contribute to the quantification of the nutrient loading to Chinese inland waters. This is an essential basis for analyzing the nutrient retention and accumulation in landscapes and waterscapes and nutrient export to coastal waters.

2. Methods and data

This study is based on the spatially explicit wastewater nutrient model (Morée et al., 2013; Van Drecht et al., 2009; Van Puijenbroek et al., 2019), which is part of the Integrated Model to Assess the Global Environment-Dynamic Global Nutrient Model (IMAGE-DGNM) (Liu et al., 2020; Vilmin et al., 2020). Here, we present a brief description and updates of the input data and model.

In contrast to earlier applications for either urban areas only or based one single year, we simulate the long-term wastewater nutrient flows at the scale of 31 Chinese provincial-level administrative regions (Fig. 1) for a series of years and include both urban and rural populations. The model calculates N and P emission fluxes in the human system, i.e., emissions from human excretion, urban industry, urban animal excretion and P emission from the use of detergents (in dishwasher and laundry), and the fate of the nutrients (e.g., use in agriculture, direct discharge, wastewater treatment, effluent from treatment plants, discharge to surface water, direct defecation in surface water, and emission to the atmosphere) (Fig. 2 and Table 1).

Rural and urban populations per province (*Dataset S1.xlsx,* in the Supplementary Information, SI) were obtained for the period 1970–2015 from the National Bureau of Statistics of China (National Bureau of Statistics of China, 2015). Human N excretion was assumed to be equal to protein-N consumption minus N in retail and household losses and N stored in human bodies (hair, nails, skin, etc.) based on various sources (Neset et al., 2006; Oddoye and Margen, 1979; Takahashi, 1985). The model uses equal protein intake rates for rural and urban populations because recent data show no significant differences in China (Ju et al., 2018). Data on protein consumption were obtained from FAO (FAOSTAT, 2017; Neset et al., 2006; Takahashi, 1985). P excretion was assumed to be a fixed fraction of N ($P = 0.1$ N).

Provincial data for sewage connection rate in urban areas were obtained from the Ministry of Housing and Urban-Rural Development of the People's Republic of China (Ministry of Housing and Urban-Rural Development of the People's Republic of China, 2016), assuming that households connected to a system of drinking water pipes also have a connection to a sewer (Graham and Marvin, 2002). Sewage connection in rural areas exists (Table SI1) but is not accounted for specifically (regarding nutrient removal) in the calculations.

Detailed data on 4436 WWTPs (*Dataset S1.xlsx in SI2*) were obtained from the Ministry of Ecology and Environment of the People's Republic of China (Ministry of Ecology and Environment of the People's Republic of China, 2014). The dataset includes the location, year of construction, treatment capacity, average load and technology for each WWTP in each province. These data were used to calculate the treatment capacity and actual operation loading for each WWTP and the aggregated results at the provincial scale for every year during the period 1970–2015.

The 42 treatment technologies distinguished (Fig. SI1) were grouped into four classes: primary, secondary, tertiary and quaternary treatments (Fig. 2b). Primary treatment includes the physical separation of large objects, plastic, wood, sand, etc. Secondary treatment is primary treatment plus aeration with active sludge (various organisms from archaea and bacteria to insects). Tertiary treatment is secondary treatment with additional (i) N removal by nitrification (aerobic) and subsequent denitrification (anaerobic), (ii) biological P removal by P accumulating bacteria with alternating starvation (low oxygen and nitrate) and banquet conditions (plenty of oxygen and nutrients) and (iii) chemical P precipitation with Fe and Al coagulants; in recent years also with Mg oxide or hydroxide to produce struvite. Quaternary treatment is tertiary treatment with additional steps such as sand filtration, ultra-filtration, ozone treatment, UV treatment, nano-filtration (NF), reverse osmosis (RO), and disinfection.

These treatment classes have typical removal rates, i.e., for primary, 10 % for both N and P; for secondary, 35 % for N and 45 % for P; for tertiary, 80 % for N and 90 % for P; and for quaternary, 95 % for both N and P. We recognize that these efficiencies are uncertain as they vary between installations depending on differences in design, operational and management conditions, and skills of the operators (Barat et al., 2013; Hu et al., 2012; Oleszkiewicz and Barnard, 2006b). Particularly efficient N removal requires careful and tuned management of the various treatment steps.

The model is extended to calculate the contribution from rural human emissions discharged via sanitation systems (Fig. 2, Table 1 and Table SI1). The most common systems in China include the septic tank, followed by the double-container composting system and biogas system (Ministry of Housing and Urban-Rural Development of the People's Republic of China, 2016; Standardization Administration of China, 2009). The national rural sanitation access rate for the period 1996–2014 (Fig. SI2) is used for all provinces due to a lack of provincial data. For the year 1970, the sanitation access rate is assumed to be equal to that in urban areas. For the period 1971–1996, the sanitation access rate is interpolated by the model. For rural areas, we calculated the N and P fluxes with access to sanitation systems (F_8), and the rest (F_9, F_{10}) is added to the sink "other" (Fig. 2a). The outflow from the sanitation systems is separated into fluxes to surface water (F_{18}) and agriculture (F_{17}). The difference between sanitation systems in rural areas is not accounted for specifically in the calculations. Although there are large differences in the N and P removal rates from sanitation systems in rural areas (through biological processes, soil seepage, and volatilization) depending on the prevailing sanitation system (Li et al., 2009; Wang et al., 2008), here a nutrient removal rate of 15 % is used for rural inhabitants with access to sanitation systems (76 % of rural population in 2014, Fig. SI2) for both N and P. This also includes sewage systems in rural areas, where the removal rate of 15 % is in between those for primary and secondary wastewater treatment discussed above.

For urban areas, N and P waste from inhabitants lacking a sewage connection together with retail household waste and sewerage leakage ends partially in the "other" pool (F_4, F_5) and the rest in surface water (F_3) (Fig. 2a). The wastewater from sewerage systems can be discharged to surface water directly (F_3) or treated in the WWTPs (F_2) (where the removal fraction depends on the type of wastewater treatment technologies shown in Fig. 3, Fig. SI3). The model specifically accounts for urban draught animals, their excreta, and the fate of these excreta. Animals in livestock production systems are not part of the wastewater model, but part of the calculations of diffuse sources of nutrients (Beusen et al., 2015).

The calculation method of detergent use (F_{11}, F_{13}) (Fig. 2a) is a function of GDP as described in Van Puijenbroek et al. (2019). For P content in laundry and dishwasher detergents, we used 6.25 % and 11.7

Fig. 1. Chinese provincial-level administrative regions and geographical breakdown of the four economic regions in China. (Note: the relevant estimates of Hongkong, Taiwan and Macao are not included due to the lack of data).

%, respectively (see SI4). For all the provinces in China, P detergents were linearly interpolated for the period 1970–2015. The use of dishwashers and laundry detergents in rural areas was assumed to be 50 % of that in urban areas. P concentrations of detergents are described in SI4.

The sensitivity of model results to variation of 46 model variables and parameters was calculated using Latin hypercube sampling (LHS) (Saltelli et al., 2000), using default ranges for each variable or parameter (Morée et al., 2013). We performed 1000 runs for the year 2000. Expressing this sensitivity as a standard regression coefficient (SRC) allows for ranking model variables and parameters based on their effects

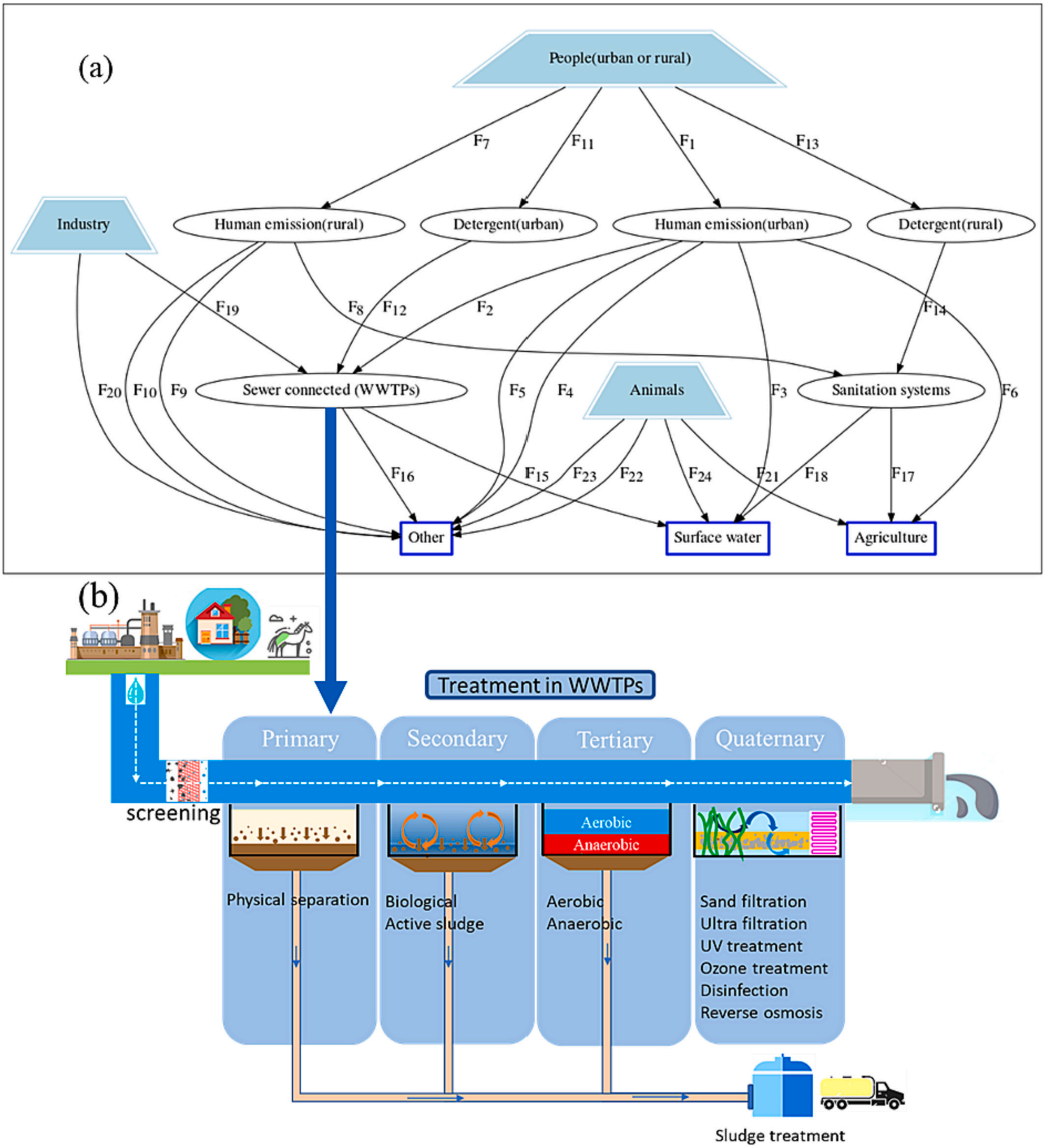


Fig. 2. (a) Scheme of the wastewater nutrient model in the IMAGE-DGNM framework. Arrows indicate the fluxes *F* of N and P (see Table 1 for details) from human excreta, animal excreta, P-based detergents and industries to the agriculture system (waste recycling in agriculture), surface water (i.e., discharge), and other compartments (including non-agricultural soils, atmosphere, and groundwater). Light blue trapezoids represent the original sources of N and P. Blue rectangles represent the sinks of N and P. (b) Scheme of wastewater treatment stages in the four sewage treatment classes. Every wastewater treatment class adds one or more treatment stages to those of the previous class(es).

Table 1
List of main nutrient flux formulas in the model.

Flux [a]	Description
F_1 and F_7	Human excreta N from people in urban and rural areas, respectively. The approach is based on protein consumption per person from Food and Agriculture Organization (FAO) (FAOSTAT, 2017). $E_{human\_N} = 0.365^{*}\ I_{human\_N}$, where $I_{human\_N} = 4 + 14\left(\frac{GDP_{PPP}}{33000}\right)^{0.3}$, where I_{human_N} is the protein N intake in g per person per day, we used an N content of 0.16 in protein (Block and Bolling, 1946), and the GDP_{PPP} is the purchasing power parity in U.S. dollars per person per year (World Bank, 2015). Human P emission is $E_{human\_P} = R_{N\_P}{}^{*}\ I_{human\_N}$, where R_{N_P} is the ratio between human P and N emissions, and here we use 1/10 which is derived from the 20th-century N and P consumption data set (USDA, 2012).
F_2	Human excreta to WWTPs through the sewer connection in urban areas. $D_{sw}^{N\_P} = E_{human\_N\_P}{}^{*}F_{sw\_con}$, where D_{sw}^{N} is the N and P discharges to WWTPs, $E_{human\_N\_P}$ is the human N and P emissions, and F_{sw_con} is the fraction of the urban population that is connected to the public sewage system.
F_3	Human excreta to surface water from the urban population lacking sewer connection.
F_4 and F_9	Retail and household waste.
F_5 and F_{10}	Loss of skin and hair.
F_6	Human excreta collection and use as fertilizer in agriculture.
F_8	Human excreta to sanitation systems in rural areas.
F_{11} and F_{13}	P emission from P-detergent use. $E_{det\_P} = E_{det}\ {}^{*}\ F_{det\_P}\ {}^{*}(1 - F_{det\_P\_free})$, where E_{det} is the use of laundry/dishwasher detergents (kg per person per year), F_{det_P} is the P content of laundry/dishwasher detergents, and $F_{det\_P\_free}$ is the fraction of P-free detergents. The P content in P-based laundry detergents F_{det_P} was calculated from 25 % sodium tripolyphosphate content in laundry detergents and 25 % P in sodium tripolyphosphate (Glennie et al., 2002),i.e., the P content fraction F_{det_P} in dishwasher detergents is assume to be 0.117 based on 30 % of phosphate (PO_4^{3-}) in detergents and 39 % of P in PO_4^{3-} (Floyd et al., 2006) The P emission from laundry detergents is calculated according to the equation below: $E_{L\_det} = 10 - 10\left(\frac{GDP_{mer}}{20000} - 1\right)^{2}$ The formula is based on a data set of 25 EU countries (Floyd et al., 2006; Van Drecht et al., 2009), where GDP_{mer} is the Chinese provincial gross domestic product per person (expressed in U.S. dollars per person per year) (National Bureau of Statistics of China, 2015). $F_{det_{P_{free}}} = \frac{GDP_{mer}}{33000}$ We do not have information for specific provinces and legislation, so we use a function of GDP_{mer}, for provinces with a per capita GDP exceeding 33,000 U.S. dollars, $F_{det\_P\_free} = 1$. The P emission from dishwasher detergents is calculated as $E_{D\_det} = 0.365^{*}F_{use}{}^{*}\frac{Dose}{A_{hou}}{}^{*}F_{pop\_con}$, where F_{use} is the frequency of the use of automatic dishwasher, *Dose* is the mass of the tablets used in automatic dishwashers (in g), F_{pop_con} is the fraction of the population with access to an automatic dishwasher (the fraction is based on the population with a sewage connection), and A_{hou} is the average number of persons per household. $F_{pop\_con} = 0.25 + 0.07\ {}^{*}\ \frac{GDP_{mer}}{20000}$, where GDP_{mer} is the Chinese provincial gross domestic product per person (expressed in U.S. dollars per person per year) (National Bureau of Statistics of China, 2015).
F_{12}	Detergent-P to WWTPs through sewer connection in urban areas. Due to the absence of data on the urban population with access to laundry washing machines. We assume the connection fraction to be the same as Fsw_con, which represents the fraction of the urban population connected to the public sewage system. For further details, please refer to F_2.
F_{14}	Detergent-P to sanitation systems in rural areas.
F_{15}	Effluent from WWTPs to surface water. $E_{sw} = D_{sw}^{N\_P}*(1 - R_{WWTPs}^{N_P}))$, where $D_{sw}^{N\_P}$ is the N and P discharges to WWTPs, $R_{WWTPs}^{N\_P}$ is the overall removal of N and P through different types of WWTPs.
F_{16}	Sludge from WWTPs to others.
F_{17}	Rural human excreta collection and use as fertilizer in agriculture.
F_{18}	Effluent from rural sanitation systems to surface water.
F_{19}	Industrial wastewater to WWTPs through sewer systems in urban areas. Due to scarcity of data, the industrial nutrient flows were assumed to be 0.5 times the urban domestic emissions in 1970 and 0.15 times the domestic emissions in 2015 using linear interpolation for the period in between.
F_{20}	Industrial waste to 'other'.
F_{21}	Animal excreta collection and use as fertilizer in agriculture. Animal numbers are from (National Bureau of Statistics of China, 2015). The model only includes equidae used for animal traction, as their excreta are typically deposited in streets and city stables. This poses a potential risk of runoff into surface water bodies or the collection of excreta for agricultural use.
F_{22} and F_{23}	Volatilization and seepage of animal excreta.
F_{24}	Animal excreta to surface water.

[a] The fluxes numbers correspond to those routes in Fig. 2. More model details can be found in Van Drecht et al. (2009) and Morée et al. (2013).

on the N and P discharge to surface waters for the year 2000 (Table 2).

3. Results and discussion

3.1. Emissions from households and industries in urban areas

The total N emission (prior to treatment) in Chinese urban areas increased from 0.5 to 5.4 Tg N yr^{-1}, and that of P from 0.05 to 0.8 Tg P yr^{-1} (Fig. 4) between 1970 and 2015. The increase in total N emission was largely due to the increase in N from human emissions caused by growing population and increasing protein consumption (improving nutrition) with a contribution that increased from 70 to 87 % (Fig. 4b, c). The increase in total P emission was also mainly due to increases of P from human emissions (with a decreasing contribution from 68 to 56 %) and detergent use (with an increasing contribution from 1 to 31 %) (Fig. 4e, f). Between 1970 and 2015, N emissions from industries (5 fold) and human waste (13 fold) increased less rapidly than the corresponding P emissions (7 fold from industries, 13 fold from human excretion, and 473 fold from detergents). The contribution of animal waste in urban areas declined from 2 % to 0.4 % for N and from 1 % to 0.3 % for P during the same period.

3.2. Provincial and regional discharge to surface water from urban areas

N and P discharge to surface waters increased throughout the period 1970–2015 in all the provinces and economic regions (Figs. 5, 6a, and c). Urban N and P fluxes to surface water varied widely among the 31 provinces (Fig. 5) and 4 economic regions (Figs. 5, 6a, and c and Movie "P load to rivers.mp4" in SI). N and P discharge to surface water increased by a factor of 21 and 27 (depending on the province) between 1970 and 2015, respectively, primarily since the year 2000. The discharge in coastal provinces in Eastern China (Figs. 5, 6a, and c) was the largest of all regions for both N and P (Fig. 6a, c) and the provinces of Guangdong, Shandong, and Jiangsu were the three dominant provinces in Eastern China in 2015 (Fig. 5b and d). In Central and Western China, Hunan and Sichuan accounted for the largest increases in both urban N and P discharge (Fig. 5b and d).

3.3. Provincial and regional discharge to surface water from rural areas

On the national scale, China's N discharge to surface water in rural areas increased from 16 Gg to 2082 Gg from 1970 to 2015, mainly stemming from human waste. For P, the discharge to surface water

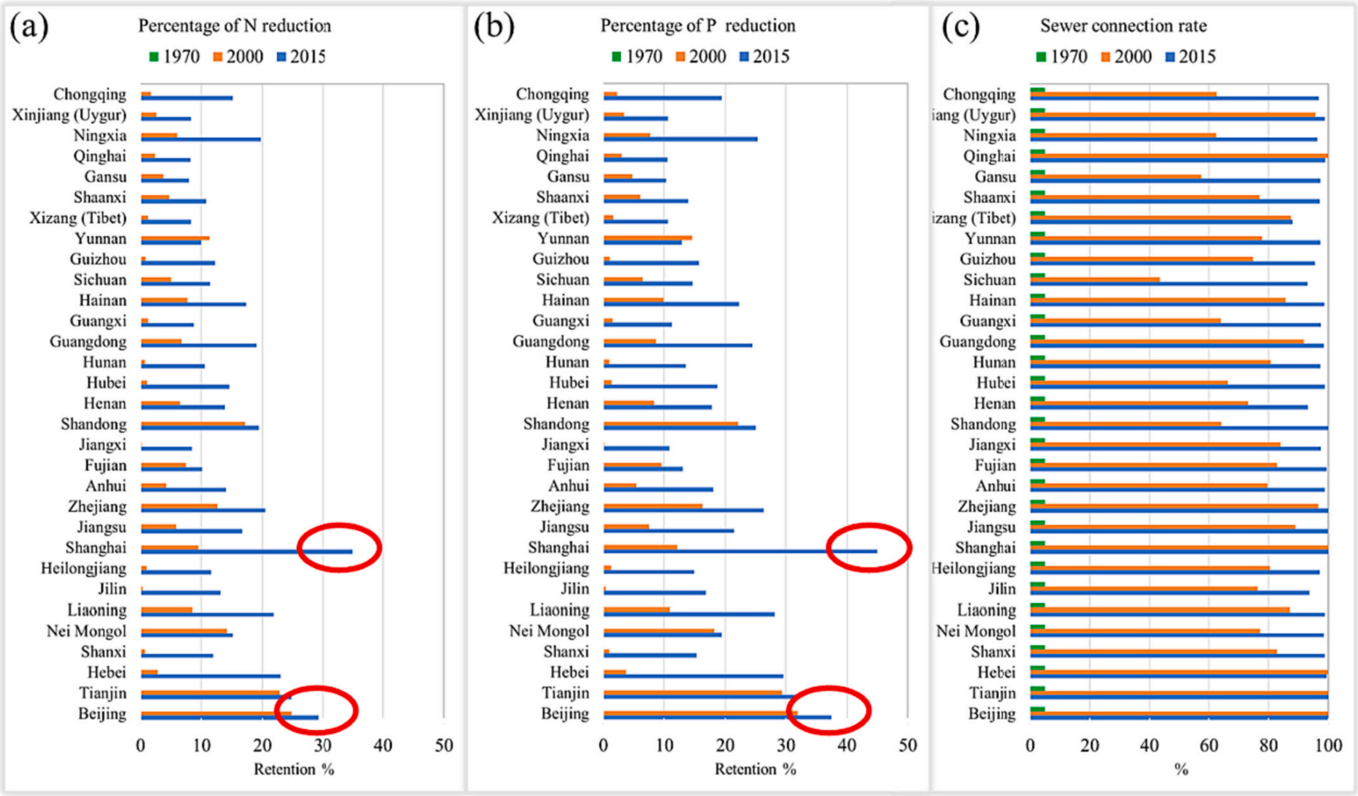


Fig. 3. Removal percentages of (a) N and (b) P for all the treatment in WWTPs and (c) sewage connection for the 31 Chinese provinces for the years 1970, 2000, and 2015. Red circles mark the removal efficiencies in Beijing and Shanghai, China's most developed provincial-level administrative regions.

Table 2

Standardized regression coefficient (SRC) representing the relative sensitivity of the model performance for 2 output variables (N and P delivery to surface water) to the variations of 46 model input variables and parameters for the year 2000.

Variables and parameters	N to surface water	P to surface water
Primary treatment connection		0.05
Secondary treatment connection	-0.12	-0.17
Tertiary treatment connection	-0.32	-0.38
Secondary treatment P removal fraction		-0.06
Tertiary treatment P removal fraction		-0.09
Secondary treatment N removal fraction	-0.05	
Tertiary treatment N removal fraction	-0.07	
Sanitation systems N/P removal fraction	-0.05	-0.06
Protein consumption	0.37	0.28
Urban population	0.20	0.23
Rural population	0.21	0.22
Rural sanitation connection	0.36	0.43
Sewer connection	0.52	0.56
Non-sewered human wastes to agriculture in urban	-0.07	-0.05
Household wasting/losses factor	-0.05	-0.04
Retail wasting/losses factor	-0.02	
Detergent use		0.22
P-free detergent use		-0.26
Industry factor of household N/P flows	0.04	0.04
Donkeys and mules N excretion	0.03	0.03
Sewer loss fraction	-0.04	-0.04
Animal feces N volatilization	**-0.04**	
Industry N loss	-0.04	
P:N ratio		0.26
N as fraction of protein	0.38	0.25

Values without color indicate $-0.2 < SRC < 0.2$; values with orange and green colors indicate values < -0.2 and > 0.2 respectively. Negative values indicate that a higher input variable or parameter value generates a lower model output variable, and positive values indicate that a higher input variable or parameter value generates a higher model output variable.

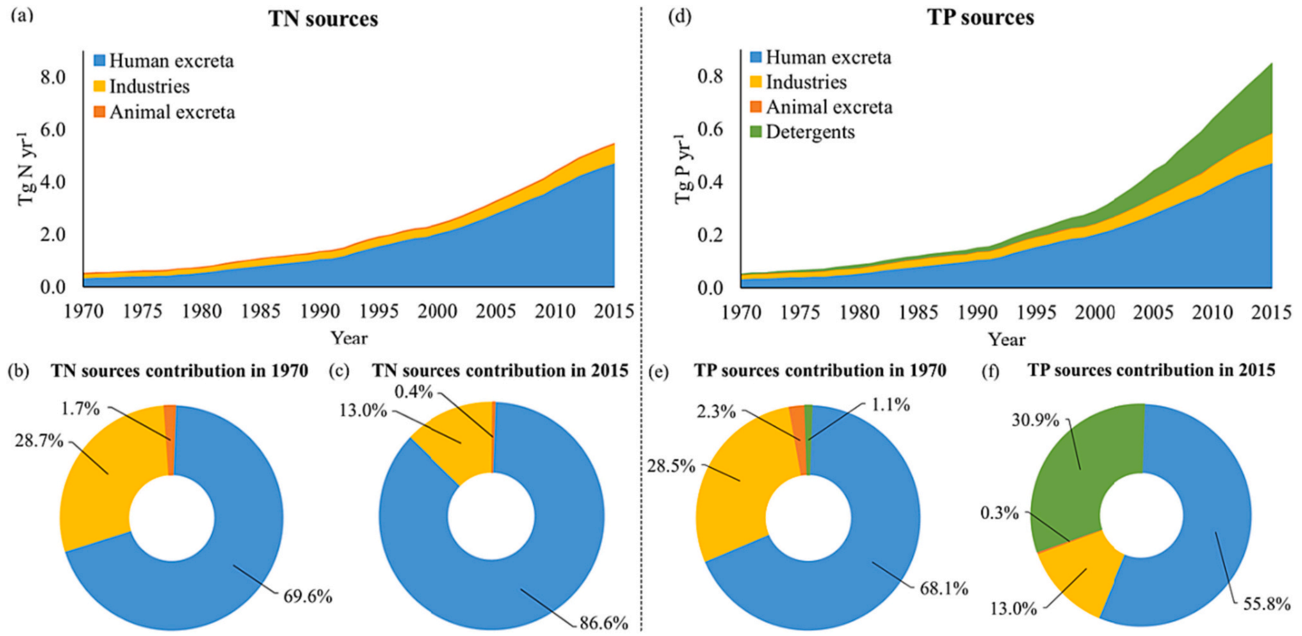


Fig. 4. (a) National N point source emissions prior to treatment from different sources for the period 1970–2015, and compositions of N sources in (b) 1970 and (c) 2015; (d) National P emissions prior to treatment from different sources for the period 1970–2015, and compositions of P sources in (e) 1970 and (f) 2015.

increased from 3 Gg to 0.4 Tg from 1970 to 2015. The contribution of rural areas to total wastewater discharge is 35 % for N and 43 % for P in 2015.

Although massive and rapid urbanization has taken place in recent decades, rural inhabitants still accounted for 43 % of the total population in 2015. The rural migrants working and living in urban areas increased from 30 million in 1989 to 277 million in 2015 (National Bureau of Statistics of China, 2015). Part of these rural migrants are still counted as rural population due to the Hukou (household) system of residency permit (Keung Wong et al., 2007), which may lead to overestimation of the contribution from rural areas and underestimation of the contribution from urban areas. Assuming that the rural migrants live in urban areas for half of the year implies an additional ~18 % nutrient emission (Fig. 2a, F_1) from urban populations in 2015.

The N discharge from rural areas (Fig. 6b) is increasing much more slowly than that from urban areas since 2000 (Fig. 6a). This slight increase in rural areas is the combined effect of the increase in protein consumption resulting from income growth and the decrease in the absolute number of rural inhabitants. Nevertheless, despite this declining rural population, the P discharge to surface water showed a rapid increase, which is mainly due to the increasing use of dishwashers and laundry detergents. In 2015, the contribution of rural areas to total wastewater discharge is 35 % for N and 43 % for P.

3.4. Sewer connection and treatment removal from urban areas

The sewer connection rates in the two largest municipalities in China, Beijing and Shanghai, both increased from 5 % in 1970 to 100 % in 2015 (Fig. 3). During the same period, the N and P removal in individual provinces showed different patterns (Fig. 3a, b) due to differences in their treatment technologies and average provincial WWTP capacities. For example, the nutrient removal in Beijing and Shanghai (red circles in Fig. 3a, b) is much more efficient than those in other provinces.

P is removed more efficiently than N in all the provinces (Fig. 3b). By aggregating all actual operational loading and technologies of the WWTPs present in the cities in three developed coastal provinces (Shanghai, Jiangsu, and Zhejiang) in 2014, their removal rates of P are 45 %, 21 % and 25 % respectively.

Even in the most developed city of Shanghai with a 100 % sewage connection rate, most wastewater treatment plants have adopted secondary technology. Therefore, the P and N treatment removal rates calculated in this study are lower than the 82 % for P and 63 % for N estimated in recent studies (Qi et al., 2020; Zhang et al., 2020), which ignored the considerable contribution of detergents and the actual capacity and loads of WWTPs. Another recent estimate (Chen et al., 2019) calculated one single year (2012) and did not capture the long-term spatiotemporal differences presented here because it used an overall sewer connection rate of 73 % in all urban areas and 1.8 % in rural areas, and ignored the differential changes over time in the various provinces. More details of the comparison with other studies are summarized in Table SI2 in SI6.

As a strategy for controlling wastewater pollution, the “Toilet Revolution” has become a buzzword in China in recent years (Cheng et al., 2018). However, relevant estimates are uncertain due to a lack of data on the removal rates in the different sanitation technologies used in rural areas, such as different septic systems and the unknown role of direct discharge of human waste to surface water.

Future developments should consider that the traditional sewage connection may not be the best option to mitigate eutrophication and the risk of wasting large quantities of economically accessible high-quality N and P (Cordell et al., 2013; Cordell and White, 2014; Van Vuuren et al., 2010), which according to our data add up to 4.7 Tg N (0.5 Tg P) for urban and 3.1 Tg N (0.3 Tg P) for rural areas in China in 2015. Sophisticated sewage systems with separate urine collection from households may be an efficient option in future urban planning to recycle nutrients. A number of biological, physical and chemical process separation systems have been developed for nutrient removal from urine (Maurer et al., 2006; Pronk and Kone, 2009; Wilsenach et al., 2007), (Zhang et al., 2014). This can substitute considerable amounts of N and P fertilizers and largely mitigate water pollution. Another option, especially for rural areas, is ecological sanitation, which combines improved sanitation with the recycling of nutrients (Langergraber and Muelleggera, 2005; Simha and Ganesapillai, 2017).

3.5. Total urban discharge to surface water and N:P ratio change

The urban N discharge to surface water increased 22-fold from 177 to 3908 Gg N yr^{-1} (For P, 29-fold from 20 Gg to 577 Gg P yr^{-1}) between

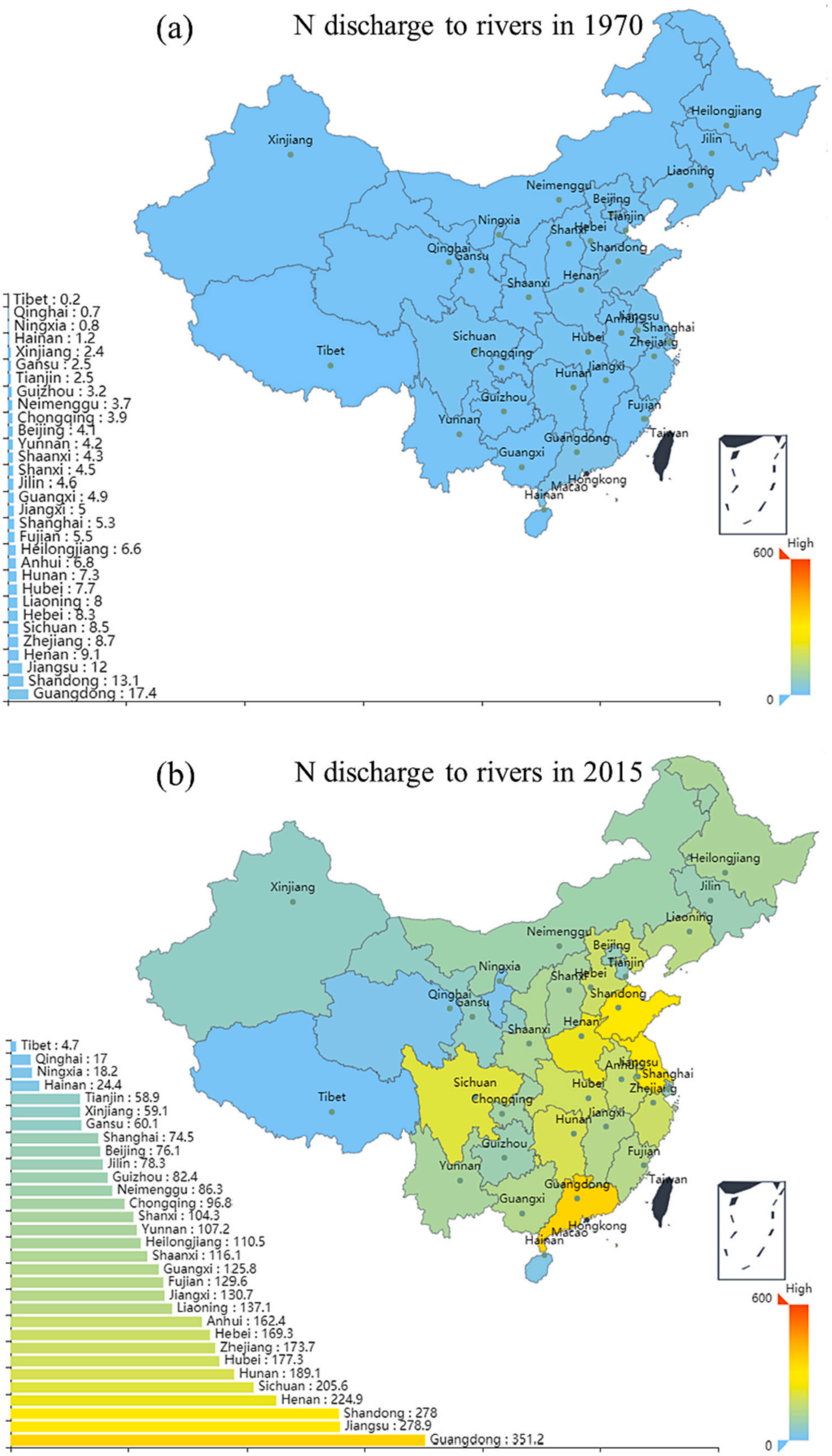
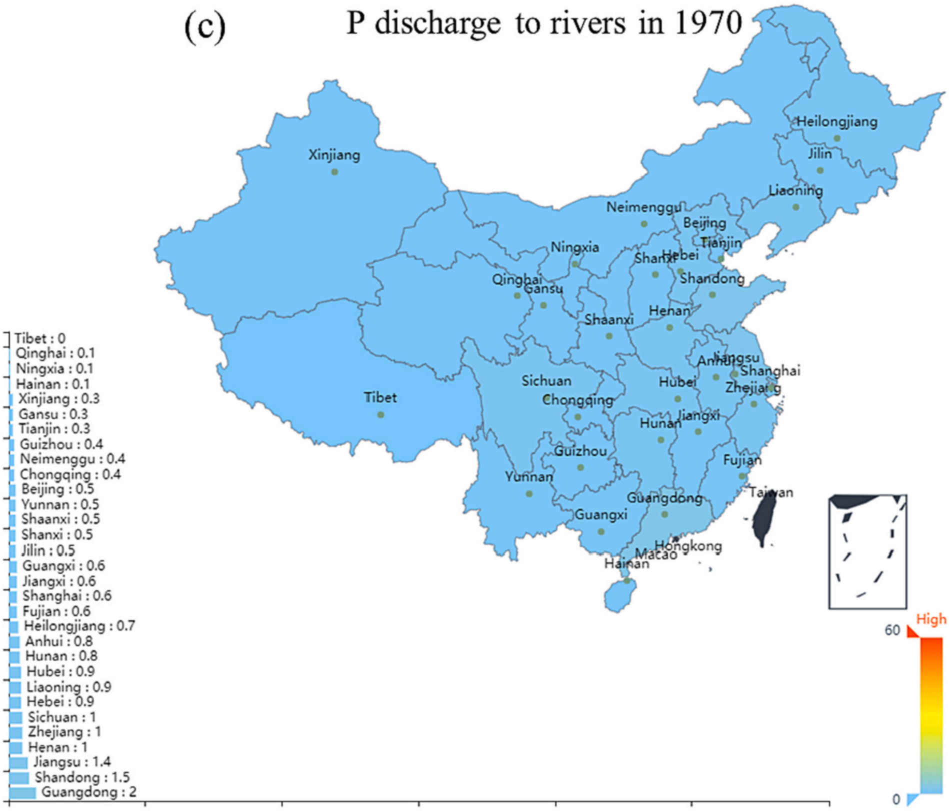


Fig. 5. Spatial distributions of nutrient discharge from urban areas to surface water for N in a) 1970 and b) 2015, and for P in c) 1970 and d) 2015. Unit is Gg yr^{-1} of N or P. Legend in the lower left corner indicates the absolute value of every province.



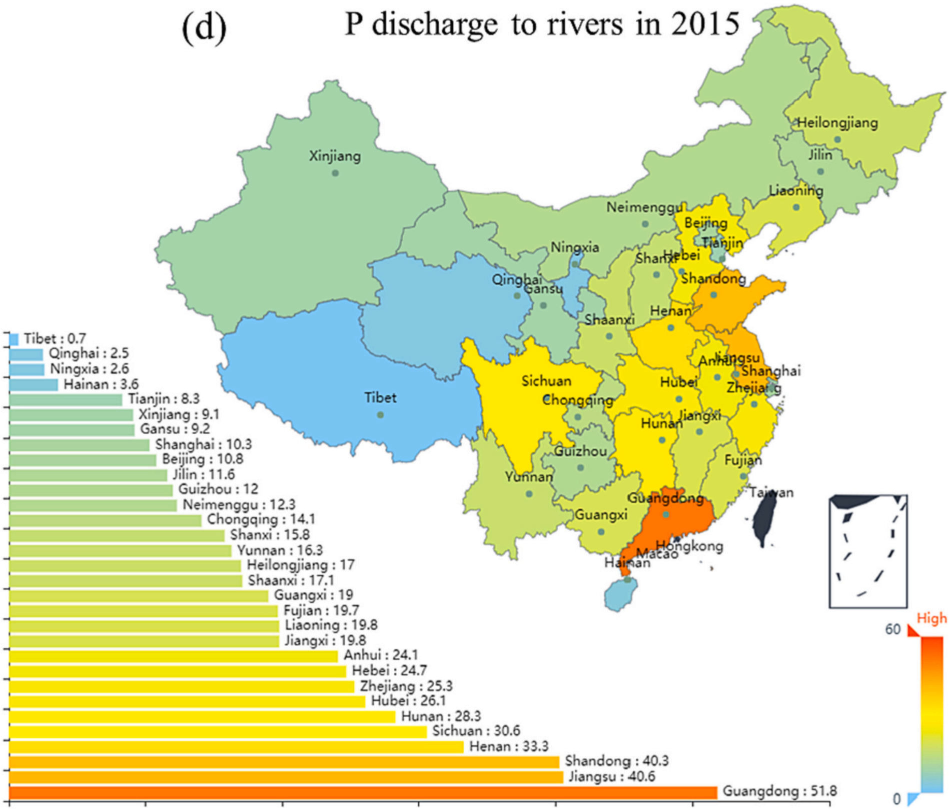


Fig. 5. (*continued*).

1970 and 2015 (Fig. 7a). For 2000, our estimated nutrient discharge of 1.7 Tg N and 0.2 Tg P (Fig. 7a) can be compared with a recently estimated discharge of 1.3 Tg N and 0.2 Tg P (Van Puijenbroek et al., 2019). The small difference is related to a scale issue, i.e. the result of two counteracting features: a higher sewer connection rate based on provincial data in the present study versus national data (Van Puijenbroek

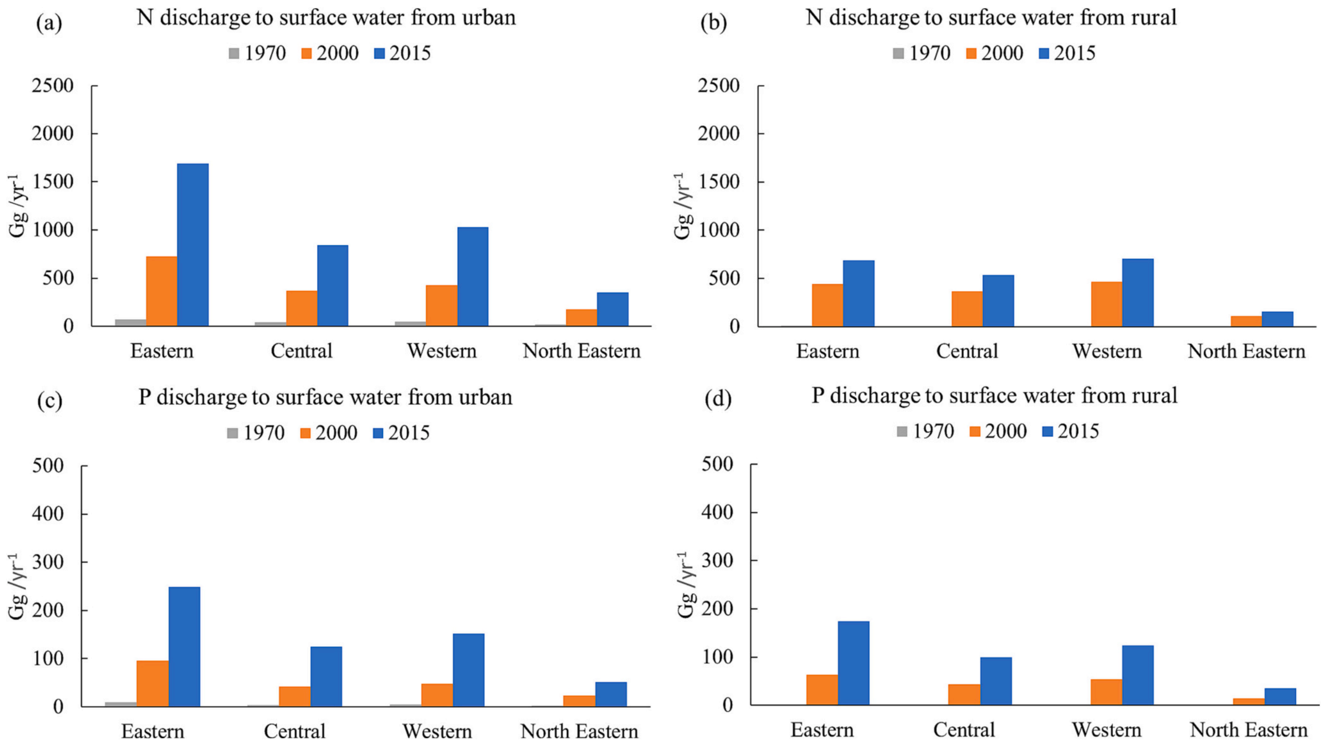


Fig. 6. N (a,b) and P (c, d) discharge to surface water from the urban (left) and rural (right) areas for the period 1970–2015.

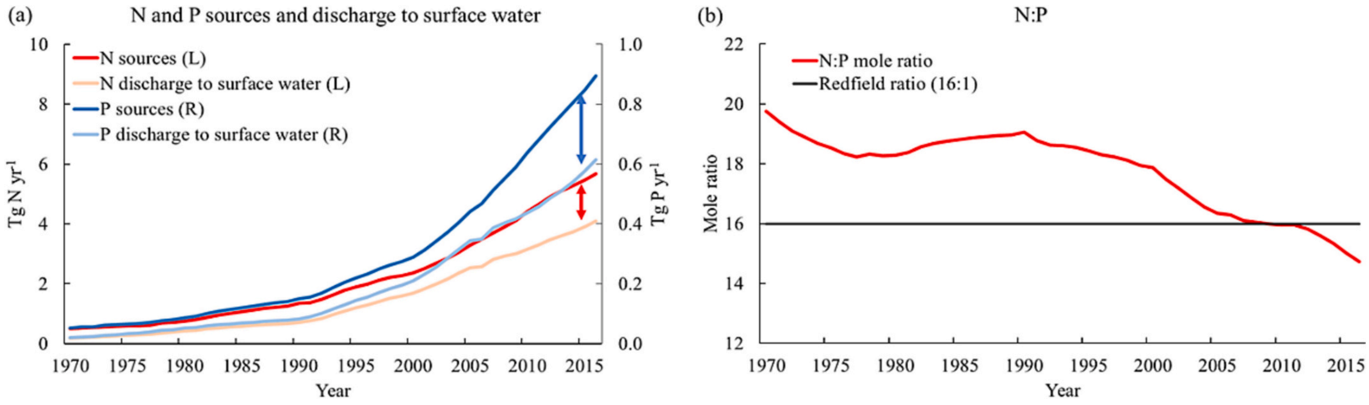


Fig. 7. (a) Urban sources of N (L, left) and P (R, right) (from human excreta, P detergents, animal excreta, and industries), and N and P discharge to surface water via WWTPs, and (b) N and P molar ratio.

et al., 2019), combined with higher removal rates (mostly secondary treatment in this study versus mostly primary treatment in Van Puijenbroek et al. (2019).

The substantial increase in the discharge of P relative to N is due to the rapid increase in the use of dishwashers and laundry detergents, which primarily occurred during 2000–2015 (Fig. 4d, f). The increase in WWTPs and upgrade of treatment technologies were not sufficient to counterbalance the increase in the P discharge from detergents.

In China, a local ban on the sale of P-based detergents started in 1999 in the Lake Tai basin, due to severe eutrophication problems (even disrupting the drinking water supply). Other regions like Lake Dianchi and Lake Chao and megacities like Shenzhen and Shanghai followed with similar bans on P-based detergents. However, there is no nationalwide ban on the use of P-containing detergents, and the existing national detergents criteria still allow for the production and sale of P-containing laundry and dishwasher detergents (Standardization Administration of China, 2012).

As a consequence of the increasing P discharge induced by the boosting detergent use, the N:P molar ratio in urban wastewater discharge decreased from 20 to 15 during the period 1970–2015 (Fig. 6b), even though the P removal efficiency is much higher than N in WWTPs (Fig. 3, Fig. SI3).

The WWTPs with tertiary and quaternary technologies currently account for 3 ‰ of total WWTPs (*Dataset S1.xlsx in SI2*). Even in the most developed cities like Beijing and Shanghai, with a 100 % sewage connection rate, most wastewater treatment technologies are secondary. In the developed western EU countries where tertiary WWTPs prevail, the P removal fractions in WWTPs range between 68 %–90 % (for N 49 %–85 %) (Hendriks and Langeveld, 2017; Longo et al., 2019; Oleszkiewicz and Barnard, 2006a; Van Puijenbroek et al., 2019). Therefore, our results suggest that the P and N treatment removal fractions in WWTPs of 81 % for P and 66 % for N reported by recent studies were overestimated (Qi et al., 2020; Zhang et al., 2020).

In regions with high rainfall, such as southern China, combined sewer overflows (CSO) can contribute a significant amount of nutrient fluxes from wastewater (Liao et al., 2015; Wu et al., 2016). It should be noted that the CSO is not simulated in our model system. CSO. Factors such as urbanization, urban water management and rainfall patterns impact CSO and their contribution to nutrient loadings (Tibbetts, 2005). Talamini et al. (2017) presented a systematic analysis of overflow causes and potential strategies of intervention, Separating the stormwater or the implementing the "Sponge City" could significantly prevent CSO. Another viable solution is real-time control (RTC) which uses sensor observations and numerical modelling to mitigating the CSO as well (Tian et al., 2022). Additionally, CSO pollution control should be fully considered as the primary aim of systematic control (Qiao et al., 2020). Given China's diverse conditions, targeted research is essential for understanding CSOs' impact on nutrient dynamics and developing effective management strategies (Burns et al., 2012).

3.6. *Sensitivity analysis*

The sensitivity analysis is helpful to rank the importance of the model variables and parameters, particularly those that can be controlled by targeted policies such as the development of wastewater treatment installations update of treatment technologies and regulating the use of P-based detergents.

The results of the sensitivity analysis (Table 2) are shown for 24 out of the 46 model input variables and parameters tested for their influence on N and P discharge to surface water. These 24 variables and parameters had significant SRC values, and the remaining 22 variables and parameters had an insignificant influence. The colored boxes in Table SI2 indicate SRC > 0.2 or SRC < -0.2, representing that their influence is larger than 4 % ($0.2^2 = 0.04$), which is considered to be an important and significant influence on the N or P discharge. The following variables and parameters exert an important influence on both N and P discharge: population both in rural and urban areas, protein consumption, sewer connection in urban, sanitation connection in rural, and access to tertiary treatment. Detergent use and the use of P-free detergents are important for exclusively P discharge. It is clear that particularly the shift from secondary to tertiary treatment has a large impact on the total N and P discharge to surface water.

4. Conclusions and policy implications

Compared with recent studies (see SI5), our analysis is a major step forward as it provides consistent data and spatial distributions of waste water nutrient flows in urban and rural areas in China, and long-term (1970–2015) changes (all the input data from the same year) in the drivers of the N and P emissions from point sources. Particularly the estimation of nutrient flows in rural areas, although there is uncertainty regarding the fate of the nutrients, contributes to a better understanding of the spatial distribution and magnitude of nutrient discharge to surface waters.

Our results aid policymakers and environmental managers in developing future strategies to reduce nutrient pollution from point sources. The main issues refer to (i) wastewater treatment technologies, (ii) regulations with regard to the use of P-free detergents and bans on P-based detergents, (iii) the importance of the rural regions in China, and (iv) the re-use of nutrients instead of wasting them.

(i) On the basis of the mass fluxes and spatial distributions of N and P from humans, animals and industries to surface water using the data on the capacity and removal efficiency for 4436 WWTPs, and based on the sensitivity analysis, our results indicate that to bend the increase in N and P discharge to surface water, the stagnant treatment technologies in WWTPs need to be incrementally improved from secondary to tertiary treatment over time, especially in the ten provinces in Eastern China.

(ii) The second recommendation is a ban on the production and sale of P-containing detergents via national legislation. This is based on our finding that the rapid increase of P discharge to surface water in rural and urban areas is primarily due to the lack of a nationwide ban of P-based detergents and the dramatic increase of the use of P-containing dishwashers and laundry detergents. The sensitivity analysis indicates that this will have a large and direct effect on the total P discharge.

(iii) The third recommendation is to consider the sanitation data for both urban and rural populations (*Dataset S1.xlsx in SI2*). Our results suggest a considerable contribution of the rural population to N and P pollution of freshwater environments in China, although the fate of the nutrients discharged through septic systems, double-container composting systems, biogas systems and sewage systems is poorly known.

The third recommendation is to consider the sanitation data for both urban and rural populations (*Dataset S1.xlsx in SI2*). Our results suggest a considerable contribution of the rural population to N and P pollution of freshwater environments in China, although the fate of the nutrients discharged through septic systems, double-container composting systems, biogas systems and sewage systems is poorly known.

(i) Comparison of WWTPs technologies in the representative provinces in different regions, rural household sanitation systems in China in 2002, N and P removal rate in WWTPs in different regions, comparison with other studies, comparison of the differences between this study and other studies, sanitation data for China, P content of detergents, and sensitivity analysis results. (ii) *Dataset S1.xlsx* includes all the 4436 WWTPs, the total 42 technologies in China, and the cumulative capacity and average loading in every province, Sanitation data of China, detergent P deduction Scenarios, rural and urban population. (iii) Movie of P load to rivers from China's wastewater. Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.168091.

CrediT authorship contribution statement

XL, AHWB, PVP and AFB designed the research and developed the model, XL collected the data for China, XZ distinguished the WWTPs technologies, JW contributed to the data collection, WJVH contributed to the visualization process, XL and AFB wrote the text. All co-authors reviewed and commented on the manuscript.

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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