GEO4-6004 - Master Thesis Water Science and Management

Exploring Nutrient Dynamics in the rivers Rhine (Europe) and Pearl (China) using the IMAGE-GNM model for the period 1900 – 2000

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Abstract:

Nitrogen (N) and phosphorus (P) dynamics in the Rhine River and the Pearl River from 1900 to 2000 are modelled using the IMAGE_GNM model. The modelling results illustrate the spatio-temporal changes of N and P delivery to rivers, exports to mouth, the contributions of nutrient sources and the in-stream retention. The modelled TN and TP exports to the sea from the Rhine River increased from 190 to 388 Gg yr^{-1} and from 22 to 51 Gg yr^{-1}, respectively, between 1900 and 1970. With policies and measures carried out to limit the nutrient load, N and P from agricultural source and sewage wastewater decreased distinctly during 1970 - 2000. The effect of improving wastewater facilities has a quick effect on cutting down the nutrient load. But the legacy effect of the N and P in soil and groundwater delays the visible effect of agricultural reduction measures. Without regulatory on agricultural fertilizer use and proper wastewater treatment, the nutrient transport in the Pearl River increased over the entire 20^{th} century. N and P exports to mouth increased up to 960 Gg yr^{-1} and 75 Gg yr^{-1} till 2000. Measures were taken to reduce nutrient load in the 21^{st} century in the Pearl River basin, mainly focusing on improving the wastewater facilities. The modelling results show that agricultural N and P account for 80% and 89% of the total loads to the Pearl River, respectively. Therefore, we recommended that regulating agricultural N and P loads should be the top priority for the Pearl River.

Keywords: Nutrient delivery; Nutrient export; Nutrient sources; Pearl River; Rhine River

1. Introduction

Nitrogen (N) and Phosphorous (P) are essential nutrients for plants. Excessive N and P released into surface water by human activities can accelerate aquatic plant production (eutrophication) and alter riverine-estuarine nutrient stoichiometry which may lead to harmful algal blooms (HABs) (Billen & Garnier, 2007). In the 20th century, global N and P loads to streams and rivers have increased rapidly from 24 to 64 Tg yr^{-1} and from 5 to 9 Tg yr^{-1}, respectively (Beusen et al., 2016). The resulting HABs have been a problem in many river basins and coastal areas around the world (Anderson et al., 2002).

The Rhine river basin is a good example, where nutrient reduction policies were implemented to effectively control the nutrient load and maintain a sustainable aquatic environment. Tracing back to the 1850s, the water quality of the Rhine river was seriously deteriorated due to the increase of agricultural fertilizer runoff and wastewater flows from household and industries (Fritjers et al., 2003). The water pollution problem continued intensifying in the first half of 20th century (Wieriks & Schulte-Wülwer-Leidig, 1997). To mitigate eutrophication, multiple measures were taken to reduce N and P loads, including improving wastewater treatment systems and limiting the amount of manure and fertilizer use in agriculture activities (Fritjers et al., 2003). Decades of efforts have distinctly improved the water quality of the Rhine River. The annual average total nitrogen (TN) concentration in the German-Dutch border Bimmen/Lobith was around 2.3 – 2.6 mg/L during 2010 - 2013 which is close to the EU standard (<2.5 mg/L) (ICPR, 2015). The annual total phosphorus (TP) concentration is still exceeded in many monitoring stations (with the standard of <3mg/L for surface water) (ICRP, 2015). However, the experience of the Rhine can still provide valuable lessons to other polluted rivers around the world.

The Pearl River, located in south China, has been facing the similar predicament that the Rhine ever faced. With the massive economic growth and urban development, excessive release of nutrients into the Pearl River estuarine frequently led to red tides in this area (Huang et al., 2003; Jin et al., 2005). In the coastal water, the average TN and TP concentrations was 7 mg/L and 8 mg/L between 2008 and 2010 (Zhang et.al., 2011). As the third largest river in China crossing 4 provinces, the Pearl water quality now urgently requires pollution control. Similar to the Rhine, nutrients from sewage systems and agriculture are thought to be the major causes for the N and P loading (Huang et al., 2003; Wong & Wong, 2004). To better learn from the nutrient management experience in the Rhine River basin, here we compare the historical nutrient dynamics between the two river basins.

Hydrographic information and monitored nutrient data for the Rhine basin were used to estimate the N and P export to the North Sea in previous research (Gömann et al., 2005; Hartmann et al., 2011; Prasuhn & Sieber, 2005). Several models were developed to analyze the detailed change of nutrient load and pollution sources in the Rhine River basin (Loos et al., 2009; Wit, 2001). For the Pearl River, the availability of nutrient monitoring data is a serious limitation. Empirical studies were based on measured concentrations of N and P in specific forms, most of which were conducted within the estuary area in a relatively short period (Dai et al., 2008; Huang et al., 2003; J. Zhang et al., 1999). The Soil & Water Assessment Tool (SWAT) model allows for analyzing nutrients in surface water and their sources on a monthly basis for relatively small-scale watersheds (Y. Wu & Chen, 2009). It has been applied to a branch of the Pearl river – the East River, and the delta area. However, the demand for the detailed input data hinders its implementation to the entire Pearl river basin. Global NEWS-2 is a modelling tool designed for a global scale and was applied to the Pearl river basin (Strokal et al., 2015) to analyze nutrients exported to mouth and their sources for two years (1970 and 2000). Global NEWS-2 is a regression model that lumps river basin data to estimate the load at the river mouth, but it fails to improve our understanding of biogeochemical processes and their drivers changing through time.

The previous studies lack spatiotemporal scales to analyze inter-annual patterns of rival nutrients sources/exports under changing human pressures for the large watersheds. The impact of nutrient mitigation strategies probably strongly depends on the location within the river basin. This is because nutrient retention depends strongly on the travel time and thus also on the presence of reservoirs which increase water retention. We therefore need a spatially explicit approach for this comparison of the rivers Rhine and Pearl. The spatially explicit, distributed process-based model IMAGE-GNM is used to evaluate the changes in the various N and P sources retention in different water bodies and exports to the coastal area from 1900 to 2000. This provides a comprehensive view of the N and P dynamics in the past. The findings can be used to further analyze the impact of nutrient-reduction policies and measures in the Rhine River basin. Based on the comparison results of nutrient dynamics of the two River basins, Rhine regulations/laws/water quality standards can be analyzed to provide implications to the environmental management in the Pearl River region.

The research questions of my study are as follows:

Q1: How did N and P in the Pearl River and Rhine River change from 1900 to 2000?

Sub1: How accurate are the modelling results compared to the monitored data for the two rivers?
Sub2: How did the nutrient sources, retention and export to the coastal seas change in the two rivers for the period 1900-2000?
Sub3: What are the differences and similarities between nutrient dynamics in the two rivers, particularly the N and P legacies?

Q2. What are the implications for the water management in the Pearl River basin?

Sub4: How did the measures taken in the Rhine River basin effectively reduce N and P concentrations in surface water?
Sub5: To ensure healthy aquatic ecosystem in the Pearl River basin, what and where are the most important nutrient sources to control for N and P?
Sub 6: Are the Rhine measures suitable for the Pearl River basin? What are the foreseeable barriers? What shall we expect if similar measures were taken?

2. Material and Method

2.1 Study area

The Rhine River is the largest river located in the west Europe and covers an area of 160,000 km^2 (Figure 1 a). It flows through 6 countries (Switzerland, Liechtenstein, Austria, France, Germany and Netherlands) and drains into the North Sea. The mean annual discharge of the Rhine River is 66 km^3 yr^{-1} at the Bimmen/Lobith station (Wang et al., 2005). It is a densely populated river basin with 45 million inhabitants in 2001 (Wit, 2001). The region is well developed and highly impacted by diverse human activities, such as agriculture and industry (Loos et al., 2009).

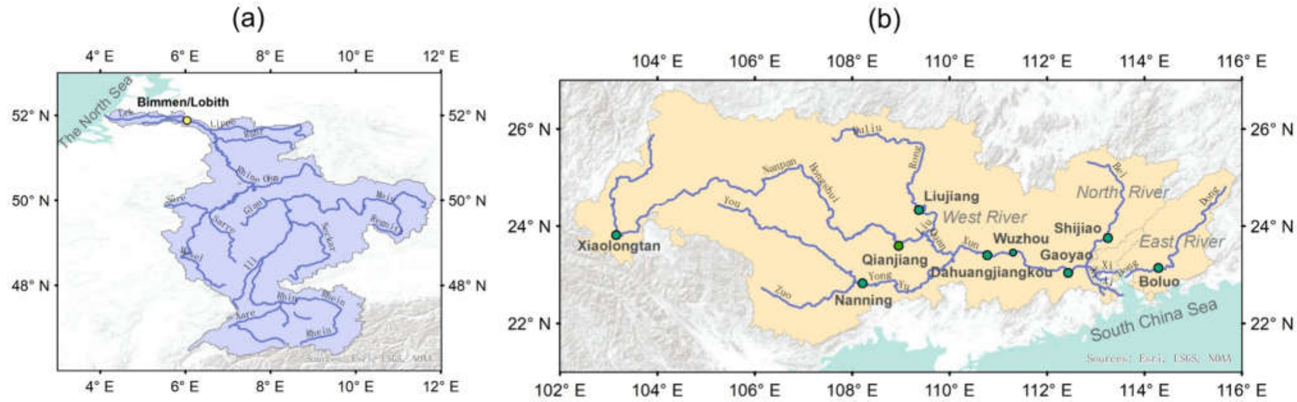

Figure 1. Map of the Rhine River (a) and the Pearl River (b) and the location of the monitoring stations

The Pearl River, located in South China, is much larger than the Rhine River. The whole basin covers 440,000 km2 with an annual discharge of 336 km3 yr-1 (sum of discharges of Gaoyao station, Boluo station and Shijiao station) (Pearl River Water Resources Committee, 1991). The Pearl River drains into the South China Sea; it has 3 main tributaries – the West River, the North River and the East River (Figure 1 b). The North River and the West River merge in the Pearl River delta and drains into the South China Sea. The East River flows into the South China Sea in a separated river mouth from the other two tributaries. The whole Pearl River flows through 4 provinces (Yunnan, Guangxi, Guizhou and Guangdong). In 1964, 100 million inhabitants lived in the Pearl River basin, which massively increased to 230 million by 2009 (C. S. Wu et al., 2012). The Pearl River serves as the major water supply for drinking water, agriculture, navigation and hydropower generation in southern China (Q. Zhang et al., 2014).

Table 1. Basic characteristics of the Rhine River and the Pearl River

	The Rhine River	The Pearl River
Basin area (km^2)	160,000	440,000
Annual discharge (km^3yr^{-1})	66	336
Related countries/provinces	6 countries (Germany, Switzerland, France, Netherlands, Austria and Liechtenstein)	4 provinces (Yunnan, Guangxi, Guizhou and Guangdong)

2.2 Model description

The Integrated Model to Assess the Global Environment – Global Nutrient Model (IMAGE-GNM) is a grid cell based model to simulate the explicit N and P delivery from land to sea and the in-stream retention in rivers, lakes and reservoirs. It can be used to analyze the yearly nutrient dynamics in worldwide rivers with a spatial resolution of 0.5 by 0.5 degree (Beusen et al., 2015). Nutrient inputs and outputs in agricultural systems are based on statistical information, which is from provincial data for China and national statistical data for the Rhine (for details see (Bouwman et al., 2017, 2005). In agricultural systems, the nutrient soil budget (all inputs minus outputs) represents the potential nutrient loss to the environment through air emissions, surface runoff, and leaching (Figure 2). For non-agricultural areas, nutrient sources include atmospheric deposition, biological N fixation in natural ecosystems; further direct sources are urban wastewater and aquaculture, and vegetation in flooded areas, as described in detail in (Beusen et al., 2015).

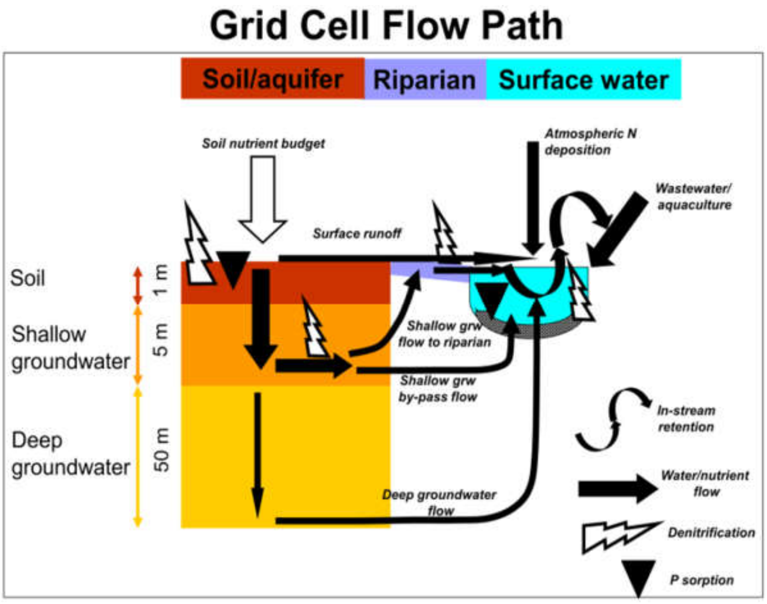

Figure 2. Scheme of water and nutrients flows within one 0.5 by 0.5 degree grid cell (Liu et al., 2018)

Figure 2 illustrates the N and P sources and the transport pathways and reaction processes simulated in each grid cell. There are 3 layers in the vertical direction, which are soil, shallow groundwater and deep groundwater. The lateral direction also includes three compartments: soil/aquifer, riparian zone and surface water. The yearly land use profile and the climate data are simulated by the IMAGE model. The PCR-GLOBWB model provides the hydrological data, including the water flux direction, residence time in water bodies, lake areas, flooding areas and information of reservoirs. The IMAGE-GNM model calculates the N and P input from different sources, including groundwater under natural vegetation and agricultural lands, surface runoff over natural or agricultural lands, rock weathering, aquaculture and sewage wastewater. Reactions including denitrification, sedimentation, absorption etc. are modeled in related land types, soil layers and water bodies. Combined with the hydrological data, the nutrient flows in rivers and the in-stream retention can be simulated explicitly. The detailed calculation methods for each process are introduced in APPENDIX1.

3. Results

3.1 Model validation

RMSEs (Root Mean Square Errors) are calculated based on the modelled results and the observed discharges and concentrations at 1 monitoring station in the Rhine and 9 stations in the Pearl. Figure 3 shows the comparison results of the observed discharges, N concentrations and P concentrations with the modelled results for 3 representative stations. More detailed validation results for all the stations are listed in APPENDIX 2.

For the Pearl River, according to the Figure A2 (d-i) the model performance of discharges varies among different stations. Discharges are well modeled for the stations of Boluo (RMSE = 17%), Gaoyao (RMSE = 11%), Qianjiang (RMSE = 11%), Dahuangjiangkou (RMSE = 18%), Liujiang (RMSE = 25%), Nanning (RMSE = 36%), Shijiao (RMSE = 33%) and Wuzhou (RMSE = 11%). The model overestimates the discharges in the stations of Xiaolongtan (RMSE = 140 %). The RMSE among all the stations for the discharge of the Pearl river is 19%. Limited by the data sources, concentrations of N and P were collected from published studies to validate the model performance for the TN and TP concentrations in the Pearl river (Figure 3 m). The simulated trend of TN concentrations matches well with the observations in Boluo station (RMSE = 19% see Figure A2 m) and Gaoyao station (RMSE = 40%, see Figure A2 n in APPENDIX 2). The RMSE values for TN and TP concentrations of the Pearl river are 39% and 132% respectively (Figure 3 n, o).

With the complete data set from 1970 to 2000 at the Bimmen-Lobith station, the model performance for the Rhine River is better than that of the Pearl. The yearly dynamics trend of discharge, TN concentration and TP concentration are well modelled according to Figure 3 (a, b, c). The RMSEs for the discharge, TN concentration and TP concentration are 14%, 15%, 25% respectively.

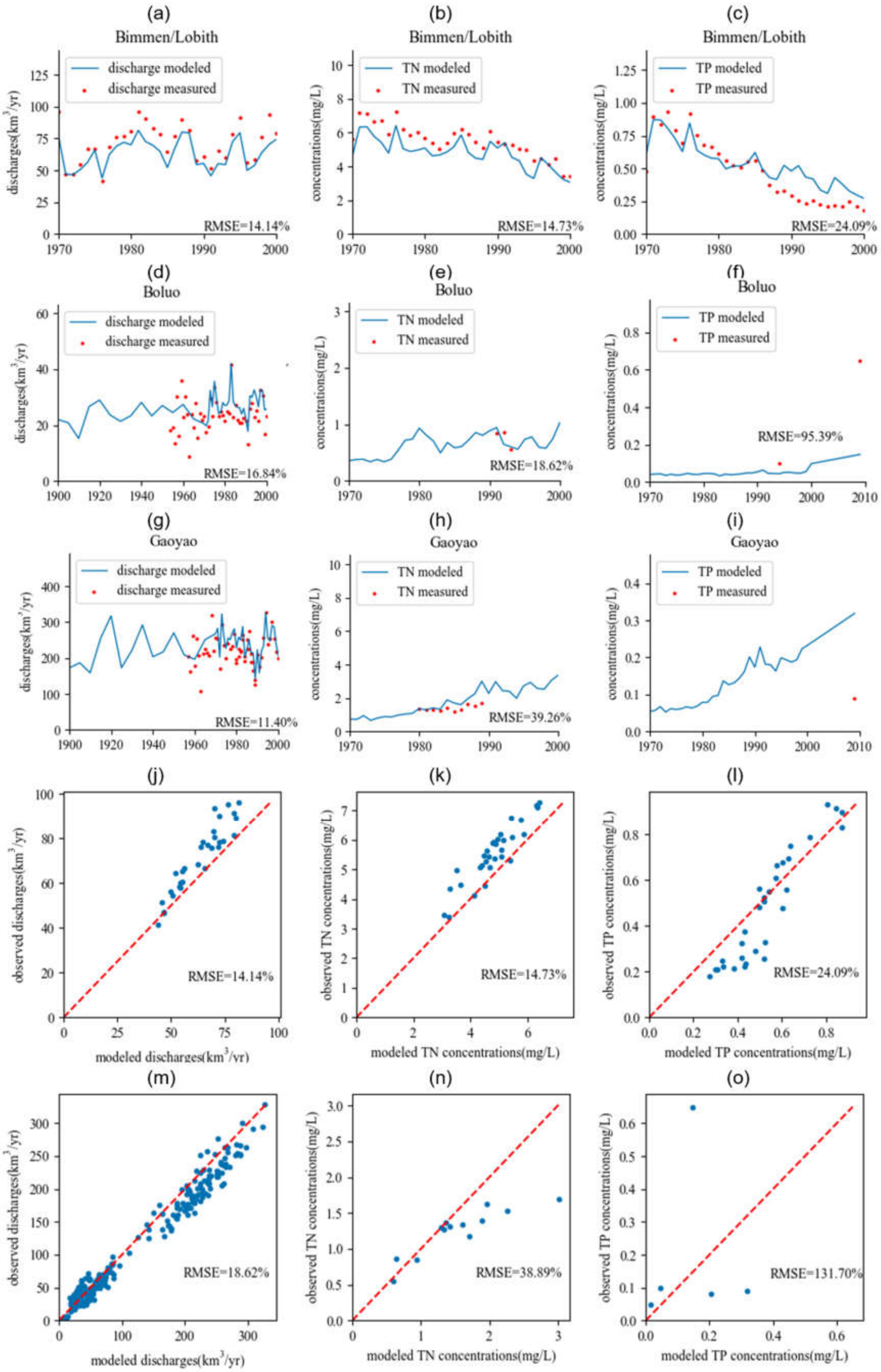

Figure 3. Comparison of measured (red spots) and modelled (blue lines) discharge / N concentration / P concentration in the Rhine River(a-c) and Pearl River (d-i); relationships between observed value and modeled value in the Rhine River(j-l) and Pearl River (m-o)

3.2 Nutrient dynamics in the Rhine River from 1900 to 2000

Figure 4 summarizes the modelling results of the N and P dynamics in the Rhine River basin from 1900 to 2000. TN and TP budget substantially increased by a factor of 3.5 (290 to 1064 Gg N yr^{-1}) and 8-fold (18 to 164 Gg P yr^{-1}), respectively from 1900 to 1980. Meanwhile, TN and TP delivery to rivers and export to mouth were doubled. From 1980 onwards, the agricultural TN and TP budgets started to decrease steeply. The TN budget decreased to 850 Gg yr^{-1} in 1990, then kept fluctuating with a slight downtrend and dropped to 670 Gg yr^{-1} in 2000. The TP budget continuously decreased to a negative value of -21 Gg yr^{-1} till 2000. However, TN and TP delivery to rivers and export to mouth decreased slowly. TP delivery to rivers and export to mouth became higher than the TP budget (close to 0) after 1995.

Figure 4 (c, d) shows the N and P loads to surface waters from different sources in the Rhine River from 1900 to 2000. TN from sewage wastewater increased from 120 to 190 Gg yr^{-1} between 1900 and 1940. Then it stabilized till 1970. In the meantime, TP from sewage wastewater increased about 3 folds rapidly from 12 to 35 Gg yr^{-1}. TN and TP from wastewater decreased significantly from 1970, which dropped to 70 Gg yr^{-1} and 7 Gg yr^{-1} respectively in 2000. TN from surface runoff and groundwater in agricultural lands increased up to 81 Gg yr^{-1} and 180 Gg yr^{-1}during 1930 – 1990 and then decreased to 66 Gg yr^{-1} and 100 Gg yr^{-1} till 2000. Similarly, TP from surface runoff in agricultural areas increased from 13 to 27 Gg yr^{-1} during 1900-1970, then dropped to 19 Gg yr^{-1} in 2000. TN from groundwater in natural lands rose slightly from 1900 to 2000 while TP from weathering and surface runoff over natural lands was relatively stable.

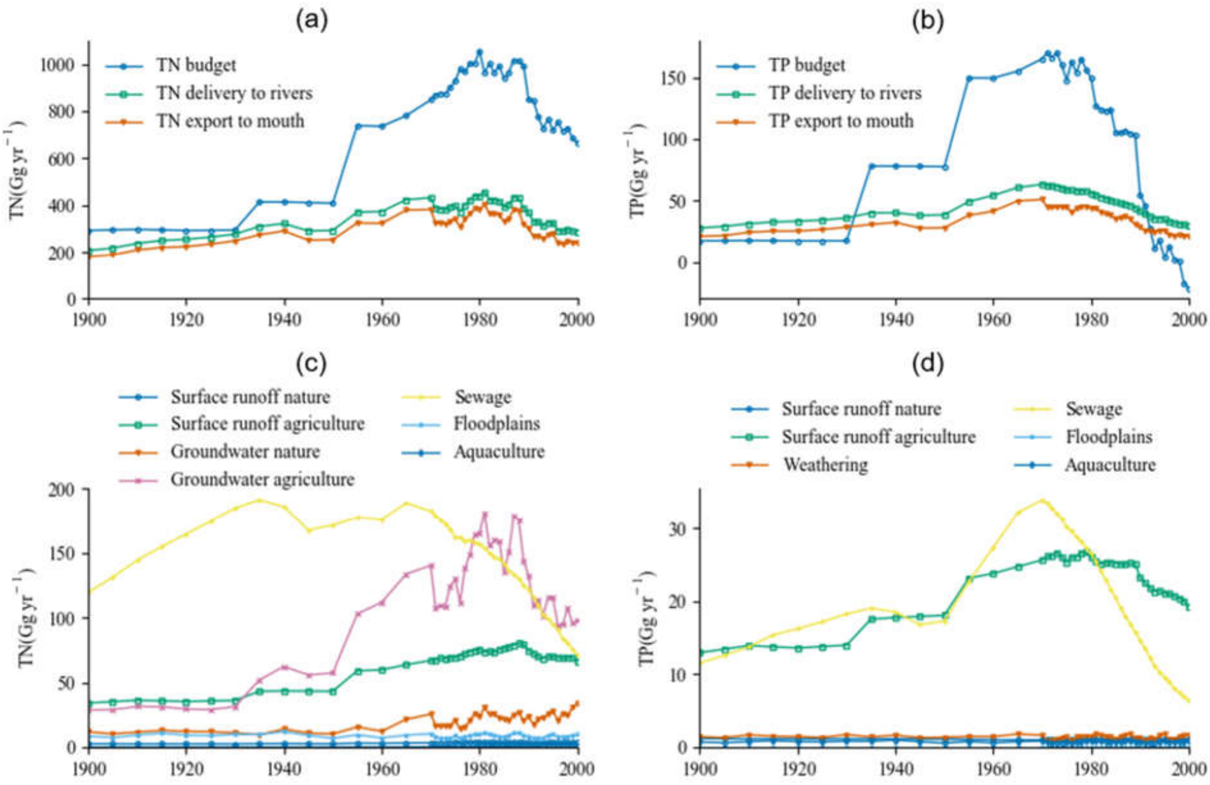

Figure 4. (a) N and (b) P soil budget, loads to rivers and exports to mouth for the Rhine River basin for the period 1900-2010; TN and TP delivery to surface water from different sources for the period 1900-2000 for the whole basin (c, d)

The changes in retention in the Rhine basin are shown in figure 5. TN retention increased from 27 to 65 Gg yr^{-1} during 1900-1990 and decreased to 48 Gg yr^{-1} till 2000. TP retention increased from 7 to 18 Gg yr^{-1} during 1900-1970, followed by a downward trend, and reached about 8 Gg yr^{-1} in 2000. TN and TP basin retention fractions were relatively stable, around 18% and 24%, respectively. The retention fraction from reservoirs initially rose in 1930 and reached up to 4% for both N and P. Retention fractions of lakes and streams were relatively stable, at around 70% and 30%, respectively.

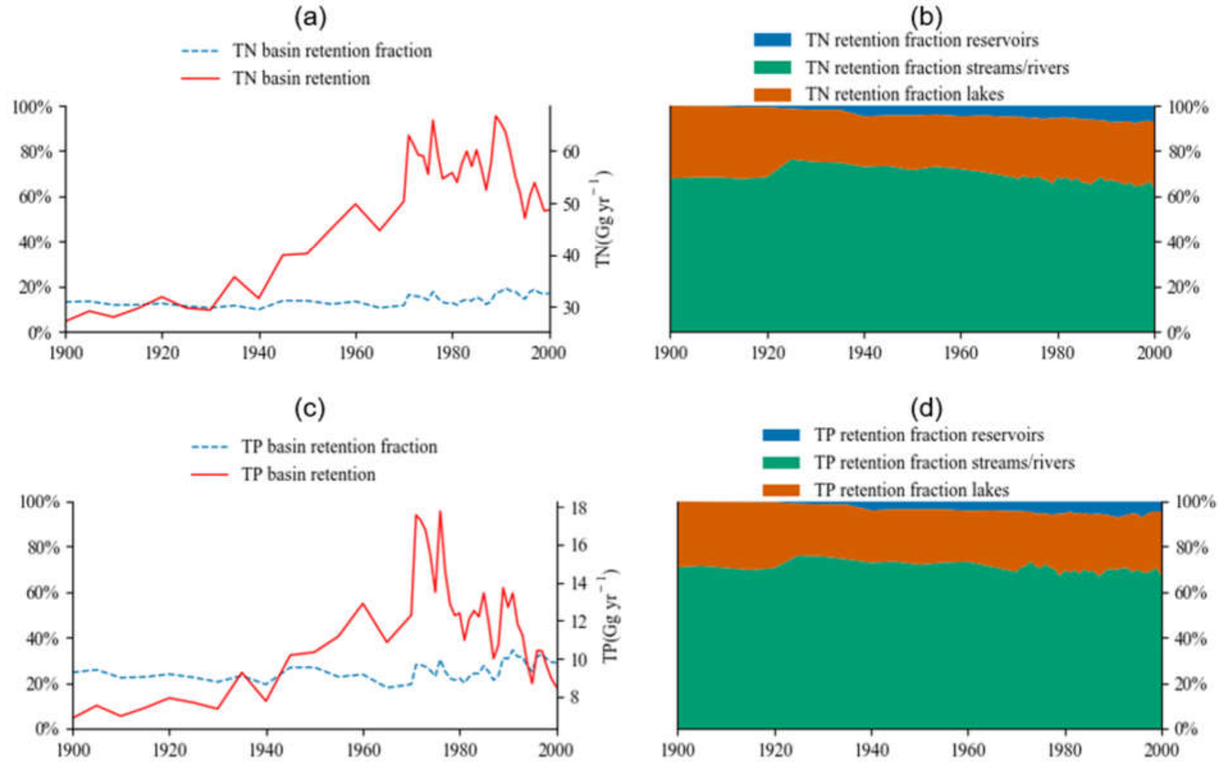

Figure 5. (a) Total N retention and N retention fraction, and (b) fraction of retention in streams/rivers, lakes and reservoirs in the Rhine River basin for the period 1900-2000; (c) total P retention and P retention fraction, and (d) fraction of P retention in streams/rivers, lakes and reservoirs in the whole basin for the period 1900-2000

3.3 Nutrient dynamics in the Pearl River from 1900 to 2000

We subdivided the Pearl river basin into two parts, the west part (the North River and the West River) and the east part (the East River), based on its hydrological characteristics. The soil N budget of the whole Pearl basin increased by more than a factor of 4 (from 423 to 1928 Gg yr^{-1} between 1900 and 2000) (Figure 6a, b). Meanwhile, the TN delivery to rivers and mouth increased from 147 to 1133 Gg yr^{-1} and 100 to 896 Gg yr^{-1}, respectively. Likewise, the TP transport in the Pearl River basin experienced a sharp rise in the period 1900-2000. The TP soil budget increased from a slight negative value of -5 to 262 Gg yr^{-1}. The TP delivery to rivers and export to mouth increased by a factor of 6.4 (from 18 to 115 Gg yr^{-1}) and 6 (from 11 to 66 Gg yr^{-1}), respectively. The results for the west and east parts of the Pearl basin are in APPENDIX 3.

Figure 6 (c, d) shows the changes in N sources for the Pearl River in 1900-2000. The dominant N source was groundwater in natural areas till 1960, contributing to 50-60% of the TN load to streams. Afterwards, N load from groundwater and surface runoff in agriculture areas became increasingly important. N load from groundwater in agriculture areas increased from 47 to 670 Gg yr^{-1}. The share of agricultural N load (sum of surface runoff and groundwater in agricultural lands) increased by up to 85 % till 2000. Besides natural and agricultural soils, sewage wastewater and aquaculture are growing N sources since 1980. TN from wastewater and TN from fresh water aquaculture increased from 23 to 58 Gg yr^{-1} and from 1 to 14 Gg yr^{-1} respectively for the period of 1980–2000. The increasing trend of sewage N load in the east part of Pearl was dramatic, which rose from 3 to 12 Gg yr^{-1} between 1999 and 2000 (Figure A3.1 (e)). The share of TN from sewage wastewater and aquaculture increased by up to 23% and 10% for the east part of Pearl in 2000.

For the Pearl River, the main P sources were weathering, surface runoff from agriculture and surface runoff from nature areas before 1960, accounting for around 30%, 35% and 20% of the TP load to river. From 1960 onwards, P from agriculture increased drastically from 10 to 87 Gg yr^{-1}. The share of TP from agriculture reached up to 83% in the year of 2000. The P from surface runoff over natural lands and aquaculture decreased during 1960 - 2000. Meanwhile, TP from weathering was stable in the range of 5-10 Gg yr^{-1}. TP from wastewater increased from 3 to 8 Gg yr^{-1} between 1980 and 2000, accounting for 9% and 8% of the TP load to the rivers. Similar to the contribution rise of TN from wastewater, a dramatic increase of TP from sewage in the east part between 1999 and 2000 can be seen in Figure A3.1(h).

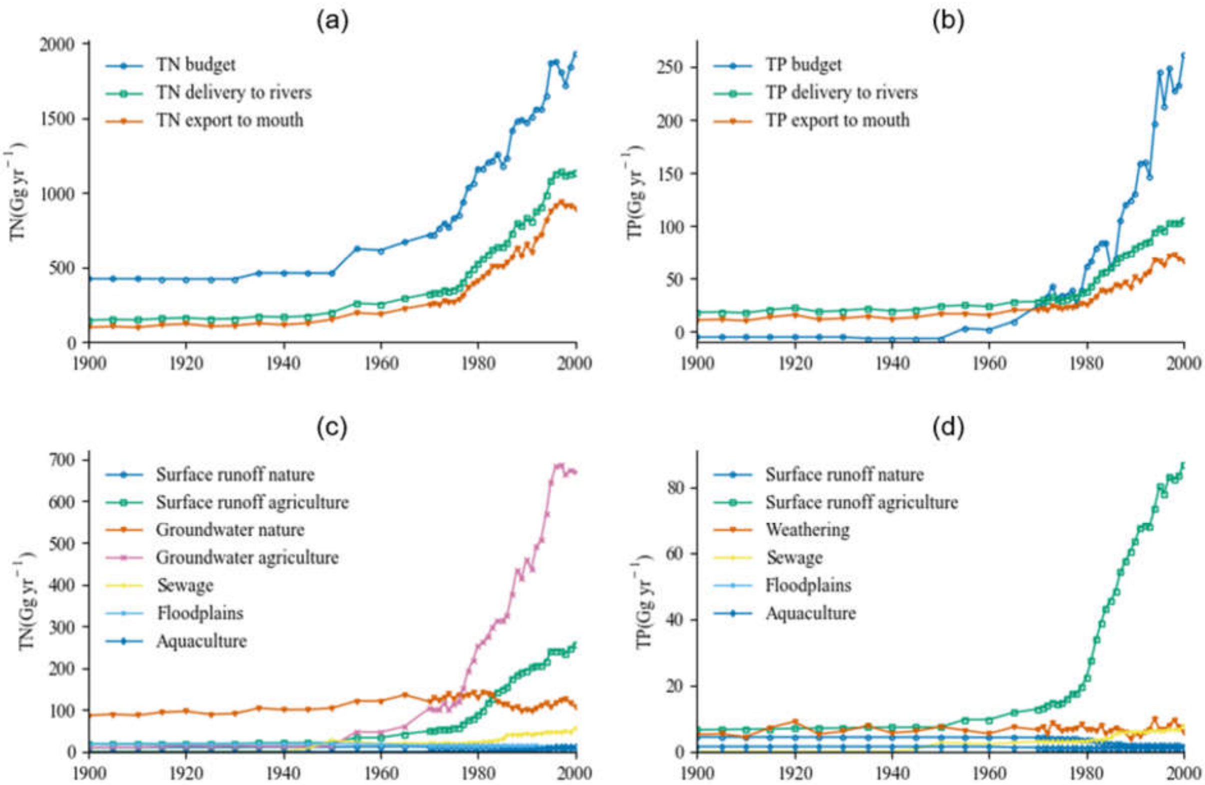

Figure 6. (a) N and (b) P soil budget, loads to rivers and exports to mouth for the Pearl River basin for the period 1900-2000; river TN and TP delivery to surface water from different sources for the period 1900-2000 for the whole basin (c, d)

TN and TP retention in whole Pearl River basin increased from 65 to 238 Gg yr^{-1} and from 8 to 39 Gg yr^{-1}, respectively, from 1960 to 2000; the retention fraction fluctuated around 30% and 40%, respectively (Figure 7 a, c). The retention fraction in reservoirs increased as more reservoirs were established in the basin (Figure 7 (b, d)). There were no reservoirs before 1960, and the retention fraction started to increase after that year to 30% and 20% for N and P in 2000. The retention fraction in reserviors is even higher in the east part of Pearl, where it reached 75% for TN retention and 65% for TP (calculated from Figure A3.2 f, h).

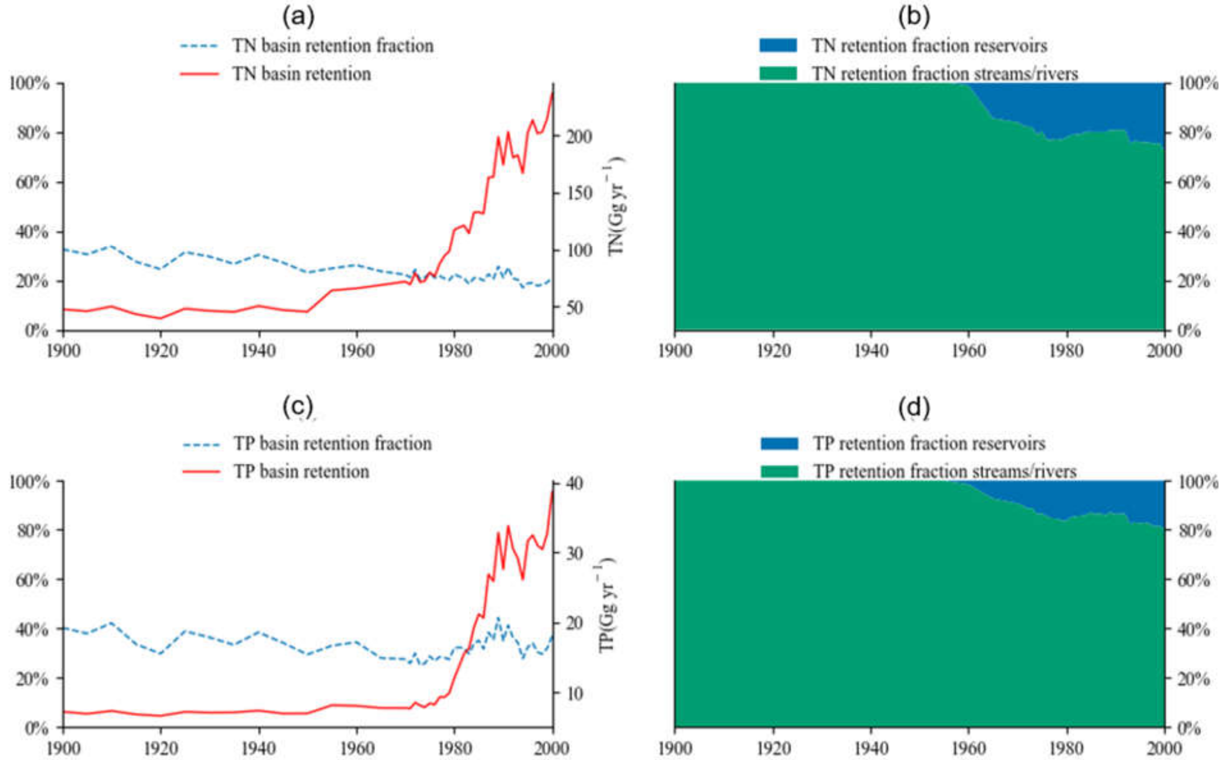

Figure 7. (a) Total N retention and N retention fraction, and (b) fraction of retention in streams/rivers, lakes and reservoirs in the Pearl River basin for the period 1900-2000; (c) total P retention and P retention fraction, and (d) fraction of retention in streams/rivers, lakes and reservoirs in the whole basin for the period 1900-2000

4. Discussion

4.1 Development of the nutrient management in the Rhine River basin

The modelled results illustrate the massive change in N and P transported in the Rhine river during 1900 – 2000. According to the results of N and P loads to the river network, the changes can be divided into 3 phases: (1) Accumulation phase: 1900 – 1970; (2) Stabilization phase: 1970 – 1980; and (3) Depletion phase: 1980 – 2000.

During the accumulation phase, the most important nutrient source was sewage wastewater. From 1900 to 1930, N from wastewater increased from 120 to 180 Gg yr^{-1}. In 1930, wastewater contributed to 65% of the TN and 51% of the TP loads to rivers. N from sewage wastewater began to stabilize after 1930. P from sewage wastewater kept increasing and peaked at 33 Gg yr^{-1} 1970. This is because chemical industries and refinery developed fast all over west Europe. Most of these factories were located in the Rhine River basin, especially in German part (Wieriks & Schulte-Wülwer-Leidig, 1997). Without proper wastewater treatment facilities, the sewage wastewater emitted from industry and domestic use were directly released into the rivers, in which industrial wastewater was the largest N and P source. From the modelling results that shows the spatio-temporal change of dominant nutrient source (Figure 8 c, d), the locations where point sources dominate in 1970 coincide with the main industrial area, pointed out in many other researches (Mostert, 2009; Wendland et al., 2005). At the same time, to increase agricultural productivity, the expanding application of chemical fertilizer and manure led to the sharp increase of the TN and TP soil budgets (Mostert, 2009). This stimulated more N and P delivery to rivers through surface runoff and groundwater flow. Our model indicated that agriculture had contributed to 52% and 41% of the TN and TP loads to rivers until 1970. The N and P loads in most part of the Rhine are dominated by groundwater and surface runoff in agriculture areas in 1970 (Figure 8 c, d). In this phase, the aquatic environment was severely deteriorated. High N and P retention in lakes and streams resulted in eutrophication, which threatened the ecology in the Rhine River basin and drinking water safety. Large amounts of nutrients exported to the coastal area led to harmful algae blooms (Prasuhn & Sieber, 2005; Wieriks & Schulte-Wülwer-Leidig, 1997). Substantial action to deal with water pollution was first started in the 1960s by the International Commission for the. Protection of the Rhine (ICPR), which was initially aimed at solving the chlorides issue (Mostert, 2009). However, the initiative contributed to the transboundary cooperation and successfully reduced the industrial emissions in 1970s. In our modelling results, we see a decrease of sewage N and P load in the same period. This led to the stabilization in the second phase of TN and TP delivery to the rivers.

In the 1980s, the Rhine Action Plan and North Sea Action Plan both aimed at reducing harmful substances to protect ecosystems and ensure drinking water safety (Mostert, 2009). At that time, N and P loads started to be specified in the pollution control strategies. The European Commission released several directives to reduce the nutrient loading to surface waters and to set standards for the nutrient concentrations within different water bodies. In the 1990s, the Nitrates Directive (CEC, 1991) intended to reduce the diffuse emissions from agriculture and the Urban Waste Water Treatment Directive (CED, 1991) aimed to improve the treatment of waste water. The Water Framework Directive (WFD) (EC, 2000) reported the ecological quality of surface water, which consists of several elements including biology factors, physic-chemical factors and specific pollutants. Major elements to describe the physic-chemical quality include the concentrations of N and P. The WFD set environmental quality standards for each nutrient specified for different types of surface water. With the enhancement of transboundary cooperation, countries had to take measures to meet the standards. Wastewater treatment system was improved to remove N and P from sewage wastewater. Best available techniques were provided to industrial activities to minimize the harmful effluents (Ackerman & Stewart, 1984). The modelling results reflected the effect of these measures: till 2000, sewage N and P nutrient loads have been lowered to 60 Gg yr^{-1} and 7 Gg yr^{-1}, respectively. Control of agricultural N and P inputs took longer than the wastewater reduction measures. Because it was difficult to change the agriculture mode to avoid soil surpluses of N and P fertilizers and manure. In terms of restrictions, limits on N and P application are set to different soil type and crop cultivation. The general limit for N from manure is 170 kg ha^{-1} yr^{-1} (Hirt et al., 2012). Each country set their own limits to manure and mineral fertilizers use both for N and P (Amery & Schoumans, 2014; Hirt et al., 2012). N and P content in soil are regularly monitored to meet the standards within farm land (Neeteson, 2000). Levies on exceeded N and P are important to help the implementation of the measures (Henkens & Van Keulen, 2001). The principle of polluter's pay gives pressure for farmers, thus some governments made market intervention to adjust the price of agricultural production as compensation (Lehn & Bahrs, 2018; Rougoor et al., 2001). Supporting measures include providing guidance to higher production efficiency and setting closed period for manure/fertilizers (Neeteson, 2000). Areas along surface water were obligated to be unfertilized, which function as riparian zones to reduce N and P leaching to surface water (Hirt et al., 2012). However, influenced by the social-economic status, the implementation progress varies in the countries. Conflicts between governments and farmers has been common (Eshuis & Stuiver, 2005). But the overall result is considerable, which can be seen in the modelling results. From 1980 to 2000, the TN soil budget decreased from 950 to 670 Gg yr^{-1}, followed by the same downwards trend for TN from groundwater in agricultural lands. The TP soil budget declined to below 0 Gg yr^{-1} in 1977 and reached -21 Gg yr^{-1} in 2000. However, the TN and TP from surface runoff in agricultural areas did not show a similar decrease. This phenomenon was particularly obvious for agricultural TP when its budget became negative (Figure 4 b) while P concentrations did not decline as rapidly. This delay of response of water quality can be explained by the N and P legacies in the soil and groundwater from the historical surpluses, which continue to leak nutrients to surface waters (Schoumans et al., 2015).

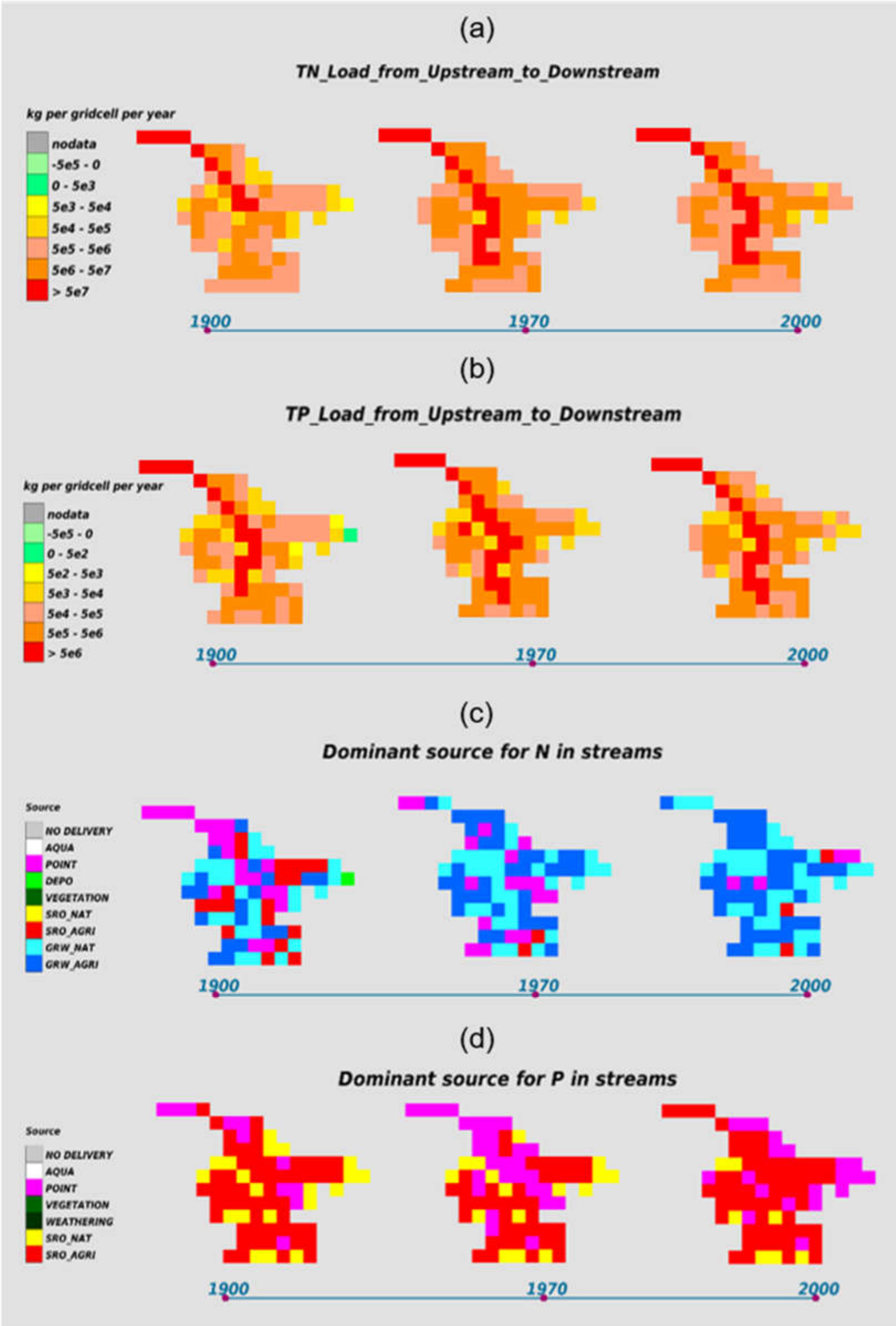

Figure 8. (a)TN load from upstream to downstream, (b)TP load from upstream to downstream, (c) Dominant source for N in streams; (d) Dominant source for P in streams for the Rhine River in 1900, 1970 and 2000

4.2 Comparison of the nutrient status of the rivers Pearl and Rhine

Before comparing the nutrient dynamics of the Rhine and Pearl, it is essential to know the basic differences between the two watersheds. Besides their basin area and annual discharge, the land use change also varies. During the 20^{th} century, the land use of the Rhine basin was almost unchanged. Only after 1970s, part of the agricultural land was restored to nature area (Figure 9 a). On the contrary, in the Pearl River basin, large part of nature area had been replaced by agricultural land till 2000 (Figure 9 b). There is limitation found in the land use data of Pearl River basin used in the IMAGE_GNM model, which is not well estimated. The proportion of grassland is overestimated than the actual land use of the Pearl River basin. This is because the basin land use is computed according to whole country's profile. Although adjustment algorithm is included to optimize the regional differences, the accuracy is not well ensured in the model. However, the expansion of crop land is well modeled, which is the most important change for the Pearl River basin in the 20^{th} century. According to the modelling results, the TN and TP delivery to the Pearl River had been increasing rapidly without deceleration till 2000. Thus, the nutrient dynamics of the Pearl River was in the accumulation phase between 1900 and 2000.

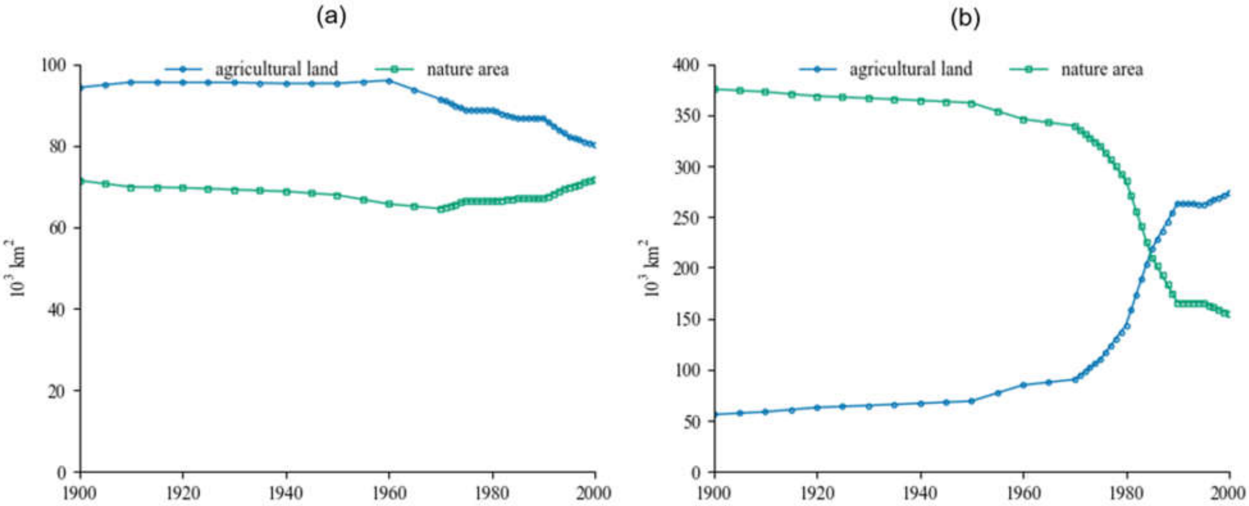

Figure 9. Area of nature agriculture land in the Rhine River basin (a) and the Pearl River basin (b) for the period 1900-2000

The most striking change in N and P dynamics in the Pearl river in the 20^{th} century was the rapid increase in the nutrient budgets, loads to rivers and export to the river mouth. Agricultural N and P loads initially rose from 1950, 5-fold and 2-fold respectively till 1975. In 1949, the P. R. China was established, followed by the expansion of agriculture and increase in manure use. In the 1970s, chemical the fertilizer industry developed quickly in China. This led to a sustained increase in synthetic fertilizer use (Gilmour & Kwiecinski, 2006). With the continuous expansion of agricultural land after 1975, we can see in Figure 6 (c, d), the TN and TP emission from agriculture was accelerated, and reached up to 730 Gg yr^{-1} of the TN and 87 Gg yr^{-1} of the TP loads at the river mouth. The share of agricultural sources reached at 86% for TN load and 82% for TP load in 2000 (Figure 6 a, b), becoming the most dominant nutrient source for both N and P in the Pearl river basin. The changes in nutrient loads and dominant sources are visualized in the basin maps shown in Figure 10. The nutrient dynamic results in the whole basin are similar to those in the west Pearl, because of its larger area than the area of east Pearl. The TP budget in the east part of the Pearl shows a fluctuating trend (Figure A3.1 (e, f)), which is different from the west part (Figure A2.1 a, b). This can be explained by the different land use change in the east Pearl basin. Agricultural land expansion stopped in this area since 1990, while it continued to expand in the west Pearl.

By comparing the modelled results of the Pearl River basin and the increasing phase of the Rhine River basin, N and P from agriculture and sewage wastewater are the most important nutrient sources in the two river basins. The difference is that agriculture expansion was much more intensive in the Pearl basin, while wastewater was more dominant for the Rhine basin. This is because of the different structure of major economic activities in the two river basins. For the Rhine, both agriculture intensification and industrial wastewater played an important role. For the Pearl, agricultural expansion and intensification was much earlier than the rise of industry in the 20^{th} century. The sewage wastewater contribution was much less in the Pearl basin than in the Rhine. But it is still an important anthropogenic T and P source, especially in the east Pearl and the delta area. After the Chinese economic reform in 1978 and the establishment of Pearl river delta economic circle in 1985, huge development and population growth took place in this area, resulting in massive wastewater emissions (Wong & Wong, 2004). At that time, the wastewater treatment facility was rather absent or very poor (Wen et al., 1999). Besides agriculture and sewage, aquaculture is another N and P source that cannot be ignored. The overall population growth and the large numbers of immigrants to the delta area motivated the aquaculture expansion, especially in the East River basin, the delta and costal area.

Our modelling results indicate that agriculture is the most dominant N source for the Pearl River basin in 1990s, which is consistent with Strokal et al. (2015). However, Strokal estimated that human waste was the dominant P source for the Pearl. The Global-NEWS-2 model calculated that Manure and fertilizers contribute to 40% of dissolved inorganic phosphorus export to coastal waters (Strokal et al. 2015). Other studies indicated agriculture and sewage are the most important nutrient sources, but without specifying their accurate contribution ratio (Dai et al., 2008; Huang et al., 2003; Wong & Wong, 2004). The uncertainty in defining the main P source could be caused by the model's weakness in reproducing the nutrient dynamic processes in the Global_NEWS_2 model and the spatial source attribution. Another possible cause is the different definition of point source in the model. In the IMAGE_GNM model, domestic wastewater in rural areas, which is typically not collected in sewer systems, is assumed to be used as manure fertilizer, while it is seen as point source in many other researches.

By analyzing the nutrient status of the accumulation phase of the Rhine (1900 - 1970) and Pearl (1900 – 2000), similarities and differences in spatial-temporal changes of nutrient load and dominant source can be seen in Figure 8 and Figure 9. During the accumulation phase of N and P loads, agriculture was more dominant for the Pearl River, while sewage and agriculture were at same level for Rhine River. As the N and P delivery to the river rose, the basin retention in the Pearl river kept increasing during 1950 and 2000. The retention fractions of the Rhine River and the Pearl river are relatively stable, despite a slight decreasing trend in the N retention ratio in Pearl. This is related to the algorithm of retention in the model, which defines a smaller retention ratio in higher TN concentration. The basin retention is allocated into different types of water bodies. Differently from the Rhine, there is no large lake in the Pearl basin. However, 32 reservoirs were built to serve for drinking water supply and to generate electric power, with a total volume reaching 40 km^3 in 2000. The proportion of TN and TP retention in reservoirs increased by 26% and 19% for the entire Pearl river basin (Figure 7 b, d). This ratio is even more dramatic in the east Pearl, which contributes up to 75% of TN and 66% of TP retention (Figure A2.2 f, h). The high retention could pose eutrophication risks within the reservoirs.

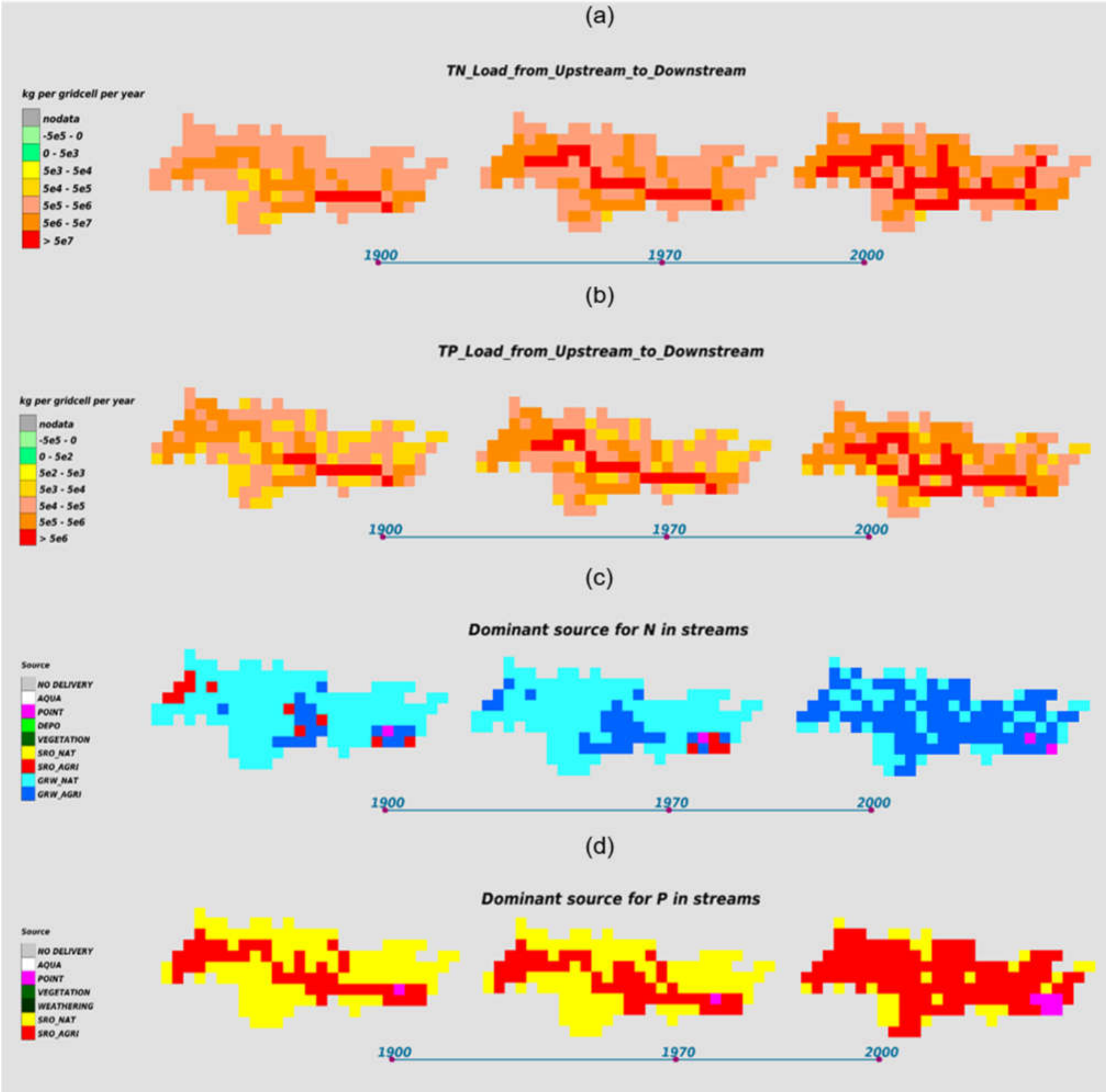

Figure 10. (a)TN load from upstream to downstream, (b)TP load from upstream to downstream, (c) Dominant source for N in streams; (d) Dominant source for P in streams for the Pearl River in 1900, 1970 and 2000

4.3 Lessons learnt from the Rhine and transfer to the Pearl

By analyzing the modelling results, the main N and P pathways and sources in the Pearl River basin are groundwater in agricultural and natural areas for N, and surface runoff for P. Sewage wastewater had been the dominant source in the delta area and expanded rapidly in the 1990s. Till 2000, sewage system was only constructed in the delta area. An increase in nutrient from sewage wastewater can be expected in the 21st century when sewage systems are built in the other parts of the river basin. To reduce the nutrient load from human activities, agricultural fertilizer and manure use and the wastewater treatment should be the most important concerns.

In terms of legislation, measures and regulations, the main nutrients management strategies of Rhine are summarized in Table 2. Whether similar measures have been taken in the Pearl River basin is checked and listed in the table. In the 20th century, there was no legislation about limiting fertilizer and manure use in the Pearl River basin. In recent years, however studies were carried out in China to solve the surplus problem of N and P in crop land. The actual regulatory measures started in 2015: the strategy named 'zero increase of fertilizer use' was aimed to stabilize the increase of fertilizer use (CMA, 2015). However, the average fertilizer applied to unit crop land of China is still 2.6 times of that in Europe (CMA, 2015). The production efficiency is much lower than that in Europe. Continuing fertilizer use reduction is still a challenge for the Pearl River basin. There are several realistic obstacles in the Chinese agricultural mode and the social background. In China, 69% of the population is in rural areas (China Statistics Year Book, 2004). Besides being engaged in agricultural production, some of them also work for local companies or in big cities. Farmers try to invest less time and still obtain high productivity. As the fertilizer prices are low in China, the small additional yield gain that can be obtained with an additional kg of fertilizer application is low, but yet profitable (Deininger et al., 2014). However, with this low efficiency of fertilizer use, surpluses of N and P in the soil are high. When the Rhine's nutrient budget peaked in 1970s, the situation for farmers was much better, because of the higher income and mechanization level. But controlling agricultural nutrient load was still the toughest challenge. N and P, the essential elements for agriculture production differ in their properties, and approaches for reduction are different. The legacies of P can remain in the soil for a long time and support plant growth, which makes it easier to cut down the P soil budget. But the legacy in soil can keep leaching to water bodies for decades. Differently, N is mobile in soil and can leach to groundwater aquifers. Aquifers have a travel time of years to decades, and the N applied long ago can now be discharged to surface waters. From the modelling results, we can distinguish the different trends of TN and TP budgets since the 1970s (Figure 4 a, b). However, the principle of regulating N and P are the same, which is by controlling fertilizer and manure use. Compared to the Rhine's measures, in the China's 'zero increase of fertilizer use' action plan, only the fertilizer application guidance has been implemented. Adapting more measures is needed to further reduce the N and P loads from agriculture.

Table 2. Comparison of the nutrient management strategies in the Rhine River basin with Pearl River basin.

	Strategies of Rhine	Pearl
Agriculture	Restrictions on N and P application to land	No*
	Guidance for optimizing fertilizer use	Yes* (since 2015)
	Monitoring the N and P balance in soil	No
	Closed periods for manure/fertilizers	No
	Unfertilized zones along surface water	No
	Levy to exceeded N and P use	No
Industry	Emission permits and corresponding standards	Yes (since 1996)
	Best available technique	Yes (since 2006)
Water quality Legislation	Setting critical values for substances in different kind of water bodies	Yes (since 2002)
Water quality monitoring	Complete monitoring system in water bodies	Yes
	Open data sources of water quality	No

*Yes means similar measures have been taken in the Pearl River basin

*No means no related measures have been taken

Wastewater from sewage is the second important anthropogenic nutrient source for the Pearl River basin, especially for the delta area. Sewage load has been increasing rapidly since 1980 and the wastewater treatment were almost absent in the 20th century (Wen et al., 1999). More than 70% of domestic wastewater was discharged into river or coastal waters directly in the Pearl River delta, and the industrial wastewater treatment ratio was even worse (Wen et al., 1999). However, since 2000, Chinese government has started to invest large amount of money to wastewater facilities. Domestic wastewater in the large cities were mostly treated, but the nutrients removal rates are still needed to be improved by technology upgrading (Sun et al., 2016). The problem remains in industrial wastewater treatment and waste flows in rural areas. Standards for wastewater emission had been set since 1996 (State Environmental Protection Administration of China, 1996), and permissions of effluent were given to industries only when they keep to meet the standards, which is similar to the emission permits policy in Rhine. But the awareness of environmental protection is lacking for some of the companies. Irresponsible industrial production continued between the regulatory gaps (Qu et al., 2016). This can be an obstacle for the Pearl River basin to regulate the sewage effluent. China has been initiating industrial transformation towards a more environmentally friendly system and higher production efficiency. Since 2006, China also adopted the Best Available Technique (BAT) to provide guidance to the industrial sectors (Wuana & Okieimen, 2011). In fact, this was a lesson learnt from the EU policies, which is aiming at balancing the economic costs and environmental benefits. In the Rhine basin, the sewage reduction was very successfully achieved in a shorter time than agriculture by improving wastewater treatment systems and optimizing production techniques. With similar approaches taken in the Pearl basin, we can expect a more sustainable wastewater treatment system in this century. In the Pearl River basin, the N and P discharge standards are set to be stricter to the area near lakes, reservoirs and other water sources that are sensitive to eutrophication (Petzoldt & Uhlmann, 2006). IMAGE_GNM model can give an explicit nutrient retention in different water bodies, which can be used to help define the sensitive area.

Besides agricultural reduction and sewage reduction measures, establishing an adequate monitoring system is also important. As the Rhine is a transboundary river, monitoring requires the agreement from the upstream countries to the downstream countries. The European Union played an important role to achieve the cooperation. ICPR and Water framework directive played important roles to enhance the cooperation and set the common standards. Complete water quality monitoring system was established in the Rhine basin, providing in time data of concentrations of substances. Annual basin reports are published on the water quality and ecological status. The Pearl river basin needs common standards and cooperation between provinces. However, the size of the Pearl basin makes monitoring the water quality in every part of the rivers a difficult task. Monitoring sites has been set up in the main streams of the Pearl River, but still not strongly developed in smaller streams. The density of monitoring sites is much lower than that of Rhine, and the monitoring data are not open to public. To ensure successful implementation of the environmental policies, besides the improved wastewater treatment and other regulations already in place, we suggest to install and improve the water quality monitoring system and parallel to this a better system of enforcement of the rules and regulations.

5. Conclusion

The IMAGE-GNM model is used to explore the N and P dynamics during 1900 – 2000. The Rhine's experience showed different effects of N and P reduction measures in agriculture and sewage wastewater. Improvement on wastewater facilities can cut down N and P load very quickly when most of the population are connected to the sewer system. But the N and P legacies in the soil and groundwater can keep leaching after agricultural nutrient inputs are reduced. These findings can help the policy-making in the Pearl River.

To analyze the applicability of the Rhine measures for the Pearl, it is certain that many other factors should also be considered, for instance their socio-economic status, governmental system and culture background etc. Here we can only give recommendations based on the potential nutrient reduction outcome when assuming that the measures were carried out in the Pearl basin. As the modelling results reveal that agriculture had been the dominant nutrient source for more than 90% of the basin area till 2000, regulating agricultural N and P loads should be the top priority. By comparing the current measures taken in the two river basins, it is found that the Pearl has been at the beginning of agricultural nutrients management. Therefore, more measures should be carried out in the future with the experience learnt from the Rhine. However, it can be expected that the effect of regulating fertilizer manure use to the N and P loads to river need time to will be visible due to the legacy effect. To support the nutrient management and related research. We recommend that the Pearl River to improve the monitoring system and make the water quality data more transparent to the public.

By applying the IMAGE_GNM model to the Rhine River and the Pearl River, this research provides the spatio-temporal changes of the N and P loads and the dominant sources within the basin, giving the focus of nutrient management strategies for each region. But limitations in the accuracy of the land use data exist in the model. The calculation of land types is now based on country-scale data. The model input of land use changes should be improved by using more detailed data in future research.

6. Acknowledgements

I really appreciate having this opportunity to research on bio-geochemistry modelling. I would like to express my gratitude to my daily supervisor Xiaochen Liu for guiding me to understand the IMAGE_GNM model. I want to thank Prof. Bouwman and Lauriane for helping me improve my thesis in both textual matter and content details.

7. Appendix

APPENDIX 1. Model description

The soil nutrient budget represents the nutrient inputs and outputs in agricultural and natural ecosystems. In agriculture, N inputs include fertilizer (Nfert), animal manure (Nman), atmospheric deposition (Ndep) and biological N fixation (Nfix). Ammonia volatilization (Nvol) and N withdrawal (Nwithdrawal) representing the vegetation consumed by animals or harvested by human are considered to be the outputs. For P, the inputs consist of fertilizer (Pfert) and manure (Pman). The P outputs of, P withdrawal (Pwithdrawal) refers to vegetation uptaken or harvested (Liu et al., 2018). The amount of N and P that can potentially enter the soil-hydrological system are computed as follows:

The soil nutrient budget represents the nutrient inputs and outputs in agricultural and natural ecosystems. In agriculture, N inputs include fertilizer (Nfert), animal manure (Nman), atmospheric deposition (Ndep) and biological N fixation (Nfix). Ammonia volatilization (Nvol) and N withdrawal (Nwithdrawal) representing the vegetation consumed by animals or harvested by human are considered to be the outputs. For P, the inputs consist of fertilizer (Pfert) and manure (Pman). The P outputs of, P withdrawal (Pwithdrawal) refers to vegetation uptaken or harvested(Liu et al., 2018). The amount of N and P that can potentially enter the soil-hydrological system are computed as follows:

$$Nbudget = Nfix + Ndep + Nfert + Nman - Nwithdrawal - Nvol \tag{1}$$

$$Pbudget = Pfert + Pman - Pwithdrawal \tag{2}$$

Nutrient entering groundwater system through leaching is a fraction of the soil N budget. The groundwater system in GNM consists of a shallow layer and deep layer, in which N residence time is accounted, so that the nutrient legacies can be simulated. Direct nutrient delivery through runoff is determined by land properties. Runoff through soils involves in the changes of nutrient contents in soil due to accumulation, depletion and erosion. Other direct nutrient delivery accounts for wastewater discharge, aquaculture, allochthonous organic material inputs, atmospheric deposition, and weathering of parent material (Liu et al., 2018).

The calculation of nutrient discharge from wastewater (from humans, animals and industries in urban areas) is described by Morée et al., (2013). Nutrient release from freshwater aquaculture is calculated using the country-scale model estimates of Bouwman et al (2013, Bouwman et al., 2011), for finfish and for shellfish using FAO (Organization, 2013) data for the period 1950 - 2010. Before 1950 aquaculture production was negligible for China.

The allocation of aquaculture nutrient release is related to population density, presence of surface water bodies, and mean annual air temperature (Beusen et al., 2015). Atmospheric N deposition is included in the soil budgets. Direct N deposition on water bodies can be calculated in each grid cell. Nutrient inputs from allochthonous organic material can be calculated as a fraction of the net primary production using N and P contents of the material depending on vegetation type. P release from weathering is associated with runoff, lithological class, the soil-shielding state and the impact of temperature (Goll et al., 2014)

For N, the main contributors to in-stream retention are denitrification, sedimentation, and uptake by aquatic plants. For P, the main contributors are sorption by sediment and sedimentation. Nutrient retention is calculated using the spiraling approach (Newbold et al., 2010; Wollheim et al., 2008). The hydraulic load and the net uptake velocity can be calculated separately for N and for P. The effect of concentration on denitrification as a result of electron donor limitation in the case of high N loads is dependent on temperature (Beusen et al., 2015). Nutrient retention is calculated using the spiraling approach (Newbold et al., 2010; Wollheim et al., 2008).

$$R = 1 - exp(\frac{-v_f}{H_L}) \quad (3)$$

H_L is the hydraulic load (m yr-1, eq. 4), and v_f is net uptake velocity (m yr-1), which is calculated separately for N and for P. The hydraulic load is calculated as follow:

$$H_L = \frac{D}{t} \quad (4)$$

where D is the waterbody depth (m), t is the residence time (yr) which is calculated by 102 volume V (m3) divided by discharge Q (m3 yr-1).

The net uptake velocity for P (eq. 5), and N (eq. 6) are calculated as follows:

$$v_{f,p} = 45f(t) \quad (5)$$

$$v_{f,p} = 35f(t)f(c_N) \quad (6)$$

where c_N is total N (TN) concentration in the waterbody considered, $f(c_N)$ describes the effect of concentration on denitrification as a result of electron donor limitation in the case of high N loads and *f(t)* describes the effect of temperature:

$$f(t) = \alpha^{(t-20)} \quad (7)$$

where α is 1.0717 for N (Wollheim et al., 2008) and 1.06 for P (Marcé & Armengol, 2009).

APPENDIX 2. Validation results per station

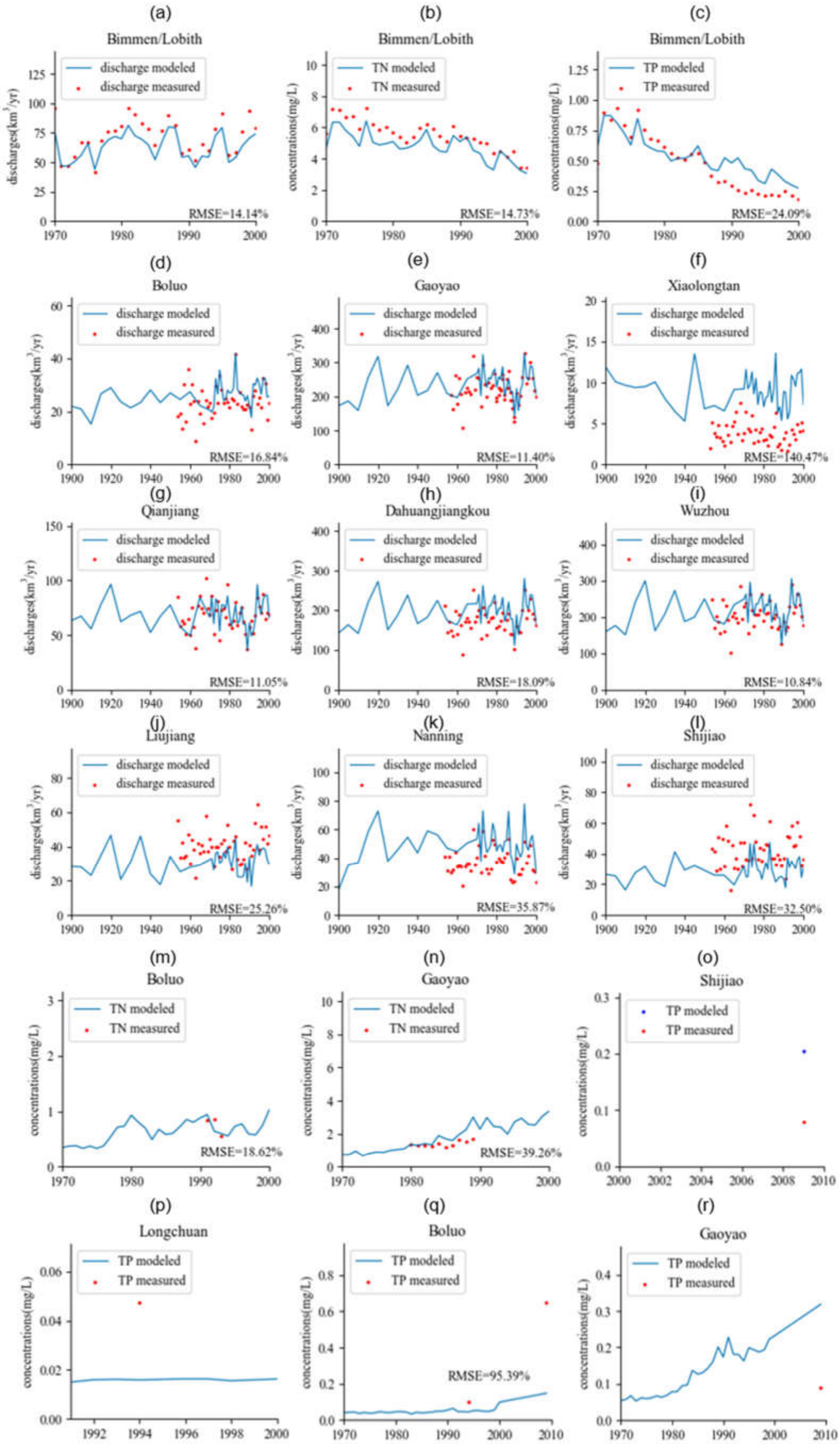

Figure A2. Comparison of measurements (red spots) and modeled (blue lines) discharge / N concentration / P concentration in the Rhine River(a-c) and Pearl River (d-r)

APPENDIX 3. Modelling results of West and East part of Pearl River

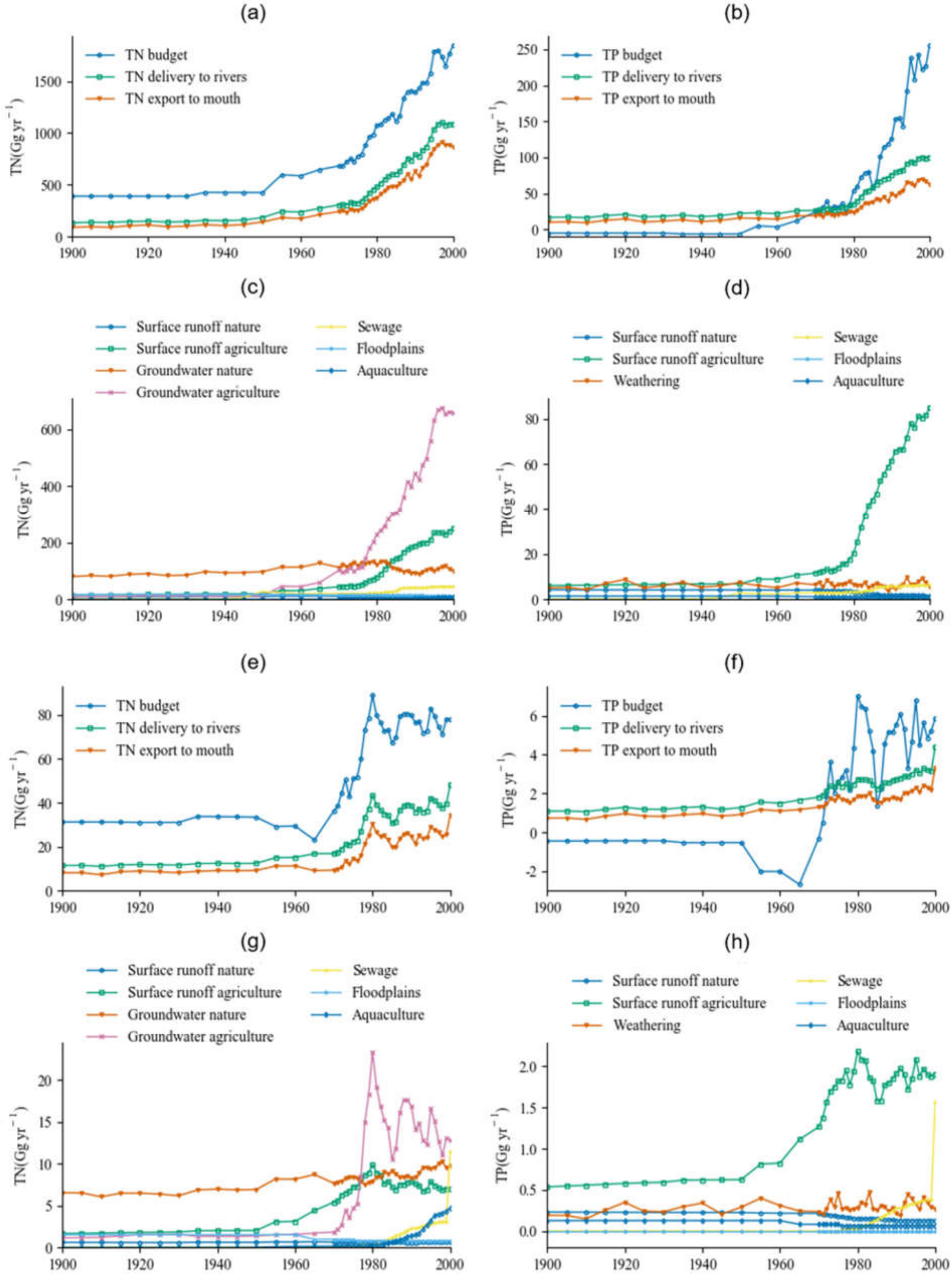

Figure A3.1. N and P soil budget, loads to rivers and exports to mouth for the west part (a, b) and east part (e, f) of Pearl River basin for the period 1900-2010; river TN and TP delivery to surface water from different sources for the west part (c, d) and east part (g, h) of Pearl River;

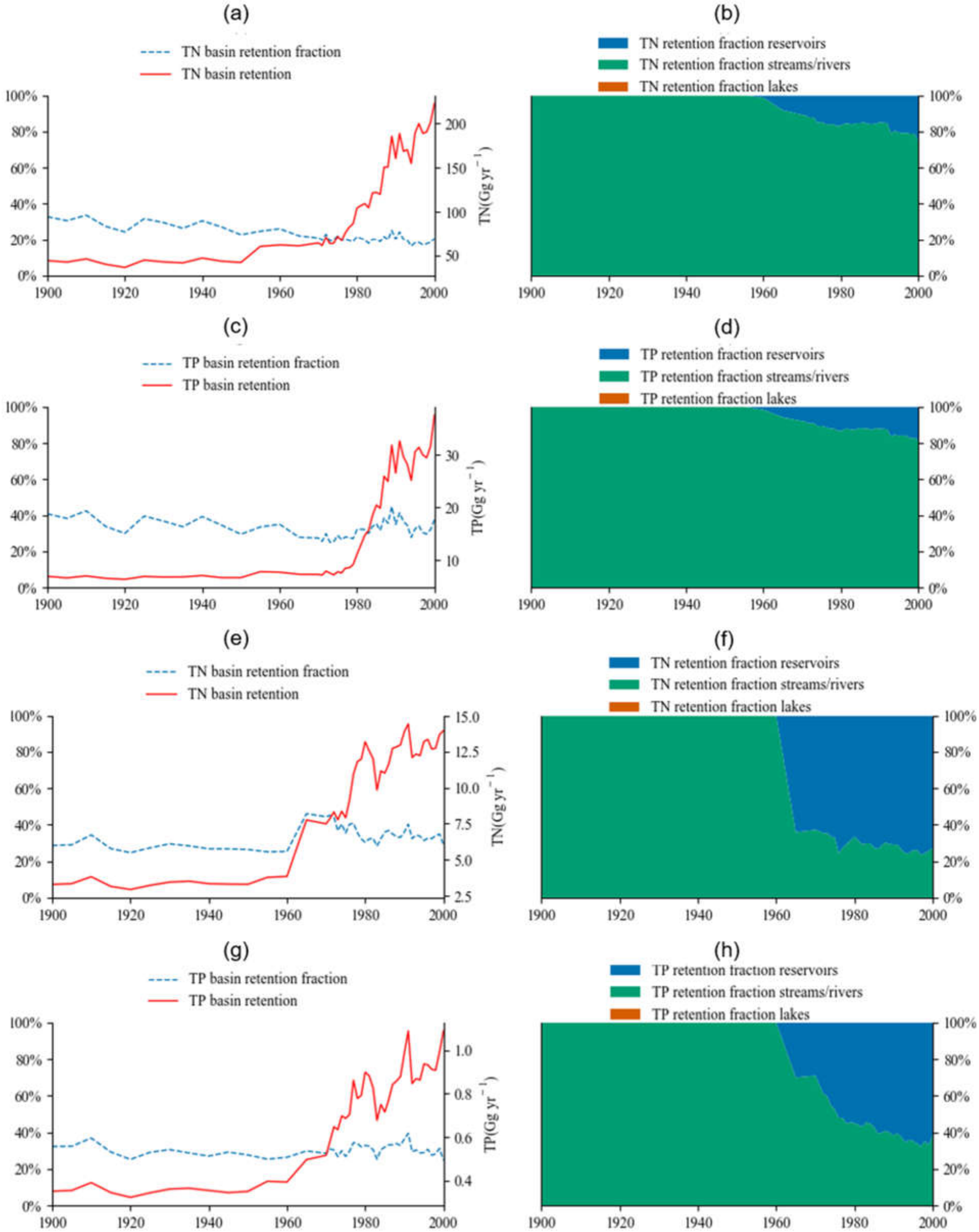

Figure A3.2. Total retention of N and the river basin N retention fraction, and fraction of retention in streams/rivers, lakes and reservoirs in the west part (a, b) and east part (e, f) of the Pearl River basin; total retention of P and the river basin P retention fraction, and fraction of retention in streams/rivers, lakes and reservoirs in the west part (c, d) and east part (g, h) of the Pearl River basin

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