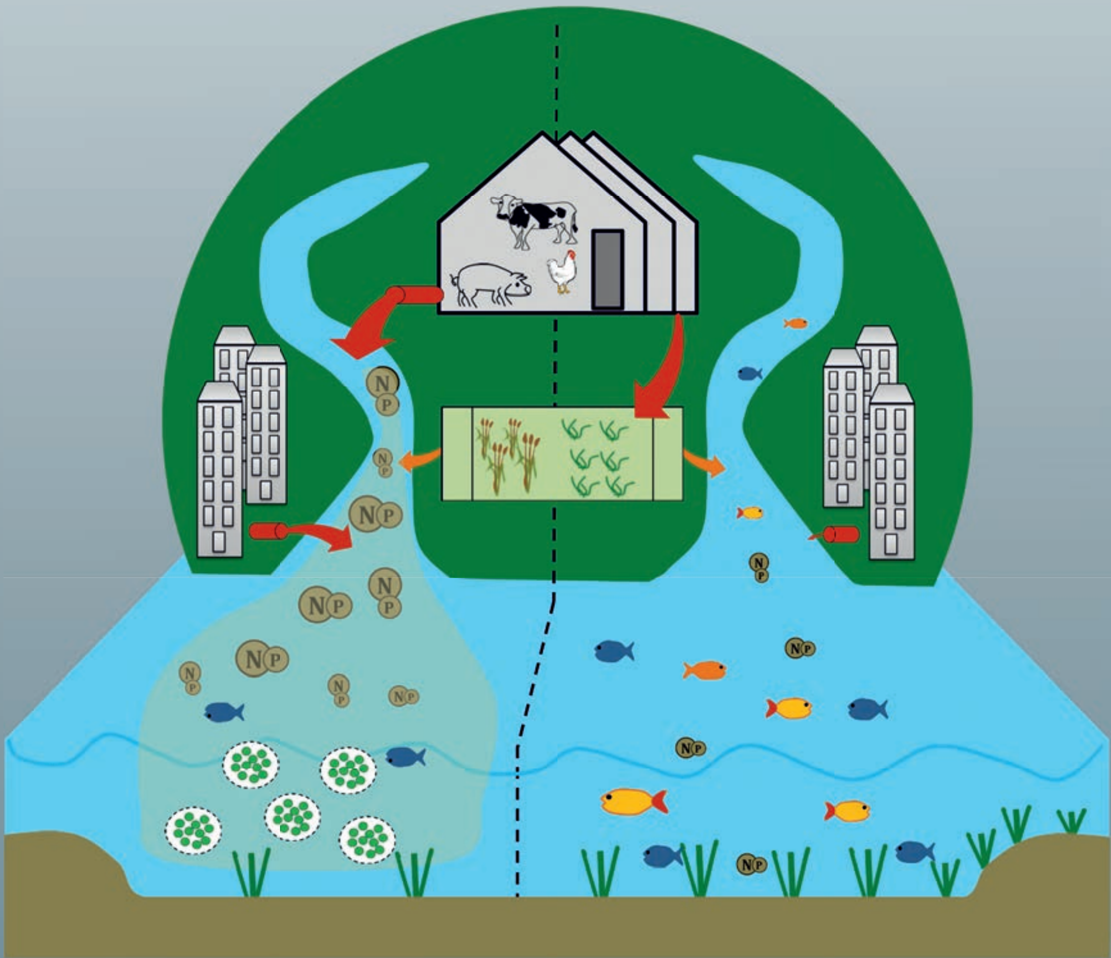
River export of nutrients to the coastal waters of China:

The *MARINA* model to assess sources, effects and solutions



Maryna Strokal

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Thesis committee

Promotor
Prof. Dr C. Kroeze
Professor of Water Systems and Global Change
Wageningen University & Research

Co-promotors
Prof. Dr S. Luan
Professor at Shenzhen Graduate School
Peking University, China

Prof. Dr L. Ma
Professor at the Center for Agricultural Resources Research, Institute of Genetic and Developmental Biology
Chinese Academy of Sciences, China

Other members:
Prof. Dr A.A. Koelmans, Wageningen University & Research
Prof. Dr W. Ma, Agricultural University of Hebei, China
Dr M. L. McCrackin, the Baltic Sea Centre, Stockholm University, Sweden
Prof. Dr H.H.M. Rijnaarts, Wageningen University & Research

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Maryna Strokal

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Table of contents

Chapters 2-5 have been published as peer reviewed scientific articles. Chapter 6 has been submitted and is under revision after positive reviews. The text, figures and tables of the published and submitted articles have been adjusted to the PhD thesis format. Throughout the thesis English names are used for Chinese rivers. Editorial changes were made for reasons of uniformity of presentation in this thesis. References should be made to the original articles.

Chapter 1.

General introduction

1

"Clean, accessible water for all is an essential part of the world we want to live in."
Sustainable Development Goals, 2015

1.1 Motivation

Society needs ample clean water for, among others, drinking, food production, recreation and energy production. Clean water is also a prerequisite for well-functioning ecosystems. However, clean water is not always adequately available everywhere and this may become worse in the future (Vörösmarty et al. 2010). The availability of clean water is further threatened by pollution as many rivers nowadays are polluted by excess nutrients (Diaz & Rosenberg 2008; Sutton et al. 2011). River export increases the nutrient level in coastal waters (Seitzinger et al. 2010) and this causes frequent eutrophication problems like blooms of harmful algae. Such environmental problems threaten not only ecosystems by killing fish through anoxic conditions but also affect people's health via toxic algae (Diaz & Rosenberg 2008). This likely brings economic losses to sectors like tourism and fishery.

China has rapidly developed economically over the past few decades (Liu & Diamond 2005; Ma et al. 2013b). This not only increased Chinese fresh water use (Piao et al. 2010), but also heavily polluted Chinese main rivers (Xu et al. 2014) and thus eutrophication is abundant in Chinese coastal waters (Tang et al. 2006a). In my PhD research, I therefore focus on understanding the trends in nutrient export by Chinese rivers, causes of river pollution and assess solutions to reduce the consequent eutrophication problems in Chinese coastal areas. I develop and apply an integrated model to simulate all relevant processes in Chinese rivers. Outcomes of my research will hopefully contribute to an increase in the future availability of clean water for the current and next generations in China. This is also essential in meeting the recently accepted UN Sustainable Development Goals (Griggs et al. 2013). In the following sections I introduce in detail the research problem, its knowledge gaps and approaches to fill these gaps. This chapter will define the thesis' objectives that are elaborated in the subsequent chapters.

1.2 Background

1.2.1 Nutrient cycling

The cycling of nitrogen (N) and phosphorus (P) involves processes that influence export of these nutrients by rivers. These processes transform N and P from organic forms to inorganic and vice versa. N and P in rivers are in dissolved inorganic (DIN, DIP), dissolved organic (DON, DOP) and particulate N and P (PN, PP) forms. Dissolved forms are more bioavailable than other forms (Garnier et al. 2010).

N cycling is controlled by various processes such as N fixation, mineralization, ammonification, nitrification, and denitrification (Bouwman et al. 2009). Most of these processes are bacterial, taking place under aerobic or anaerobic conditions. Only certain bacteria can convert inert N from the atmosphere into ammonium and organic forms of N. This way, they make nitrogen available for other organisms in soils and water (Galloway et al. 2008; Smil 2004). In nitrification, ammonia is converted to nitrate, which is another bioavailable element (Bouwman et al. 2009). During N cycling some N is lost to the atmosphere via denitrification or to water via leaching. Anaerobic bacteria denitrify biologically available N back to unreactive N. When this denitrification process is incomplete, other gaseous N compounds, like nitrous oxides (NOx) can also be released.

P cycling is controlled by weathering, mineralization and adsorption or desorption (Bouwman et al. 2009; Schoumans 2015). Weathering of rocks releases inorganic P to soils and water. Living organisms absorb this inorganic P and return organic matter when they die and decay. Certain bacteria mineralize organic matter back to bioavailable P forms. P has a strong ability to accumulate in soils and sediments because of strong chemical adsorption/desorption processes. These processes are largely controlled by the availability of calcium, iron and aluminum oxides. These chemicals bind P (adsorption) and form insoluble compounds. Desorption is the process of releasing bound P to soil solutions (Schoumans 2015). Thus, the adsorption/desorption processes influence P release to aquatic systems (Bouwman et al. 2009; Schoumans 2015).

Anthropogenic activities can influence N and P cycles in two main ways. The first way is adding nutrients as part of fertilizers that ends up in soils and rivers (Galloway et al. 2008; Sutton et al. 2013; Vitousek et al. 2009). The other is related to human impacts on rivers, including water withdrawal and dam construction (Maavara et al. 2015; Van Cappellen & Maavara 2016). Depending on the degree of these human influences nutrient export by rivers can either increase or decrease.

1.2.2 Trends in nutrient export by rivers

China has many rivers. Sixteen rivers flow through around 40% of the country to the Bohai Gulf, Yellow Sea and South China Sea (Qu & Kroeze 2010). They flow through areas with intensive socio-economic developments (see Section 1.2.3). The Yangtze (Changjiang), Yellow (Huanghe) and Pearl (Zhujiang) rivers have the largest drainage areas (see Section 1.5 on the study area).

Several studies indicate increasing trends in nutrient export by Chinese rivers since the 1970s. For example, Qu and Kroeze (2010) analyzed nutrient export by sixteen rivers for N and P in different forms. Results show that river export of dissolved N and P increased by a factor of 1.3 to 4.0 between 1970 and 2000. River export of particulate N and P changed slightly whereas dissolved silica (Si) somewhat decreased during this period. Other studies are mostly for large individual rivers and nutrients. Examples include studies for the Yellow (Tao et al. 2010; Tong et al. 2016), Yangtze (Li et al. 2014b; Li et al. 2011b; Li et al. 2011c; Powers et al. 2016; Sun et al. 2013a; Sun et al. 2013b; Wang et al. 2015; Yan et al. 2010; Yu et al. 2011) and Pearl (Huang et al. 2003; Yin et al. 2004). In general, these studies confirm an increase in fluxes of dissolved nutrients by rivers since the 1970s, except for Powers et al. (2016) who showed a declining trend in P export by the Yangtze since 1970. Some experimental studies also report on nutrient pollution problems for inland water systems (Huang 2015; Xu et al. 2014) and estuaries (Liu et al. 2009b; Tong et al. 2015). For example, Xu et al. (2014) indicate that many Chinese rivers like Huai, Hai, Yellow and river deltas today have concentrations of nutrients at levels that do not allow for human consumption. However, these studies lack analyses of trends in nutrient export by Chinese rivers.

Clearly the abovementioned studies differ in their focus, study area and nutrient forms. Thus, a comprehensive analysis of trends in the export of different nutrient forms by Chinese rivers with comparable data and methodologies is still lacking.

1.2.3 Causes of nutrient pollution in rivers

Agriculture and urbanization are important causes of increasing nutrient export by Chinese rivers (Ju et al. 2005; Liu & Diamond 2005; Ma et al. 2013b). The rapid economic development and fast growing population have intensified agriculture (Zhang et al. 2013) and stimulated urbanization (Liu & Diamond 2005; Qu & Kroeze 2010). For example, the annual income per capita increased by around seven times and the total population by half, between 1970 and 2000 (Qu & Kroeze 2010). Today China accommodates around 1.3 billion people, which is one-fifth of the world's population (Ma et al. 2010). Nearly half of the total Chinese population is urban (WHO/UNICEF

2014). Figure 1.1 shows pathways of how important human activities contribute nutrients to Chines rivers and thus to coastal waters.

Urban waste is an important source of river pollution (Figure 1.1). Although urbanization develops fast in China (Liu & Diamond 2005), not all urban people are connected to sewage systems (WHO/UNICEF 2014). In 2000, only one-third of the total wastewater was treated in China. This increased up to almost half in 2005 (Liu & Qiu 2007). By 2012 around three thousands wastewater treatment plants (WWTPs) were available in China for municipal wastewater (Jin et al. 2014). However, their treatment efficiency is not always effective in removing N and P (Jin et al. 2014). Furthermore, treatment of sludge and its management need improvements (Jin et al. 2014). Uncollected urban waste can end up either in nearby water systems or used as a fertilizer for fruits and vegetables in neighboring semi-urban areas (Figure 1.1). This also happens in rural areas where people lack access to sewage systems (Ju et al. 2005; Ma et al. 2012; Miao et al. 2010; WHO/UNICEF 2014; Xing & Yan 1999). Thus, human waste from urban and rural areas can be diffuse as well as point sources of nutrients in rivers (see above, Figure 1.1).

Agriculture is a source of pollution because of large losses of nutrients to the environment. The North China Plain (NCP) is a typical example of areas with intensive crop production, contributing to around half of the national wheat production (Kendy et al. 2003). However, in China arable farmers prefer to use synthetic fertilizers in considerable amounts instead of animal manure. This has resulted in over-fertilization of soils and has adverse effects on crop yields and water quality (e.g., Chen et al. 2014; Ju et al. 2009). Thus the efficiency of N and P use in crop production is relatively low in China compared to the world average (Ma et al. 2010). Crop production is considered as a diffuse source of nutrients in rivers. This is because nutrients enter rivers from fertilized soils via leaching and runoff (Figure 1.1).

Animal production has started industrializing since the 1990s for food security reasons (Bai et al. 2014; Chadwick et al. 2015). The number of pigs, poultry, cattle, sheep and goats increased by a factor of 1.5–8 between 1970 and 2000. And in 2000 around 10 - 40% of the animals were housed in industrial farms. This increased to 40-80% for pigs and poultry in 2010 (Bai et al. 2013; Bai et al. 2014; MOA 2011b). Manure management in industrial farms differ from that in small, traditional farms (Chadwick et al. 2015). Traditional farms were more dominant in the past. Those farms collected manure and usually recycled it for crops, vegetables and fruits. Today, there are still many small farms growing animals, but their meat products are mainly for local consumption (Chadwick et al. 2015). Industrial farms today are largely disconnected from crop production. Management of manure in industrial farms is such that considerable amounts of manure is directly discharged to nearby water systems (Chadwick et al. 2015). Some manure is collected and can be composted, however, it is rarely brought back onto the land. This is a result of constrains like transportation costs, outdated application techniques, labor demand and limited knowledge (Chadwick et al. 2015; Ma et al. 2013b). Thus, animal production in China can be diffuse and a point source of nutrient pollution (Bai et al. 2015; Bai et al. 2013; Bai et al. 2014; Chadwick et al. 2015; Ma et al. 2012). A diffuse source is when manure is used as fertilizer. A point source is when manure is discharged to water (Figure 1.1).

The abovementioned studies largely emphasize the importance of diffuse sources of N pollution in Chinese rivers because of soil over-fertilization. Point sources are considered large polluters of P in rivers as a result of inadequate treatment (e.g., Bao et al. 2006; Liu et al. 2008a; Qu & Kroeze 2010; Qu & Kroeze 2012; Shen et al. 2012; Xing & Zhu 2002). Some studies indicate the importance of atmospheric N deposition on land (Liu et al. 2013) and on seas (Xu et al. 2015) as well as of aquaculture (Biao & Kaijin 2007; Cao et al. 2007; Liu et al. 2009a). Direct N deposition on seas and aquaculture are point sources of nutrients in local areas of China. Human waste that is not collected and industrialization of animal production take, however, place across China (Chadwick et al. 2015; Ma et al. 2012).

However, the importance of uncollected human waste and manure discharges from animal production in the total amount of river pollution is less well studied. Uncollected human waste can be an important source of nutrients in Chinese rivers since many rural people are still without proper sanitation facilities (WHO/UNICEF 2014). Chadwick et al. (2015) indicate that approximately half of the manure from industrial farms is directly discharged to water systems. This makes animal production a potentially large polluter of Chinese rivers. This is in line with some other national (Ma et al. 2010) and provincial (Ma et al. 2012) studies on nutrient flows in the food chain of China. Unfortunately, river-specific estimates do not exist. Studies on river export of nutrients often do not account for such sources (e.g., Li et al. 2011b; Qu & Kroeze 2010; Yan et al. 2010). As a result, existing studies may underestimate nutrient pollution of rivers and coastal waters in China.

Furthermore, how human activities in upstream parts of a river affect water quality downstream is not studied well, but it is important for the management of coastal eutrophication. This is especially needed for large rivers like the Yangtze, Pearl and Yellow (see Section 1.5) as their contribution to coastal water pollution might be considerable. A few studies divided the large drainage area into sub-basins for better spatially explicit analyses (Bao et al. 2006; Chen et al. 2016; Liu et al. 2008a). But these studies are mainly for the Yangtze and focus more on nutrient budgets of agricultural systems. Sub-basin analyses are more common for hydrological studies in China (Cui et al. 2007; Huang et al. 2009a; Niu & Chen 2010; Wang et al. 2010; Yang & Lu 2014b; Zhou et al. 2013) while scarce for nutrient export analyses.

In addition to human activities, water withdrawal and dams affect export of nutrients from upstream to downstream (Figure 1.1). An example is the Three Gorges Dam, which is located in the middlestream of the Yangtze. It is one of the largest dams in the world. Its reservoir retains nutrients and thus reduces nutrient export downstream of the Yangtze (Sun et al. 2013a; Sun et al. 2013b). Results of Qu and Kroeze (2010) also indicate effects of dams especially on river export of particulate N and P, but their analyses are at the basin scale. Sub-basin analyses of the Yellow exist for the effects of water consumption on nutrient fluxes (Tong et al. 2016). The drainage area of the Yellow is drier than of the Yangtze and Pearl (Piao et al. 2010; Zhai et al. 2005). And part of the Yellow drainage area is located in the NCP where agriculture is intensive (see above). Water demand for human needs is thus high (Li et al. 2008). Tong et al. (2016) show the considerable effect of high water consumption on removing nutrients from the river. Piao et al. (2010) indicate an increase in water withdrawal from Chinese rivers. Clearly, the amount of nutrients reaching coastal waters of China is the combined effect of human activities with retentions and removal of nutrients in rivers.

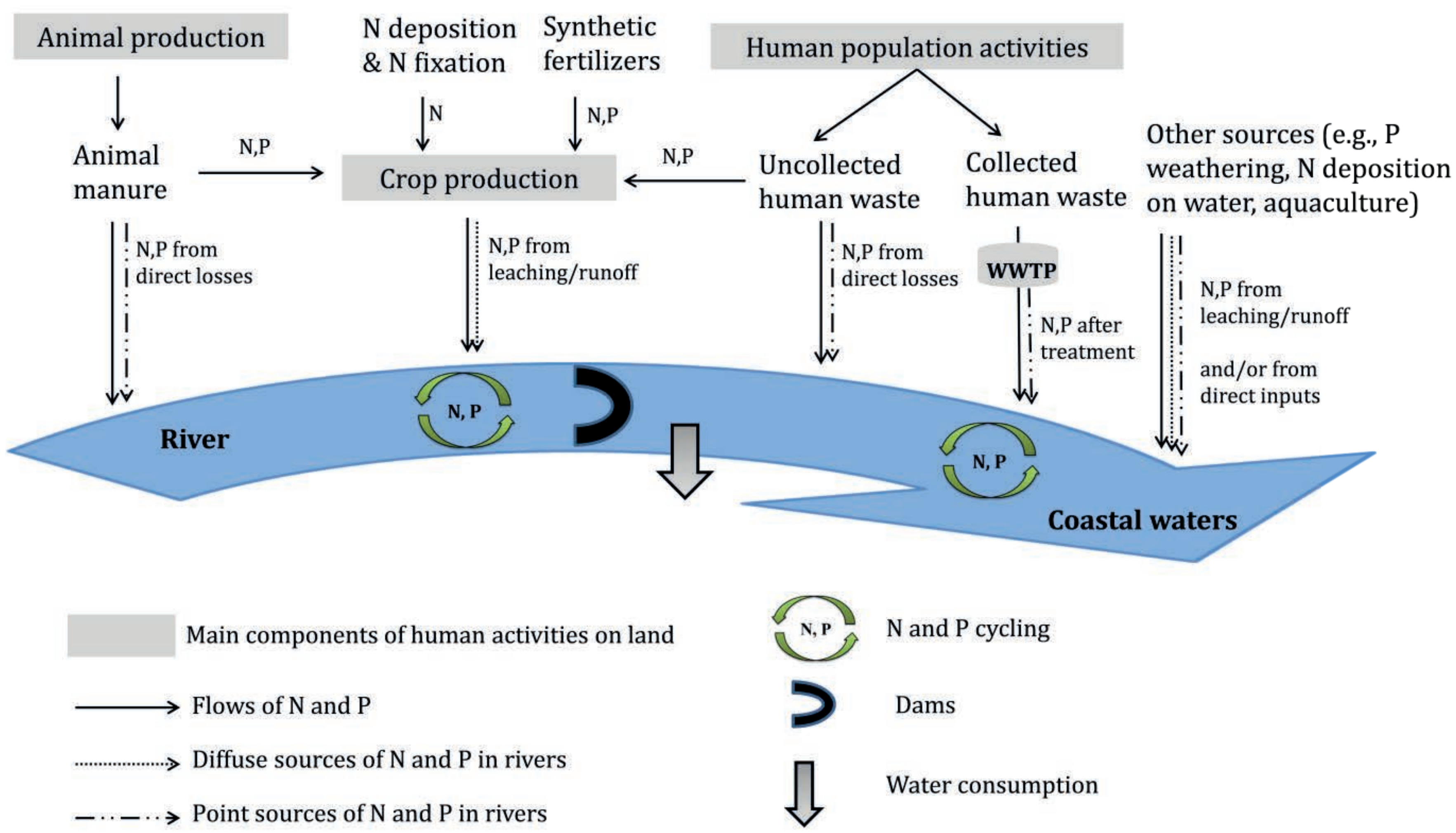

Figure 1.1. *Main pathways of nitrogen (N) and phosphorus (P) flows to rivers from different sources. WWTPs are wastewater treatment plants. Other sources of nutrients in rivers also cover non-agricultural land. This overview is made based on various existing studies (see Sections 1.2.2 and 1.2.3).*

1.2.4 Impacts on coastal waters

Increasing river export of nutrients can potentially result in impacts such as coastal eutrophication and its symptoms. Eutrophication refers to the water enrichment by N and P. Blooms of harmful algae can develop in such eutrophic systems. Algae or microscopic phytoplankton sustain primary production in aquatic systems. Siliceous diatoms are examples of a large group of primary producers. The Redfield ratio of N:P:Si (16:1:20) characterizes the nutrient requirements for diatom growth in marine systems (Garnier et al. 2010). However, when rivers deliver too much N and P, then the ratio of N:P:Si is unbalanced. A shortage of Si occurs with increasing loads of N and P. In such conditions diatom growth is limited and new non-siliceous algae can develop. These are often undesirable algae. They can bloom and thus deplete oxygen (Billen & Garnier 2007). This often results in hypoxic conditions and leads to the deaths of living organisms such as fish (Diaz & Rosenberg 2008). Some non-siliceous algae like cyanobacteria have toxins and thus can pose a threat for people (Berdalet et al. 2016). Other factors such as light and the presence of algae consumers also play a role in controlling algal growth together with nutrients (Conley et al. 2009; Davis et al. 2009; Paerl et al. 2001).

In China the number of recorded events with algal blooms increased from less than five before the 1970s to more than a hundred in the 2000s (Tang et al. 2006a). This affected around 14 thousands km^2 of the Chinese coastal area in 2003. In particular, coastal eutrophication and blooms of harmful algae were reported for the Yellow Sea (Tang et al. 2006a), South China Sea (Wang et al. 2008) and the Bohai Gulf (Tang et al. 2006b). The serious effects of these environmental problems like fish kills and poisoned people by toxic algae have already been indicated (Tang et al. 2006a).

Some studies relate river export of nutrients to eutrophication problems in China, but mainly for individual rivers and nutrient forms. For example, Li et al. (2014b) related decreasing trends in Si and increasing trends in DIN export by the Yangtze river with the development of harmful algae. Huang et al. (2003) and Yin et al. (2004) link the P limited Pearl River with high concentrations of chlorophyll a in the estuary of the river. Liu et al. (2009b) report on decreasing ratios of Si to DIN and DIP in estuaries of the three main seas of China. Liu et al. (2016) analyzed P losses to water and related these losses to freshwater eutrophication.

Clearly, a comprehensive analysis that relates river export of nutrients to eutrophication of the Chinese seas in China is still lacking. Studies for other regions (e.g., Dupas et al. 2015; Kroeze et al. 2013; Sattar et al. 2014) do this by using the Indicator for Coastal Eutrophication Potential (ICEP) (Garnier et al. 2010). It relates riverine fluxes of total N and P to fluxes of Si to determine the potential for coastal eutrophication. So far, application of ICEP in China has been limited to a few individual rivers (Liu et al. 2012; Liu et al. 2011; Wang et al. 2013a).

1.2.5 Scenarios for nutrient exports and coastal eutrophication

Urbanization and industrialization of animal production will likely continue in the coming years in China. More people are expected to move to cities for better jobs (Qu & Kroeze 2012). By 2050 around two-thirds of the total Chinese population is expected to be urban (Qu & Kroeze 2010). This will increase the demand for food in urban areas. Thus, more industrial farms are expected to emerge around cities to produce enough meat products. As a result, the impact of urbanization and industrialization on water quality in China may increase unless human and animal waste is properly managed.

Scenario analyses can help to test the effectiveness of options to reduce adverse effects of human activities on future water pollution. This is done with the help of modeling tools (see Section 1.2.6). Currently, several sets of global scenarios exist. For instance, there are scenarios from the Spatial Report Emissions Scenarios (SRES, Nakicenovic et al. 2000), Millennium Ecosystem Assessment (MEA, Alcamo et al. 2005), Shared Socioeconomic Pathways (SSPs, O'Neill et al. 2014) and Representative Concentration Pathways (RCPs, Van Vuuren et al. 2011). These scenarios can serve as a basis to account for socio-economic drivers of population development and for changes in climate that influences hydrology and thus river export. MEA scenarios have been widely used in the river export of nutrients for many world regions (Sattar et al. 2014; Strokal et al. 2014c; Suwarno et al. 2013; Yasin et al. 2010; Zinia & Kroeze 2015) including China (Li et al. 2011b; Qu & Kroeze 2010; Qu & Kroeze 2012; Yan et al. 2010). SSPs and RCPs are relatively new scenarios and thus their applications to nutrient export by Chinese rivers are, so far, rare.

MEA includes four scenarios for 2030 and 2050. These are the Global Orchestration (GO), Adapting Mosaic (AM), Order from Strength (OS) and TechnoGarden (TG) (Alcamo et al. 2005). The main differences among these scenarios are associated with globalized (GO, TG) or regionalized (AM, OS) trends in socio-economic development and with a reactive (GO, OS) or proactive (AM, TG) approach to manage environmental problems (Alcamo et al. 2005; Seitzinger et al. 2010). These scenarios were quantitatively interpreted and integrated into Global *NEWS*-2, a river export model, short for Nutrient Export from WaterSheds (Mayorga et al. 2010; Seitzinger et al. 2010) (see Section 1.2.6 for a model description).

MEA scenarios can be used as the basis to develop alternative scenarios to test the effectiveness of additional options to reduce future water pollution up to 2050. This was done for Chinese rivers in a few studies. For example, Qu and Kroeze (2012) analyzed effects of nutrient management options in agriculture and sewage. These management options focused mainly on reducing over-fertilization of arable soils and on increasing the removal efficiency of nutrients during treatment. Reducing soil over-fertilization is more effective to reduce N pollution and increasing nutrient removal is more effective to reduce P pollution in Chinese rivers. However, direct inputs of animal manure to rivers from industrial farms and uncollected human waste are not accounted for in this study as well as in other similar river export studies (Li et al. 2011b; Yan et al. 2010). Thus, the effectiveness of the tested management options on reducing river pollution might be overestimated. Furthermore, these studies did not analyze the potential for future coastal eutrophication as affected by river export of nutrients.

Scenario studies that account for both direct manure discharges and uncollected human waste do not quantify river export of nutrients. They focus on nutrient use efficiencies in crop and animal production at the national (Bai et al. 2015; Ma et al. 2013b), provincial (Ma et al. 2012) and county (Wang et al. under review; Wang et al. in preparation) scales. They take FAO (Food and Agriculture Organization) projections for future developments in agriculture up to 2030. They often assume business-as-usual scenarios and compare them with scenarios assuming improvements in nutrient management. These improvements are largely motivated by emerging environmental policies (e.g., MOA 2015) and by experimental studies to increase crop yields (Chen et al. 2011; Chen et al. 2014; Ju et al. 2009; Miao et al. 2010). These scenario studies conclude the following: integrated nutrient management that combines management options in crop and animal production is more effective than individual options in reducing nutrient losses from agriculture to the environmental.

Scenario analyses that encompass the whole range from worst to best cases contribute most to a better understanding of water pollution (Figure 1.2). However, existing studies tend to be somewhere in between best and worst cases and typically focus on the effects of selected individual or combined management options (e.g., Bai et al. 2015; Ma et al. 2013a; Qu & Kroeze 2010; Qu & Kroeze 2012). Worst-case scenarios typically take a pessimistic approach and represent a worst case for the environment (high pollution, Figure 1.2). Examples can be scenarios with lack of effective environmental policies for China such as GO in the MEA (Alcamo et al. 2005; Seitzinger et al. 2010). In contrast, the best-case scenarios typically take an optimistic approach and represent a best case for the environment (low pollution, Figure 1.2). Examples can be scenarios that assume full implementation of the best technologies to reduce water pollution. These optimistic scenarios are currently lacking for river export of nutrients to coastal waters of China.

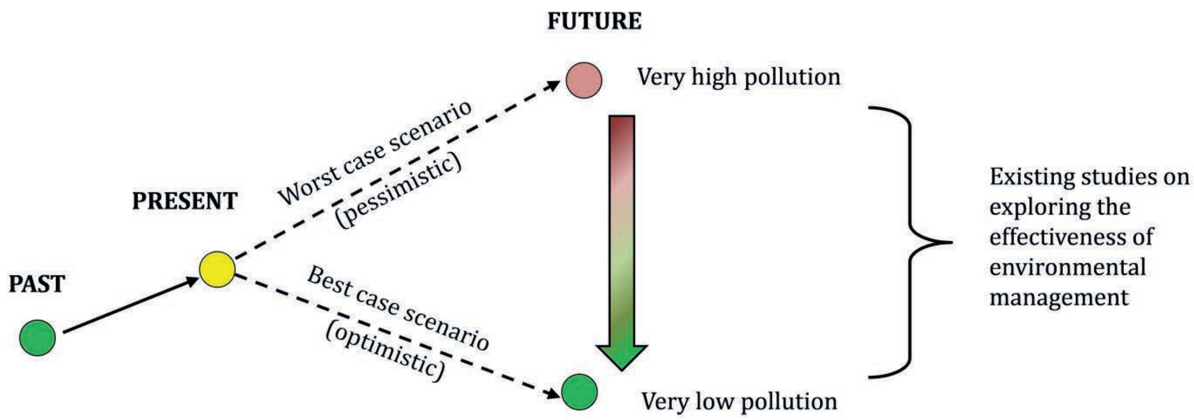

Figure 1.2. *Schematic illustration of two extreme perspectives in scenario analyses to explore futures for water pollution by nutrients: worst and best cases scenarios. The worst case takes a pessimistic perspective and the best case takes an optimistic perspective.*

1.2.6 Models for nutrient exports and coastal eutrophication

Models can be used to better understand causes of high river export of nutrients to coastal waters of China. Models are used to explore solutions based on scenarios. In fact, various nutrient-related models exist today (e.g., Beusen et al. 2015a; Ma et al. 2010; Mayorga et al. 2010; Strokal & de Vries 2012). Here I present briefly illustrative examples of models for nutrient fluxes. These are IMAGE-GNM (Integrated Model to Assess the Global Environment–Global Nutrient Model, Beusen et al. (2015a)), Global *NEWS*-2 (Mayorga et al. 2010), SPARROW (SPAtially Referenced Regressions On Watersheds, Schwarz et al. 2006), RVERSTRAHLER (Sferratore et al. 2005), SWAT (Soil and Water Assessment Tool, Arnold et al. 2012), NUFER (NUtrient flows in Food chains, Environment and Resources use, Ma et al. 2012) and AGNPS (AGricultural Non-Point Source Pollution, Jianchang et al. 2008).

These models are similar in some aspects, and different in others. These aspects are, for example, the aim, spatial and temporal level of detail as well as nutrient sources. Most of the abovementioned models (except NUFER) quantify the river export of nutrients while accounting for human activities on land and nutrient retentions during traveling from land to river outlet. NUFER quantifies nutrient flows in the food chain (Ma et al. 2012). The temporal level of detail is annual for Global *NEWS*-2 (Mayorga et al. 2010), IMAGE-GNM (Beusen et al. 2015a) and SPARROW (McCrackin et al. 2013). RVERSTRAHLER (Ruelland et al. 2007) and SWAT (Arnold et al. 2012) allow for inter-annual analyses of nutrient fluxes. AGNPS focuses on events such as storms (Jianchang et al. 2008). Recently a seasonal version of Global *NEWS*-2 has been developed for DIN export by world rivers (McCrackin et al. 2014).

The spatial level of detail is basin for Global *NEWS*-2 (Mayorga et al. 2010). RIVERSTRAHLER (Ruelland et al. 2007; Sferratore et al. 2005) and SPARROW (Alexander et al. 2007; McCrackin et al. 2013; Schwarz et al. 2006) can take sub-basin and basin scales for calculations. This depends on the drainage area of a river and the objective of the study. IMAGE-GNM quantifies nutrient fluxes from land to sea by grid, 0.5 latitude by 0.5 longitude (Beusen et al. 2015a; Beusen et al. 2015b). AGNPS also takes the grid scale for calculations, but with flexible sizes (Jianchang et al. 2008). SWAT runs at hydrological response units, which are further aggregated to basins (Arnold et al. 2012; Gassman et al. 2014). In contrast, NUFER runs at the provincial (Ma et al. 2012) or county (Wang et al. in preparation) scale.

Some of the models account for both diffuse and point sources of nutrients in rivers. Figure 1.1 provides examples of such sources. These are IMAGE-GNM (Beusen et al. 2015a), Global *NEWS*-2 (Mayorga et al. 2010), SPARROW (Schwarz et al. 2006) and RVERSTRAHLER (Ruelland et al. 2007). SWAT and AGNPS focus more on diffuse source pollution from agriculture (Shen et al. 2012). NUFER accounts for both diffuse and point sources of nutrients in rivers from agriculture (Ma et al. 2010; Ma et al. 2012). Uncollected human waste and manure direct discharges to surface waters are accounted for in NUFER. IMAGE-GNM, Global *NEWS*-2 and SPARROW provide the source attribution for nutrient river export.

However, only IMAGE-GNM, Global *NEWS*-2 and NUFER cover all of China. And of these, only IMAGE-GNM and Global *NEWS*-2 focus specifically on river export of nutrients. The other models have been applied to individual basins. SWAT and AGNPS were applied to several small Chinese rivers (e.g., Huang et al. 2009b; Shen et al. 2012; Wu & Chen 2013). RIVERSTRAHLER is mainly for European basins (Billen & Garnier 2000; Billen et al. 2005; Ruelland et al. 2007) with an application to the Red River in Vietnam (Garnier & Billen 2002). SPARROW is more for rivers of the United States (Alexander et al. 2007; McCrackin et al. 2013).

The IMAGE-GNM and Global *NEWS*-2 are global models covering over 6000 river basins. These models differ in several aspects. One of them is the source attribution for nutrients in rivers. Global *NEWS*-2 makes a separation among diffuse sources: synthetic fertilizers, animal manure, atmospheric N deposition, and biological N fixation (Mayorga et al. 2010). IMAGE-GNM lumps these sources and provides the source attribution in terms of sub-surface and surface runoff (Beusen et al. 2015a). Unlike Global *NEWS*-2, IMAGE-GNM accounts for some other sources such as aquaculture, vegetation in floodplains and industrial wastewater. Both models ignore direct discharges of animal manure to rivers.

Another difference is in modeling approaches for nutrient cycling. IMAGE-GNM is a non-calibrated, dynamic model (Beusen et al. 2015b). It includes spiraling for in-river nutrient retentions. Spiraling accounts for nutrient uptake by plants, denitrification and sedimentation. Reservoirs are considered as sinks for nutrients. In contrast, Global *NEWS*-2 is a globally calibrated, steady state model (Mayorga et al. 2010). It considers denitrification and reservoirs for nutrient losses from and retentions in rivers. In addition, it also considers water consumption (Mayorga et al. 2010), which can be considerable in Chinese areas with intensive crop production (Li et al. 2008).

An important difference is also in the nutrient forms and application. IMAGE-GNM models fluxes of total N and P. The model has been recently developed and applied for global analyses over the period of the 21st century (Beusen et al. 2015a; Beusen et al. 2015b). Thus, its separate application to Chinese river basins is, so far, limited. In contrast, Global *NEWS*-2 quantifies river export of multi-elements in different forms for past and future years. The model includes N, P, Si and carbon (C) in dissolved inorganic, dissolved organic and particulate forms for the period of 1970-2050. Furthermore, the model quantifies the potential for coastal eutrophication, ICEP. This allows the analyzing of the impact of human activities on coastal water pollution. Future trends are analyzed based on four MEA scenarios (Alcamo et al. 2005; Seitzinger et al. 2010). This gives an opportunity to explore solutions to reduce coastal water pollution that is associated with river export of nutrients.

Global *NEWS*-2 has been widely applied not only for global analyses, but also for regional analyses of river export of nutrients. Examples of such regional analyses are river export of nutrients to coastal waters of Indonesia (Suwarno et al. 2013; Suwarno et al. 2014a; Suwarno et al. 2014b), the Bay of Bengal (Sattar et al. 2014), the Black Sea (Strokal & Kroeze 2013; Strokal et al. 2014c), North America (McCrackin et al. 2013), Africa (Yasin et al. 2010) and Europe (Thieu et al. 2010). This model has also been applied to Chinese basins (Li et al. 2011b; Qu & Kroeze 2010; Qu & Kroeze 2012; Yan et al. 2010; Yu et al. 2015). However, the basin scale limits spatially explicit analyses of nutrient sources within a basin (Mayorga et al. 2010; Seitzinger et al. 2010).

1.3 Knowledge gaps

From the above review I identified the main knowledge gaps for China to address trends in river and coastal water pollution. These are:

1. Lack of information on impacts of human activities in upstream basin areas on downstream water pollution;
2. Incomplete information on the relative importance of some missing sources of nutrients in Chinese rivers. This holds especially true for sources that might be important in the total water pollution. These are uncollected human waste and direct discharges of manure to rivers. Ignoring these sources hinders identification of main causes of coastal eutrophication;
3. Lack of optimistic views on solutions to reduce water pollution. This is needed to cover the full range from worst to best cases to better understand water pollution in China.

These knowledge gaps exist because existing models (Section 1.2.6) are limited in representing the Chinese context. Global models do not account for the importance of upstream pollution in downstream impacts and ignore nutrient sources that are important for China. Other (non-global) models have the disadvantage that they do not cover all of China, and that their applications are often very specific for the local situation. Clearly, there is a need for a model that is developed in particular for China to fill in the knowledge gaps.

In this thesis, I took Global *NEWS*-2 as a starting point. So far, Global *NEWS*-2 is the only model that quantifies past and future trends in river export of nutrients in different forms (of N, P, C and of Si) by source and the associated coastal eutrophication simultaneously (Section 1.2.6). However, Global *NEWS*-2 needs improvements in two major aspects for China: the spatial level of detail and the nutrient sources.

For the spatial level of detail a new modeling approach is needed to be able to analyze the impacts of upstream activities on water quality downstream (<u>Knowledge Gap 1</u>). I aim for a rather simple approach that can be applied easily to large basins in China, but is also able to provide the needed information to realize Knowledge Gap 1. A modeling approach at the sub-basin scale can be a solution here (see Section 1.2.3). It can serve as the first step in understanding the importance of upstream pollution in downstream impacts. And, it does not require as many spatial detailed inputs as, for example, gridded modeling. For nutrient sources approaches of Global *NEWS*-2 need to be modified to account for the missing sources of nutrients in Chinese rivers (<u>Knowledge Gap 2</u>). In addition, Global *NEWS*-2 also requires an update in terms of information for dams in China since this is a global model.

A new model for China can result from the sub-basin scale modeling and improved approaches of Global *NEWS*-2 with other extra information. This new model can then be used to explore solutions to reduce river export of nutrients and thus coastal eutrophication in the future in China. This can be done for both worst and best case scenarios (Knowledge Gap 3). Optimistic scenarios can represent the best cases. I give more information about scenarios in Section 1.5.

1.4 Research objectives

This PhD research aims to address the three abovementioned knowledge gaps. The overall research objective is, therefore, to better understand trends in river export of nutrients to the coastal waters of China by source from sub-basins, and the associated coastal eutrophication. To this end, the Global *NEWS*-2 model is used as a starting point. Five research sub-objectives are formulated to achieve the overall objective (Figure 1.3):

1. To analyze the original Global *NEWS*-2 model for river export of nutrients and the associated coastal eutrophication;
2. To develop a sub-basin scale modeling approach to account for impacts of upstream human activities on downstream water pollution, taking the Pearl River as an example (Knowledge Gap 1);
3. To quantify the relative share of manure point sources to nutrient inputs to rivers at the sub-basin scale (Knowledge Gap 2);
4. To quantify the relative share of sources to river export of nutrients at the sub-basin scale (Knowledge Gaps 1 and 2);
5. To explore optimistic futures to reduce river export of nutrients and coastal eutrophication in China (Knowledge Gap 3).

1.5 Study area and methodology

Each research sub-objective forms a chapter of this PhD study. Figure 1.3 shows the outline of the PhD chapters with a focus on novelties and methods.

In Chapter 2, I analyzed nutrient export by Chinese rivers and coastal eutrophication using Global *NEWS*-2 and ICEP (the first research sub-objective, Figures 1.3 and 1.4). The novelties of this chapter include new insights on the effects of river export of nutrients on coastal eutrophication. I studied sixteen rivers that drain into the Bohai Gulf, Yellow Sea and South China Sea. Rivers that drain into the Bohai Gulf are the Yellow, Liao, Hai, Luan and Dalinghe and Xiaoqinghe. Rivers that drain into the Yellow Sea are the Yangtze, Huai, Fuchunjiang, Menjiang, Yalu and Oujiang. The Pearl, Hanjiang and Jiulong He drain into the South China Sea. The Pearl River consists of the Zhujiang and Dongjiang rivers in Global *NEWS*-2 (Figure 1.4). The drainage areas of these rivers exceed 4 grid cells of 0.5 longitude by 0.5 latitude in Global *NEWS*-2 (Mayorga et al. 2010; Vörösmarty et al. 2000b). The drainage areas of the Yellow is 0.89 million km^2, for the Yangtze it is 1.8 million km^2 and for the Pearl is 0.44 million km^2. Drainage areas of the other Chinese rivers range from 0.01 to 0.27 million km^2.

In Chapter 3, I addressed the second research sub-objective (Figure 1.3). The main novelty is the new sub-basin scale modeling approach. I used the Pearl River to test this new approach. I developed this approach for DIN and DIP export over the period of 1970-2050. I did it by applying approaches of Global *NEWS*-2 with reservoir information from the Global Reservoir and dam Database (GRanD) (Lehner et al. 2011a; Lehner et al. 2011b). This approach quantifies the relative share of sub-basins in the total river export. And this is done by source. The sub-basins of the Pearl were classified into up-, middle- and downstream. In this way it is easier to analyze impacts of upstream activities on downstream water quality.

In Chapter 4, I addressed the third research sub-objective (Figure 1.3). Here, the main novelty is new insights on the relative importance of manure point sources in the total water pollution in China at the sub-basin scale. For this, I extended the sub-basin model to other basins in China and included manure point sources using information from NUFER (Ma et al. 2012). Inputs of total dissolved N and P to Chinese rivers by source at the sub-basin scale are quantified. Here, the study area covers the six main rivers (Tong et al. 2015). These are the Yellow, Hai, Liao, Yangtze, Huai and Pearl (Figure 1.4). The large drainage areas of the Yangtze, Yellow and Pearl rivers are divided into sub-basins. Hai, Huai and Liao are considered as individual sub-basins in this study. In total the study area consists of 25 sub-basins that are classified into up-, middle- and downstream.

In Chapter 5, I addressed the fourth research sub-objective (Figure 1.3). Here the main novelties are (1) the newly developed *MARINA* model for China and (2) new insights on the relative share of sources in coastal water pollution at the sub-basin scale. The new model quantifies river export of dissolved N and P (inorganic and organic) by source from sub-basins for past and future years and the coastal eutrophication. This *MARINA* model combines modeling approaches of Chapters 3 and 4 with extra new components (Figure 1.3). First, the sub-basin scale modeling approach of Chapter 3 was improved and extended to other basins and nutrient forms. Second, the model quantifies the relative importance of the sources including manure point sources (from Chapter 4) and uncollected human waste and includes updated information on dams. Uncollected human waste was included based on information and approaches of the NUFER model for the rural population (Ma et al. 2012) and of Morée et al. (2013) for the urban population. Coastal eutrophication is quantified using ICEP.

In Chapter 6, I addressed the fifth research sub-objective (Figure 1.3). Here I developed optimistic scenarios representing the best case for Chinese rivers. I took GO of the MEA as an illustrative example of the worst case for Chinese rivers. This is because of poor manure and human waste management that is assumed in GO for urbanization and industrialization of animal production. I also explored the effects of the recently introduced environmental policy to reduce river pollution from agriculture (MOA 2015). I analyzed effects of these scenarios on coastal eutrophication using ICEP (Garnier et al. 2010).

In addition to modeling and ICEP, I used sensitivity analyses and ArcGIS in the chapters. ArcGIS was used to visualize model results and to prepare model inputs at the sub-basin scale. Most model inputs were from datasets of Global *NEWS*-2 at the grid scale of 0.5 latitude by 0.5 longitude (Bouwman et al. 2009; Fekete et al. 2010; Mayorga et al. 2010; Seitzinger et al. 2010; Van Drecht et al. 2009). Some inputs were from NUFER (Ma et al. 2012) and other sources (Lehner et al. 2011b; Morée et al. 2013). Global *NEWS*-2, literature and expert knowledge are sources for model parameters.

1.6 Outline of the PhD thesis

The PhD thesis consists of seven chapters (Figure 1.3). In Chapter 1, I provide the background information and thesis' objectives. In Chapters 2-6, I focus on research objectives. And in Chapter 7, I discuss thesis' modeling approaches and I draw main conclusions. From my research the reader can gain a better understanding of the main causes of river and coastal water pollution and how to reduce this pollution in the coming years in China. This information is important when developing effective environmental policies. Hopefully, this will contribute to increase the future availability of clean water. I believe that my research can serve as an example for other regions with similar environmental problems.

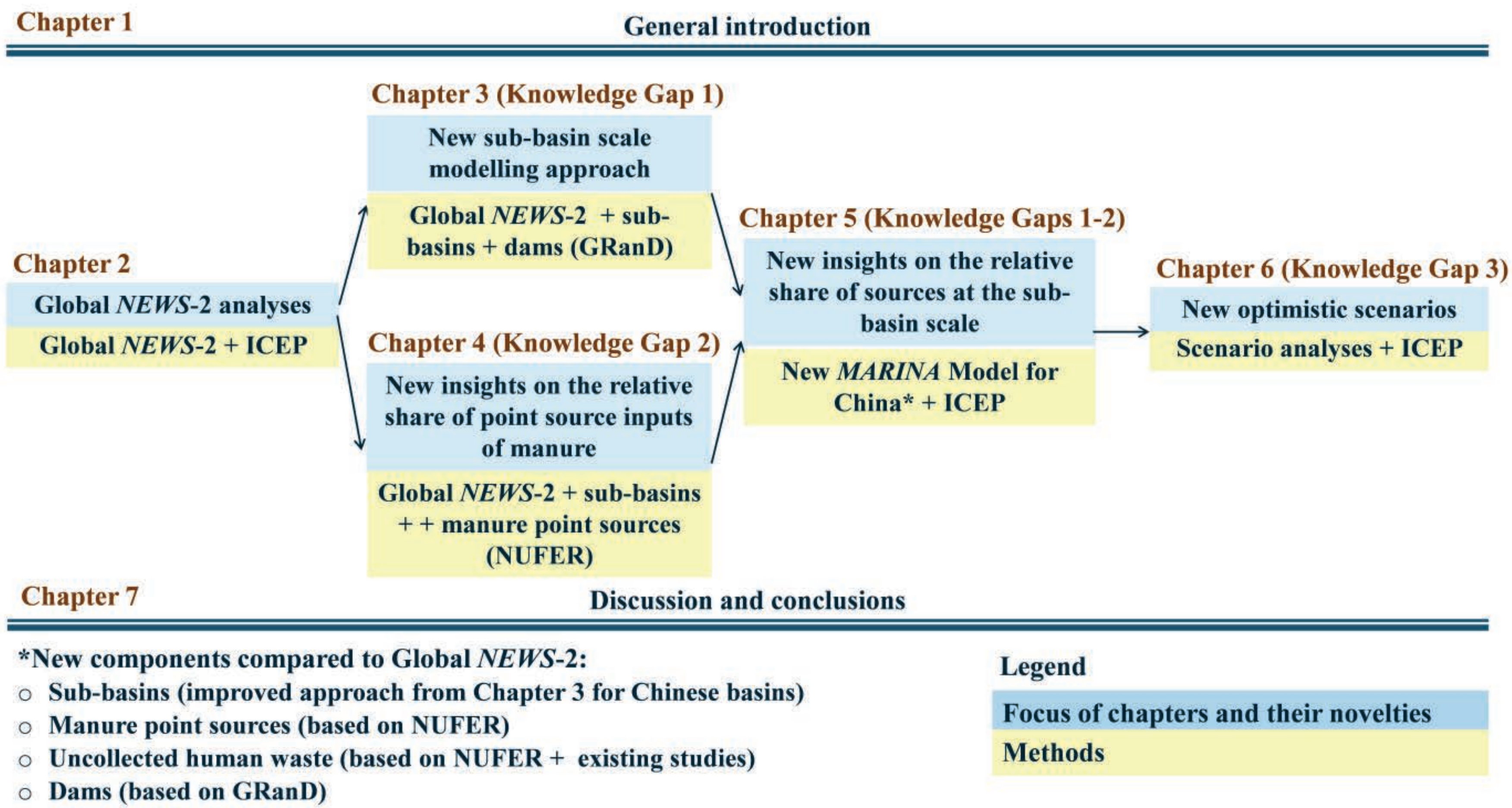

Figure 1.3. *An overview of the PhD thesis structure with the focus of chapters, their novelties and methods. Knowledge gaps are given in Section 1.3. Global NEWS-2 is brief for Nutrient Export from WaterSheds (NEWS) (Mayorga et al. 2010; Seitzinger et al. 2010). NUFER is in brief for NUtrient flows in Food chains, Environment and Resources use (Ma et al. 2010; Ma et al. 2012). GRanD is the Global Reservoir and dam Database (Lehner et al. 2011a; Lehner et al. 2011b).*

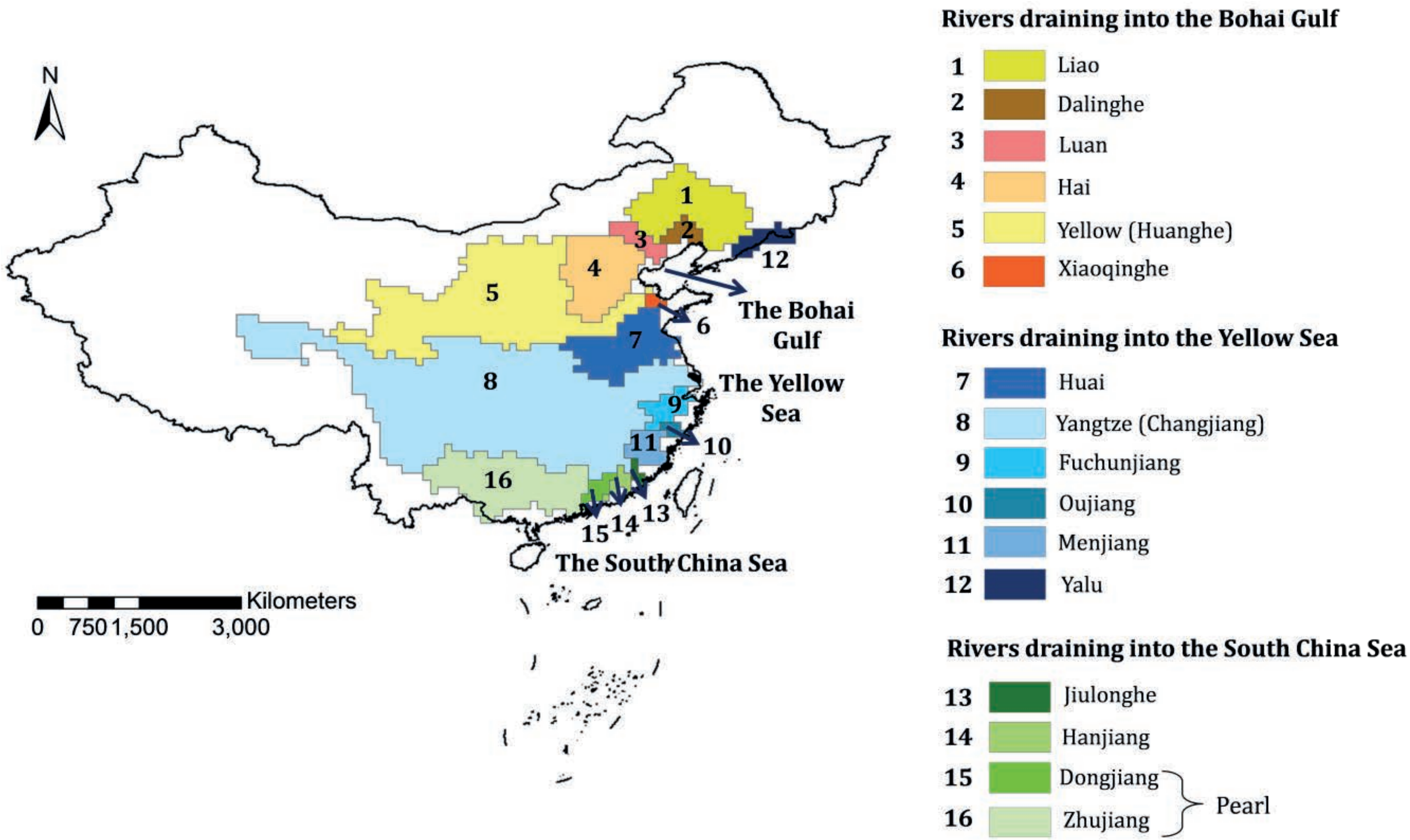

Figure 1.4. *Sixteen rivers draining into the Bohail Gulf, Yellow Sea and South China Sea in China. Dongjiang and Zhujiang basins form the Pearl basin in this PhD study. Source: Global NEWS-2 (Mayorga et al. 2010; Vörösmarty et al. 2000a).*

Chapter 2.

Increasing eutrophication in the coastal seas of China from 1970 – 2050

2

Abstract

We analyzed the potential for eutrophication in major seas around China: the Bohai Gulf, Yellow Sea and South China Sea. We model the riverine inputs of nitrogen (N), phosphorus (P) and silica (Si) to coastal seas from 1970 – 2050. Between 1970 and 2000 dissolved N and P inputs to the three seas increased by a factor of 2-5. In contrast, inputs of particulate N and P and dissolved Si, decreased due to the damming of rivers. Between 2000 and 2050, the total N and P inputs increase further by 30-200%. Sewage is the dominant source of dissolved N and P in the Bohai Gulf, while agriculture is the primary source in the other seas. In the future, the ratios of Si to N and P decrease, which increases the risk of harmful algal blooms. Sewage treatment may reduce this risk in the Bohai Gulf, and agricultural management in the other seas.

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2.1 Introduction

The Chinese coastal waters have been receiving increasing amounts of nutrients from rivers due to the rapid economic development and population growth (Liu et al. 2012; Ma et al. 2012; Qu & Kroeze 2010; Qu & Kroeze 2012; Sutton et al. 2013; Weng 2007). Currently, approximately 1.3 billion people live in China (Ma et al. 2010), with annual incomes of approximately 3500 US dollars per person (Qu & Kroeze 2010). The population density increased by 50% between 1970 and 2000, while the per capita income increased by approximately 600% in the large river basins in China (Qu & Kroeze 2010); these changes generated environmental pressures. The increased availabilities of nitrogen (N) and phosphorus (P) are included among these pressures and are largely associated with trends in agriculture, urbanization and waste management. Increasing the nutrient availability in aquatic systems may lead to eutrophication. Coastal eutrophication is an enrichment of coastal waters by nutrients, such as N and P (Richardson & Jørgensen 1996). This nutrient enrichment may increase the production of phytoplankton, such as harmful algae (Carstensen et al. 2007), leading to harmful algal blooms (HABs).

Increased food production has increased the nutrient inputs to the environment. The production and consumption of animal- (e.g., eggs, meat, milk) and crop-derived (e.g., cereals) products have increased with the growing population and economy (Ma et al. 2012; Riedel et al. 2012). For example, grain production was 70% higher in 2005 than in the 1980s (Ma et al. 2012). After 30 years of rapid economic growth, the living standards have improved. Many people have shifted their dietary preferences toward animal-derived products (Li et al. 2011a; Ma et al. 2012). Consequently, meat consumption has more than doubled since 1982 (Qu et al. 2005). These dietary changes are associated with increased nutrient inputs for agriculture, allowing rivers to transport these nutrients further to coastal waters and increasing the risk of harmful algal blooms (Heisler et al. 2008). Food production and consumption in China may continue to increase in the future (Bouwman et al. 2009).

Urbanization is another important source of N and P inputs to the rivers and coastal waters in China (Ma 2012; Qu & Kroeze 2010). Sewage systems are urban point sources for the nutrients in rivers. The urban population increased from approximately 100 million people in the 1970s to approximately 400 million people in the 2000s, demonstrating a growth rate twice as fast as the world average during the same period (e.g., Qu & Kroeze 2010). In 2008, the urban population reached 590 million (Ma 2012). Furthermore, animal production seems to move from rural areas toward more urban areas due to the increased demand for meat in cities (Ma et al. 2010; Ma et al. 2012). The urban population may continue to increase in the coming years (Ermolieva et al. 2009; Qu & Kroeze 2010); this increase generates more sewage systems and discharges additional human waste into aquatic systems. However, wastewater treatment may not be able to keep up with the urbanization progress. Between 2000-2005, approximately 30-45% of the total amount of wastewater was treated before being discharged into waters. Although wastewater treatment has improved in recent years, the absolute amount of sewage discharge continues to increase dramatically every year as the urbanized areas continue to expand (Liu & Qiu 2007).

While the riverine inputs of N and P to Chinese seas have increased in recent decades (Qu & Kroeze 2010), the inputs of dissolved silica (DSi) to coastal seas have continued to decrease worldwide due to the damming of rivers (Billen & Garnier 2007). This decreases the nutrient ratios (N:P:Si) in aquatic systems, favoring the development of HABs (e.g., cyanobacteria) in estuary systems. These events may be associated with hypoxic events (oxygen depletion) and fish death (Carpenter et al. 1998; Galloway et al. 2008; Smith & Schindler 2009; Sutton et al. 2013). HABs develop when coastal waters do not contain sufficient silica levels as the N and P loads increase (Billen & Garnier 2007; Garnier et al. 2010). Some studies (e.g., Diaz & Rosenberg 2008; Selman et al. 2008; Sutton et al. 2013; Wang et al. 2008; Xiao et al. 2007) have reported an increase in areas of harmful algal growth along the Chinese coastal zones. In the 2000s, > 100 algal bloom events were reported for the entire Chinese coast (e.g., Tang et al. 2006a).

So far, few studies have linked coastal eutrophication to land-based drivers for the three major seas in China: the Bohai Gulf, Yellow Sea and South China Sea. The riverine inputs of N and P to the coastal waters of China were studied (Qu & Kroeze 2010; Qu & Kroeze 2012); however, the environmental impacts of these nutrients on coastal waters were poorly addressed, and no sea-specific analyses of nutrient inputs were performed. Various studies have focused on land-based sources of N and P in rivers at the national and/or provincial levels (Ma et al. 2010; Ma et al. 2012; Sutton et al. 2013). Some studies analyzed specific locations, such as the Pearl River estuaries (Ho et al. 2010; Xu et al. 2010a), while focusing on the current management of water resources (Cui et al. 2007; Lu et al. 2007; Weng 2007); others involved the Yangtze River (Li et al. 2012). In addition, eutrophication in selected lakes in China has been studied (Qu et al. 2005). However, none of these studies have addressed eutrophication in the three major seas of China. An integrated analysis of the main drivers and sources of coastal eutrophication in the Bohai Gulf, Yellow Sea and South China Sea is absent. Consequently, sea-specific management strategies to reduce nutrient inputs to rivers and coastal seas have not been explored.

Integrated model, such as Global *NEWS*-2 (Nutrient Export from WaterSheds) may contribute to the integrated analyses of causes, effects and solutions. The Global *NEWS*-2 model was developed to understand the relationship between human activities on land and nutrient enrichment in coastal waters (Mayorga et al. 2010). This model has been utilized in many studies to evaluate inputs of N, P, carbon (C), and Si in different forms to coastal seas (Bouwman et al. 2009; Fekete et al. 2010; Seitzinger et al. 2010; Strokal & de Vries 2012; Strokal & Kroeze 2013; Suwarno et al. 2013; Thieu et al. 2010; Van der Struijk & Kroeze 2010; Van Drecht et al. 2009; Yan et al. 2010; Yasin et al. 2010). The model assesses the potential for coastal eutrophication through the ICEP (Indicator for Coastal Eutrophication Potential) approach (Billen & Garnier 2007; Garnier et al. 2010). This approach has been widely accepted and used in various studies for global (Garnier et al. 2010) and regional (Crosswell et al. 2012; Dauvin et al. 2008; Liu et al. 2012; Romero et al. 2012; Strokal & Kroeze 2013; Thieu et al. 2011; Wang et al. 2013a) analyses of coastal eutrophication.

The primary objective of this study is to assess the potential eutrophication in the three major seas of China as influenced by riverine inputs of nitrogen, phosphorus and silica in 1970 – 2050. Toward this purpose, we applied the Global *NEWS*-2 model and ICEP approach. Global *NEWS*-2 can be used to determine the riverine nutrient inputs to coastal seas, which is referred to here as nutrient export by rivers. First, we analyzed the past and future trends of the main drivers for the export of nutrients to coastal seas through rivers (Section 2.3.1). Second, we analyzed the trends in the river export of N, P and Si in different forms (dissolved inorganic, dissolved organic and particulate), as well as their main sources (Section 2.3.2). Third, we evaluated the eutrophication potential in the three major seas of China: the Bohai Gulf, the Yellow Sea and the South China Sea from 1970-2050 (Section 2.3.3). Finally, we discussed future management of eutrophication in these three seas by illustrating some possible sea-specific management options (Section 2.3.4).

2.2 Methodology

2.2.1 Study area

The study area includes 16 rivers that drain into the coastal waters of the Bohai Gulf, Yellow Sea and South China Sea in China (Figure 2.1). These rivers are the Global *NEWS*-2 (version 2) rivers with basin areas exceeding 4 grid cells of 0.5^0 longitude by 0.5^0 latitude in size. We group the selected river basins into three regions that drain into the Bohai Gulf (or Pohai Gulf in Global *NEWS*-2), Yellow Sea and South China Sea (or North-South China Sea in Global *NEWS*-2). The names and delineation of the river basins are taken from the Global *NEWS*-2 model (see Section 2.2.2).

The river basins draining into the Bohai Gulf cover approximately 35% of the total study area (Figure 2.1a). These basins include the Yellow (or Huanghe, the largest river of this region), Liao (or Hun), Hai (or Yongding), Luan, Dalinghe and Xiaoqinghe (Figure 2.1b, c). The river basins draining into the Yellow Sea cover approximately 55% of the study area and include the Yangtze (or Changjiang, the largest river in China), Huai, Fuchunjiang (or Qiang Tang), Menjiang (or Minjiang), Yalu and Oujiang. The East China Sea is considered part of the Yellow Sea in this study. The remaining study area includes the river basins of the South China Sea: the Zhujiang, Dongjiang, Hanjiang and Jiulong He (Figure 2.1). The Zhujiang and Dongjiang form the Pearl River basin in this study.

2.2.2 Global *NEWS*-2 model

2.2.2.1 Model description

We used the Global *NEWS*-2 (Nutrient Export from WaterSheds, version 2) model to analyze the export of nutrients through the rivers to the coastal waters of the Bohai Gulf, Yellow Sea and South China Sea. The Global *NEWS*-2 model estimates river export of nitrogen (N), phosphorus (P), carbon (C) and silica (Si) at the river mouth in different forms: dissolved inorganic (DIN, DIP, DSi), dissolved organic (DON, DOP, DOC) and particulate (PN, PP, DOC) (Mayorga et al. 2010; Seitzinger et al. 2010). Below, we briefly describe how the river exports of N, P and Si are modeled. Detailed information is provided by Mayorga et al. (2010). The models for the individual nutrient forms are explained by Dumont et al. (2005) for the dissolved inorganic N, Harrison et al. (2010) for the dissolved inorganic P, Harrison et al. (2005a) for the dissolved organic N and P, Beusen et al. (2005) for the particulate N and P, and Beusen et al. (2009) for the dissolved Si.

<u>The river export of dissolved N and P</u> is quantified based on mass-balance (Mayorga et al. 2010). Box 2.1 summarizes the main formulas. The dissolved N and P export by rivers (Yld_F, kg km^{-2} year^{-1}) is estimated as the sum of the nutrient export to the rivers from diffuse ($RSdif_F$, kg km^{-2} year^{-1}) and point ($RSpnt_F$, kg km^{-2} year^{-1}) sources corrected for losses from and retention within the river systems ($FEriv_F$, 0-1) (Box 2.1).

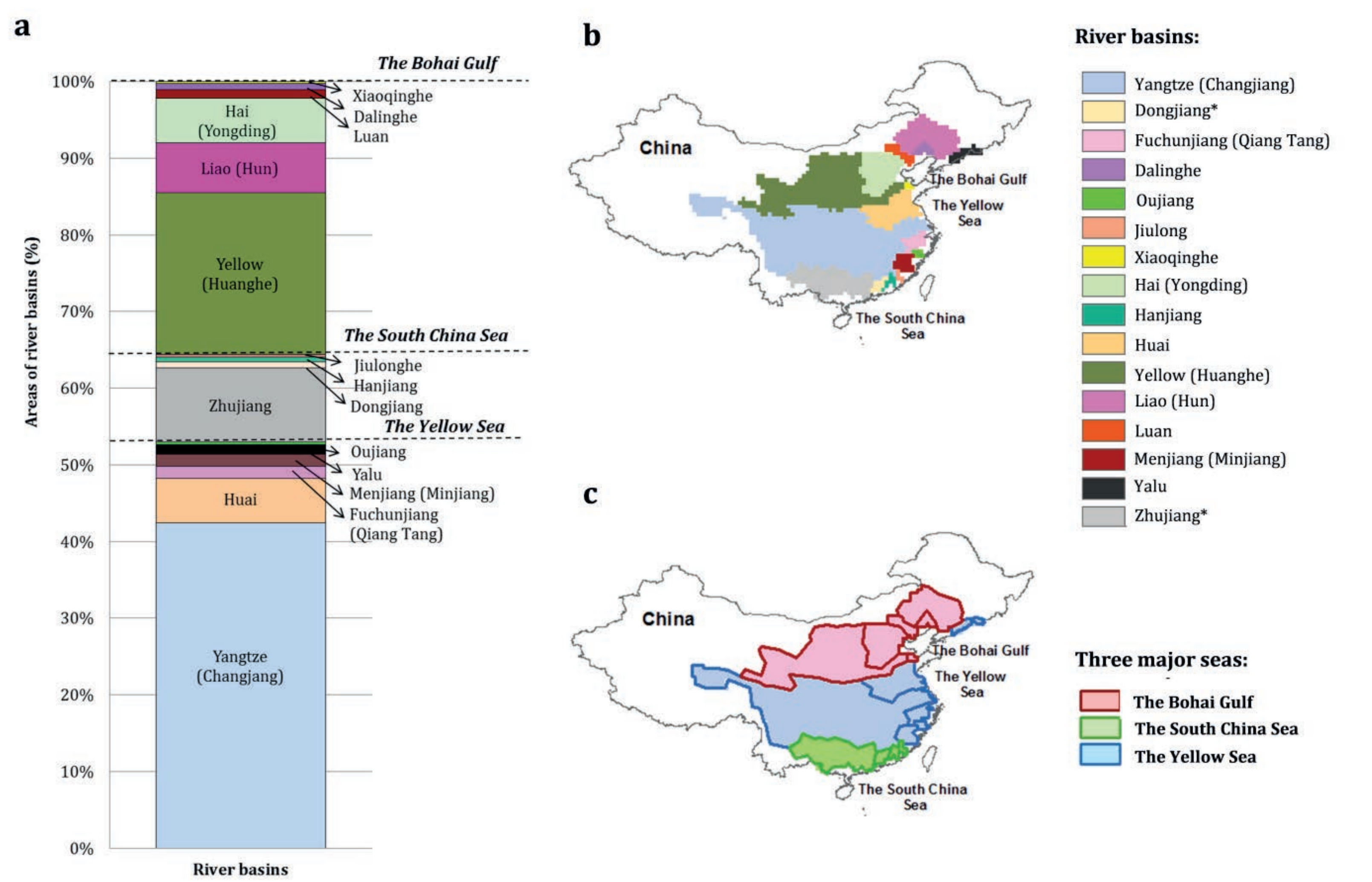

Figure 2.1. *Shares of the rivers in the total studied basin area (a), their locations in China (b), and the drainage basins of the Bohai Gulf, Yellow Sea and South China Sea (c). Based on Global NEWS-2 (Mayorga et al. 2010). * The Zhujiang and Dongjiang form the Pearl River basin in Global NEWS-2.*

Diffuse sources of dissolved nutrients for rivers ($RSdif_{F}$, kg km^{-2} year^{-1}) originate from agricultural ($RSdif_{ant.F}$, kg km^{-2} year^{-1}) and non-agricultural ($RSdif_{nat.F}$, kg km^{-2} year^{-1}) areas (see Box 2.1). For the agricultural areas, the dissolved N and P export to rivers ($WSdif_{ant.E} \cdot FE_{ws.F}$ in Box 2.1) is modeled as a function of the net nutrient inputs to watersheds ($WSdif_{ant.F}$, kg km^{-2} year^{-1}) from synthetic fertilizers ($WSdif_{fe.F}$, kg km^{-2} year^{-1}), the excretion of animal manure ($WSdif_{ma.F}$, kg km^{-2} year^{-1}) (for dissolved N and P), atmospheric N deposition ($WSdif_{dep\ ant.N}$, kg km^{-2} year^{-1}), biological N fixation by crops ($WSdif_{fix.ant.N}$, kg km^{-2} year^{-1}) (for dissolved N) corrected for nutrient export ($WSdif_{ex.F}$, kg km^{-2} year^{-1}) via crop harvesting and animal grazing, and for nutrient retention within watersheds (land) ($FE_{ws.F}$, 0-1). The DIP inputs to rivers from the weathering of P-minerals and dissolved organic N and P inputs to rivers from soil leaching are estimated as a function of the annual mean runoff from the land to the surface waters (an export-coefficient approach, $RSdif_{ec.F}$, kg km^{-2} year^{-1}) with a correction for the fraction of agricultural areas (Ag_{fr}, 0-1) (see Box 2.1). For the non-agricultural areas, the DIN inputs to the rivers are modeled as a function of the N inputs to the watersheds from atmospheric N deposition ($WSdif_{dep.nat.N}$, kg km^{-2} year^{-1}) and biological N fixation by crops ($WSdif_{fix.nat.N}$, kg km^{-2} year^{-1}) with a correction for the N retention within watersheds ($FE_{ws.nat.F}$, 0-1). The dissolved inorganic P from weathering and the dissolved organic N and P from leaching of organic materials to rivers are modeled through an export-coefficient approach ($RSdif_{ec.F} \cdot (1\text{-}Ag_{fr})$ in Box 2.1).

Nutrient inputs to rivers from point sources ($RSpnt_{F}$, kg km^{-2} year^{-1}) are calculated as a function of the N and P emissions to watersheds that originated from wastewaters (for N and P: $WShwExc_{F}$, kg km^{-2} year^{-1}) and detergents (for P: $WShwDet_{P}$, kg km^{-2} year^{-1}) (Box 2.1). These N and P emissions are corrected for the fraction of nutrient removal via wastewater treatment ($hw_{frem.E}$, 0-1), population with a sewage connection (I, inh km^{-2} year^{-1}) and the export fraction of the element (N, P) emitted to rivers as a form ($FEpnt_{F}$) (see Box 2.1).

The nutrient losses from and retention within the river network are represented by three terms in the model (Box 2.1): (i) nutrient losses in the river itself, such as via denitrification (L_{F}, 0-1, for DIN only); (ii) nutrient retention caused by dams/reservoirs (D_{F}, 0-1 for dissolved inorganic N and P); and (iii) nutrient losses via water consumption (FQrem, generic for all nutrient forms, 0-1) for different human activities, such as irrigation and hydropower. L_{F} is estimated based on the area of the basin. The $D_{F.i}$ for each reservoir (i) in a basin is calculated as a function of the changes in the annual water residence time. The D_{F} for each basin is the discharge-weighed retentions of each reservoir in this basin with a correction for the actual water discharge by the basin (Qact, km^{3} year^{-1}) (see Box 2.1). The FQrem is the ratio between the actual water discharge

(Qact, after the water is removed for human activities, km^3 $year^{-1}$) and the natural water discharge (Qnat, before water is removed, km^3 $year^{-1}$).

The river export of particulate N and P (Yld_F, kg km^{-2} $year^{-1}$) is modeled through a regression analysis that accounts for the relationship with the total suspended solids (TSS) (Beusen et al. 2005; Mayorga et al. 2010). The principle involves the determination of the fluxes in the nutrient form (F: PP, PN) in the pre-dam situation ($Yld_{F.pred}$, kg km^{-2} $year^{-1}$) corrected by the fraction of TSS retained in reservoirs / dams (D_{TSS}, 0-1):

$$Yld_F = (1 - D_{TSS}) \cdot Yld_{F.pred} \tag{2.1}$$

The D_{TSS} is quantified as a function of the changes in the water residence time. The $Yld_{F.pred}$ is based on the estimated TSS pre-dam yield from a regression analysis, which accounts for the major factors controlling TSS export. These controlling factors are the precipitation intensity, relief, lithology (e.g., carbonate rocks, basalt, sand/sandstone), wetland rice and marginal grassland (Beusen et al. 2005; Mayorga et al. 2010).

The river export of dissolved Si is based on an empirical relationship with TSS or DIP (Beusen et al. 2009), which is similar to the approach used to estimate particulate N and P. The pre-dam DSi fluxes are derived on the basis of TSS or DIP retention in reservoirs. The main controlling factors of DSi export are the annual precipitation, bulk density, volcanic rocks, slope, and land use type (e.g., wetland, arable areas). These factors were used to estimate the DSi export at the river mouth through a regression method (Beusen et al. 2009).

Several sources were used to derive the input data for the Global *NEWS*-2 model (Seitzinger et al. 2010). The input data for diffuse sources of nutrient inputs to rivers (e.g., synthetic fertilizers, animal manure, atmospheric N deposition and biological N fixation, export by harvesting and grazing) are described by Bouwman et al. (2009). Van Drecht et al. (2009) describes the input data for point sources of nutrients in rivers (N and P in human waste and detergents, nutrient removal, and population with a sewage connection) and their socio-economic drivers (total population density, urban population, GDP), while Fekete et al. (2010) explains the input data for the hydrology and climate (e.g., annual precipitation, water discharges). Most of the input data were provided on a gridded scale (0.5 by 0.5 degrees in resolution) by the IMAGE (Integrated Assessment model) and WBM (Water Balance Model) models. These inputs were aggregated to a river basin scale (Seitzinger et al. 2010).

The model has been implemented for the years 1970, 2000, 2030 and 2050. For the future years (2030, 2050), the storylines for four Millennium Ecosystem Assessment (MEA) scenarios (Alcamo et al. 2005; Carpenter et al. 2006) were interpreted quantitatively to produce the model inputs for the Global *NEWS*-2 model (Seitzinger et al. 2010). These scenarios are the Global Orchestration (GO), TechnoGarden (TG), Order from Strength (OS) and Adapting Mosaic (AM). These scenarios address three main aspects toward nutrient management: agricultural practices (e.g., synthetic fertilizer use and animal manure excretion) (Bouwman et al. 2009), sewage management (e.g., nutrient treatment, population with a sewage connection) (Van Drecht et al. 2009), and hydrology and climate (Fekete et al. 2010). In this study, we focused on the GO and AM because they represent different views on the socio-economic development (globalization in GO and regionalization in AM) and different approaches toward managing environmental issues (reactive in GO and proactive in AM). Below, we describe the GO and AM scenarios. The details describing the TG and OS can be found elsewhere (Alcamo et al. 2005; Carpenter et al. 2006; Seitzinger et al. 2010).

The GO scenario assumes that the world is globalized with reactive nutrient management. In this scenario, economic growth is high due to the globally connected markets. The population growth will increase (8.2 billion people globally in 2050), but not as much as in the regionalized AM scenario (9.6 billion people globally in 2050) (Alcamo et al. 2005; Bouwman et al. 2009). Agricultural practices will be diversified to improve human well-being and to sustain a growing population (Alcamo et al. 2005; Bouwman et al. 2009; Seitzinger et al. 2010). Hydropower production may also increase due to the globalization trends, resulting in higher water demand. This change will drive the construction of new dams (Fekete et al. 2010; Seitzinger et al. 2010), decreasing the nutrient export through sediment trapping. Sanitation will be improved, and more people will have access to sewage systems. Furthermore, the current wastewater treatment (e.g., mechanical treatment) method will be replaced by more advanced processes (e.g., biological treatment), increasing the efficiency of the N and P removal. Consequently, the N and P inputs to rivers may decline (Seitzinger et al. 2010; Van Drecht et al. 2009).

The regionalized AM scenario with proactive nutrient management assumes that the economic growth is not as high as in the GO scenario because the markets are restricted to regional or national boundaries. The agricultural productivity depends on economically visible technologies combined with local knowledge and experience (Alcamo et al. 2005; Carpenter et al. 2006; Seitzinger et al. 2010). On one hand, this scenario moderately increases the agricultural productivity compared to GO. On the other hand, the nutrient management in agriculture may become more efficient: synthetic fertilizers will be partially replaced by animal manure and human waste from households (Bouwman et al. 2009; Seitzinger et al. 2010). These changes may lower the nutrient inputs to rivers from agricultural activities compared to other scenarios. The water demand for different human practices (e.g., hydropower, irrigation) will increase as in GO (Fekete et al. 2010; Seitzinger et al. 2010). People will not have enough access to sewage systems, and the improvements for the sanitation systems are not as dramatic as in GO (Seitzinger et al. 2010; Van Drecht et al. 2009). The N and P inputs to rivers may increase from sewage systems unless sewage treatment is efficient. Long-term solutions to reduce coastal eutrophication are obvious in this scenario due to the assumed proactive management approach. However, those solutions are financially limited due to regionalization (Alcamo et al. 2005; Carpenter et al. 2006; Seitzinger et al. 2010).

2.2.2.2 Model performance

Global *NEWS*-2 was calibrated and validated for rivers worldwide. The validation results indicate that the Global *NEWS*-2 model explains 50-70% of the variation in the observations of the dissolved N and P for large rivers in the world, including Chinese rivers (e.g., Yangtze, Pearl) (Mayorga et al. 2010). Beusen et al. (2009) evaluated the model for dissolved silica river export worldwide. Beusen et al. (2005) reported on the performance of the model for particulate forms of N and P. These studies revealed an acceptable model performance for these N and P forms worldwide. Some studies validated the model at the regional scale, including the Black Sea (Strokal & Kroeze 2013), South America (Van der Struijk & Kroeze 2010) and Africa (Yasin et al. 2010). These validation results revealed good performance by the model for these regions. For the Chinese river basins, the model was validated by Qu and Kroeze (2010) using 11 rivers (including large rivers: the Yangtze, Yellow and Pearl) for dissolved inorganic and organic N and P. The modeled dissolved N and P values are generally in line with the observed values.

In this study, we explored the performance of the model further using three indicators: Pearson's coefficient of determination (R_P^2), the Nash-Sutcliffe efficiency (R_{NSE}^2) and Model error (ME). The Pearson's coefficient of determination (R_P^2) is the square of the correlation coefficient ($r_{x,y}$), and it indicates the proportion of the variance in measured data explained by the model (Moriasi et al. 2007): the closer R_P^2 is to 1, the better the fit between measured and modeled values. The Nash-Sutcliffe efficiency (R_{NSE}^2) determines how well the plots of the measured and modeled values fit the 1:1 line. Values between 0 and 1 are acceptable levels of performance (Moriasi et al. 2007). The Model Error (ME) measures the difference between the measured and modeled values as a percentage (Alexander et al. 2002a; Moriasi et al. 2007). Values below 25 indicate that the model displays good performance (Moriasi et al. 2007).

Box 2.1. *Summary of Global NEWS-2 equations from Mayorga et al. (2010) used to quantify dissolved inorganic (DIN, DIP) and dissolved organic (DON, DOP) nitrogen (N) and phosphorus (P) exported at the river mouth (kg km^{-2} year^{-1}).*

DISSOLVED NUTRIENT FORMS (F) IN YIELDS (Yld, kg km^{-2} year^{-1}) EXPORTED BY RIVERS AT THE RIVER MOUTH

$$Yld_F = (RSdif_F + RSpnt_F) \cdot FEriv_F$$

Nutrient inputs to rivers from diffuse sources ($RSdif_F$)

$\mathbf{RSdif_F} = RSdif_{ant.F} + RSdif_{nat.F}$	*For N & P forms (F)*
$RSdif_{ant.F} = (WSdif_{ant.E} \cdot FE_{ws.F}) + (RSdif_{ec.F} \cdot Ag_{fr})$	
$WSdif_{ant.N} = WSdif_{fix.ant.N} + WSdif_{dep.ant.N} + WSdif_{fe.N} + WSdif_{ma.N} - WSdif_{ex.N}$	*For N forms*
$WSdif_{ant.P} = WSdif_{fe.P} + WSdif_{ma.P} - WSdif_{ex.N}$	*For P forms*
$RSdif_{ec.F} = EC_F \cdot R_{nat}$	*For DIP / DOP / DON forms*
$FE_{ws.F}$ = function of R_{nat}	
$RSdif_{nat.F} = (WSdif_{nat.E} \cdot FE_{ws.nat.F}) + (RSdif_{ec.F} \cdot (1-Ag_{fr}))$	
$WSdif_{nat.N} = WSdif_{fix.nat.N} + WSdif_{dep.nat.N}$	*For DIN only* *(DON / DOP / DIP/ = 0)*

Nutrient inputs to rivers from point sources ($RSpnt_F$)

$\mathbf{RSpnt_F} = [(1 - hw_{frem.E}) \cdot I \cdot WShw_E] \cdot FEpnt_F$	*For N & P forms (F)*
$WShw_N = WShwExc_N$	*For DIN / DON*
$WShw_P = WShwExc_P + WShwDet_P$	*For DIP / DOP*
$FEpnt_F$ is a linear, empirical function of $hw_{frem.E}$	*For DIN only*
calibrated or default constant	*For DIP / DOP/ DON*

Nutrient losses within the river network ($FEriv_F$)

$\mathbf{FEriv_F} = (1-L_F) \cdot (1-D_F) \cdot (1-FQrem)$	*For N & P forms*
L_{DIN} is a function of basin area	*For DIN only*
$D_F = (1/Qact) \cdot \sum_{i=1...n}(Qact_i \cdot D_{F.i})\quad 0 \le D_F \le D_{F.max}$	*For DIN / DIP*
$D_{F.i}$ is a function of water residence time	
$FQrem = 1 - Qact/Qnat$	*For N & P forms*

Abbreviations

Yld_F – river export of nutrient form (F: DIN, DIP, DON, DOP) at the river mouth in yields (Yld: kg km^{-2} year^{-1}).

$RSdif_F$ – inputs of nutrient form (F) to rivers from diffuse sources (kg km^{-2} year^{-1}) over agricultural ($RSdif_{ant.F}$, kg km^{-2} year^{-1}) and non-agricultural ($RSdif_{nat.F}$, kg km^{-2} year^{-1}) areas. $WSdif_{ant.E}$ – net inputs of nutrient element (E: N, P) to watersheds over agricultural areas (kg km^{-2} year^{-1}), resulting from N-fixation (for N: $WSdif_{fix.ant.N}$, kg km^{-2} year^{-1}), N-deposition (for N: $WSdif_{dep.ant.N}$, kg km^{-2} year^{-1}), synthetic fertilizer use ($WSdif_{fe.F}$, kg km^{-2} year^{-1}) and the excretion of animal manure ($WSdif_{ma.F}$, kg km^{-2} year^{-1}) on land minus nutrient export via crop harvesting and animal grazing ($WSdif_{ex.F}$, kg km^{-2} year^{-1}). $WSdif_{nat.E}$ – net inputs of nutrient element (E: N, P) to watersheds over non-agricultural areas (kg km^{-2} year^{-1}), resulting from N-fixation ($WSdif_{fix.nat.N}$, kg km^{-2} year^{-1}) and N-deposition ($WSdif_{dep.nat.N}$, kg km^{-2} year^{-1}). $FE_{ws.F}$ and $FE_{ws.nat.F}$ – the fractions of nutrient inputs to rivers exported from agricultural watersheds (land) ($FE_{ws.F}$, 0-1) and non-agricultural watersheds ($FE_{ws.nat.F}$, 0-1) as nutrient form (F). $RSdiff_{ec.F}$ – inputs of nutrient form (F) to rivers from weathering of P-contained minerals (for DIP) and leaching of organic materials (for DON, DOP) from agricultural (Ag, 0-1) and non-agricultural (1-Ag, 0-1) areas, estimated based on an export-coefficient method as a function of annual mean runoff from land to streams (kg km^{-2} year^{-1}).

$RSpnt_F$ – inputs of nutrient form (F) to rivers from point sources (kg km^{-2} year^{-1}). $WShw_E$– inputs of nutrient element (E) to watersheds (land) resulted from human excrements (for N/P: $WShwExc_{N/P}$) and detergents (for P: $WShwDet_P$, kg km^{-2} year^{-1}). These are estimated based on the relationship with gross domestic products (GDP) (Van Drecht et al. 2009). $hw_{frem.E}$ – removal of nutrient element (E) via waste water treatment plants (fraction, 0-1). I – population density connected to sewage systems (fraction, 0-1). $FEpnt_F$ – the fraction of sewage effluent exported to river as nutrient form (F) (fraction, 0-1).

$FEriv_F$ – the fraction of nutrient form (F) that is exported at the river mouth (fraction, 0-1). L_F – losses of nutrient form (F, for DIN only) from the river system by, for instance, denitrification, metabolic processes in the river etc (fraction, 0-1). D_F – discharge-weighted retention of nutrient form (F) by reservoirs / dams with the river network (fraction, 0-1). FQrem – losses of nutrients (generic for each nutrient form) from the river system by withdrawing water for different purposes (e.g. irrigation, hydropower production) (fraction, 0-1). It is estimated based on natural (before water is consumed, Qnat, km^3 year^{-1}) and actual (after water is consumed Qact, km^3 year^{-1}) water discharge at the river mouth.

We calculated the three indicators for the dissolved inorganic N and P because most of the total N and P entering the Chinese seas are in dissolved inorganic forms (see also Figure 2.2) (Qu & Kroeze 2010). Another important reason is the scarcity of the data for the other forms of nutrients. We compared the measured and modeled total annual DIN and DIP exports (yields, kg km^{-2} $year^{-1}$) by the Yellow, Liao (covering approximately 80% of the Bohai Gulf drainage basin), Yangtze, Fuchunjiang (covering approximately 80% of the Yellow Sea drainage basin) and Pearl (covering 85% of the South China Sea drainage basin). The measured annual yields of DIN for Yangtze and Zhujiang, and of DIP for Yangtze, Yellow, Zhujiang, Liao, and Fuchunjiang were provided by Mayorga et al. (2010). The measured annual DIN yields for the Yellow were taken from Dumont et al. (2005); the values for the Fuchunjiang and Liao were obtained from the GEMS/GLORI database (Meybeck & Ragu 1995) in terms of the nitrate and ammonium concentrations ($N\text{-}NO_3$; $N\text{-}NH_4$ in mg L^{-1}). The modeled DIN and DIP yields for the selected river basins were derived from the Global *NEWS*-2 model and refer to the year 2000 (Mayorga et al. 2010).

The Global *NEWS*-2 model for DIN and DIP performed well based on the three indicators. We calculated an $R_P{}^2$ of 0.96, indicating that 96% of the variance in the measured DIN and DIP yields is explained by the Global *NEWS*-2 model for the selected rivers. Our $R_{NSE}{}^2$ value lies within 0-1 (0.42), indicating an acceptable performance by the Global *NEWS*-2 model. The calculated ME for the DIN and DIP export by the rivers is 18%, indicating good performance.

These three indicators have been widely accepted to evaluate model performance, and they have different weaknesses and strengths. For instance, $R_{NSE}{}^2$ is sensitive to extremely high values due to the squared differences (Moriasi et al. 2007). A drawback of $R_P{}^2$ is that only the dispersion is quantified (Krause et al. 2005), rendering it insensitive toward the additive and proportional differences between the measured and modeled values (Legates & McCabe 1999). Our combination of these three indicators builds trust in the model performance for the DIN and DIP. We argue that these results, when combined with the previous validations for the other N and P forms, support the performance of the Global *NEWS*-2 model for Chinese rivers.

2.2.3 ICEP: An Indicator for Coastal Eutrophication Potential

The ICEP is the Indicator for Coastal Eutrophication Potential. This indicator was developed by Billen and Garnier (2007); it reflects the production of non-siliceous phytoplankton (e.g., potentially harmful algae), which are sustained in coastal waters by the N and P discharged by rivers, while accounting for the nutrient requirements for growth (represented by the Redfield ratios). Changes in the nutrient ratios may alter the phytoplankton population, for example, shifting from siliceous phytoplankton to non-siliceous (harmful) phytoplankton. Specifically, excess N and P in the waters relative to Si stimulate the growth of harmful algae that generally occurs with eutrophication.

The ICEP is estimated based on the Redfield ratio of C:N:P:Si=106:16:1:20. Either N-ICEP or P-ICEP (kg C-eq. km^{-2} day^{-1}) is estimated depending upon which nutrient is limiting. We followed the ICEP approach by Garnier et al. (2010), as implemented in Global *NEWS-2* (Strokal & Kroeze 2013):

$$\text{N-ICEP} = [TN_{flx} / (14 \cdot 16) - DSi_{flx} / (28 \cdot 20)] \cdot 106 \cdot 12 \quad N{:}P<16 \text{ (N is limiting)} \qquad (2.2)$$

$$\text{P-ICEP} = [TP_{flx} / 31 - DSi_{flx} / (28 \cdot 20)] \cdot 106 \cdot 12 \quad N{:}P>16 \text{ (P is limiting)} \qquad (2.3)$$

2

TN_{flx}, TN_{flx} and DSi_{flx} are the fluxes of total N (sum of DIN, DON, and PN), total P (sum of DIP, DOP, and PP) and dissolved Si, respectively (kg km^{-2} year^{-1}). Positive ICEP values indicate rivers with the "potential" for coastal eutrophication because they export N and P to coastal waters in excess relative to Si. Negative values indicate a low risk for coastal eutrophication, but they should not be interpreted as zero risk for harmful algal blooms because ICEP reflects the average annual basin values. This indicator does not account for sub-basin and seasonal variations.

Another indicator for potential coastal eutrophication is silica deficiency. A silica deficiency in combination with increased amounts of N and P is a favorable condition for the growth of harmful algae (Billen & Garnier 2007; Garnier et al. 2010). Therefore, we analyzed the ratios between N, P and Si following Turner et al. (2003). We estimated the ratios of the total N to total P (TN:TP), the dissolved Si to total N (DSi:TN) and the dissolved Si to total P (DSi:TP) for the 16 selected rivers using their fluxes at the river mouth (kg km^{-2} year^{-1}) and their atomic weights. The TN fluxes were calculated as the sum of the DIN, DON and PN. The TP fluxes are the sum of the DIP, DOP and PP. The nutrient fluxes were taken from the Global *NEWS-2* model (Section 2.2.2). We compared the TN:TP ratios with the DSi:TN and DSi:TP ratios (see Figure 2.5 in Section 2.3.3). P-limited rivers are identified when their TN:TP ratios exceed a Redfield value of 16 (N:P = 16:1, vertical lines in Figure 2.5), while N-limited rivers are identified when these ratios are below 16. A silica deficiency is then identified based on the TN and TP fluxes. Rivers with a DSi deficiency relative to the TN fluxes are calculated when their DSi:TN ratios are below the Redfield value 1.25 (SI:N = 20:16, horizontal lines in Figure 2.5). A Si deficiency relative to the TP fluxes is calculated when the DSi:TP ratios are below a Redfield value of 20 (Si:P = 20:1, horizontal lines in Figure 2.5).

2.2.4 Scenario analysis

We analyzed the effects of alternative sea-specific management options to reduce future N and P inputs to the Bohai Gulf, Yellow Sea and South China Sea. We developed three alternative scenarios (S1, S2, and S3) that address the management options for agriculture and sewage taking the GO for 2050 as the baseline (Table 2.1).

Alternative Scenario 1 (S1) involves managing synthetic fertilizers and animal manure with greater efficiency (Table 2.1). Several options are available for reducing the N and P inputs from fertilizer use and animal manure excretion to rivers of the Chinese seas (Table 2.1): (i) more efficient synthetic fertilizer and animal manure use in agriculture (e.g., avoiding the overuse of fertilizers by applying them only in the required quantities); (ii) advanced technologies for fertilizer application; (iii) improved management of animal manure collection and storage (Qu & Kroeze 2012); and (iv) decreased human consumption and, consequently, production of animal-derived products (changes in human diets). We assume that these reduction options could lead to a 20 - 40% decrease in the N and P inputs to watersheds from synthetic fertilizers and a 10 - 30% decrease from the excretion of animal manure (Table 2.1). For the Bohai Gulf, lower potential reductions in N and P inputs to watersheds from fertilizers (20%) and manure (10%) were assumed compared to the Yellow Sea (30% for fertilizers, 20% for manure) and the South China Sea (40% for fertilizers, 30% for manure). This difference reflects the relative contributions of agriculture to the nutrient inputs to the coastal waters of the three seas (see Section 2.3.2). The assumed percentages for the nutrient reductions for agriculture seem feasible (Ermolieva et al. 2009; Li et al. 2011a; Vitousek et al. 2009). For example, Ju et al. (2009) showed that fertilizer N inputs can be reduced by approximately 50% without decreasing the grain yield and quality on the North China Plain (partially covering the Bohai Gulf and Yellow Sea).

Alternative Scenario 2 (S2) addresses the sewage management options to reduce N and P in rivers (Table 2.1). We focus on two options: (i) increasing the use of P-free detergents while recycling human waste and (ii) improving the N and P treatment in sewage systems (e.g., using advanced treatments) (Table 2.1). We assume that the first management option could decrease N and P inputs to watersheds from human waste and detergents by 10-40%, depending on the sea region (see Table 2.1).We consider that these reductions are feasible by 2050 because the current Chinese regulations require the use of P-free detergents in watersheds with static waters (e.g., lakes, reservoirs) (Liu & Qiu 2007). The second management option could increase the N and P removal from sewage effluents by 10-40% by 2050, depending on the sea region. These increases are technically feasible considering current technological opportunities in many industrialized countries. The Bohai Gulf exhibits the highest rates for nutrient reduction (40% decrease in nutrient inputs to watersheds from human waste and detergents) and removal (40% increase in nutrient removal during treatment) compared to the Yellow and South China Seas (see Table 2.1) due to the relatively large share of sewage sources in nutrient export (see Section 2.3.2).

Alternative Scenario 3 (S3) combines scenarios S1 and S2 (Table 2.1).

To implement scenarios S1-S3, we ran the Global *NEWS*-2 using modified model parameters for agriculture and sewage. For agriculture, we assumed that lower nitrogen and phosphorus inputs were transferred to land from fertilizer ($WSdif_{fe.N}$, $WSdif_{fe.P}$) and manure ($WSdif_{ma.N}$, $WSdif_{ma.P}$) (see Box 2.1 for abbreviations). For sewage, we used modified nitrogen and phosphorus emissions to watersheds from human excrement ($WShwExc_{N}$, $WShwExc_{P}$), phosphorus emissions from detergents ($WShwDet_{P}$), and nitrogen ($hw_{frem.N}$) and phosphorus ($hw_{frem.P}$) removal fractions in wastewater. We ran the Global *NEWS*-2 model to calculate river export of dissolved inorganic, organic and particulate N and P for each sea region, as well as the ICEP values for 2050.

2

2.3 Results and discussion

2.3.1 Drivers of river export of nutrients

Economic and population growth increased rapidly in the river basins draining into the Bohai Gulf, Yellow Sea and South China Sea between 1970 and 2000 (Table 2.2). The annual gross domestic product (GDP) was approximately seven times higher in 2000 than in 1970 (Table 2.2). The population density increased on average by approximately 50% during this period. These trends may continue in the future. Between 2000 and 2050, the GDP is projected to increase approximately five-fold in the AM scenario and eleven-fold in the GO scenario. The average population densities are projected to increase by approximately 15% in AM and stabilize in GO between 2000 and 2050. The GO scenario assumes a globalized world with global markets and improved human well-being (e.g., better education and sanitation) (Alcamo et al. 2005; Seitzinger et al. 2010), explaining the higher economic and lower population growth. We did not calculate large differences in income rates among the three sea regions. In contrast, the population densities varied considerably among regions (Table 2.2).

Table 2.1. *Options to reduce the nutrient inputs to watersheds of the river basins draining into the Bohai Gulf, Yellow Sea and South China Sea, as assumed in Scenarios S1, S2 and S3.*

Alternative scenario (S)	Reduction option	Assumed changes in the model inputs relative to the GO[(a)] level in 2050		
		The Bohai Gulf	The Yellow Sea	The South China Sea
S1: Agricultural management	Efficient use of synthetic fertilizers and animal manure [(b)]; Advanced technologies for fertilizer applications [(b)]; Improved collection and storage of animal manure [(b)]; Decreased meat consumption and production	Reduced: Synthetic fertilizer use: 20% Animal manure excretion: 10%	Reduced: Synthetic fertilizer use: 30% Animal manure excretion: 20%	Reduced: Synthetic fertilizer use: 40% Animal manure excretion: 30%
S2: Sewage management	Use of P-free detergents and recycling of human waste in sewage	Reduced[(c)]: Human waste: 40% Detergents: 40%	Reduced[(c)]: Human waste: 20% Detergents: 20%	Reduced[(c)]: Human waste: 10% Detergents: 10%
	Improved waste water treatment	Increased[(d)]: Nutrient removal: 40%	Increased[(d)]: Nutrient removal: 20%	Increased[(d)]: Nutrient removal: 10%
S3: Combination	All of the above	All of the above	All of the above	All of the above

(a) GO = Global Orchestration is one of the Millennium Ecosystem Assessment Scenarios as implemented in Global *NEWS*-2 (Seitzinger et al. 2010). **(b)** Based on Qu and Kroeze (2012), who analyzed the effects of these management options on the river export of nutrients in China. **(c)** Assumed reductions in the N and P (as elements) inputs to watersheds from human waste (for N and P) and detergents (for P). **(d)** Assumed increases in the percentage of N and P removal from the sewage effluents through treatment.

Urbanization trends are driving the development of sewage systems (Van Drecht et al. 2009). Between 1970 and 2000, the number of people with a sewage connection increased three-fold in the Bohai Gulf, five-fold in the Yellow Sea and over two-fold in the South China Sea, with a larger percentage of people connected to sewage systems in the Bohai Gulf region (Table 2.2). The total N and P inputs (ton km^{-2}) to rivers from sewage systems were over six times higher in 2000 than in 1970 for the three seas. This is a net effect of a population increasingly connected to sewage systems and low sewage treatment efficiencies (approximately 10% of N and P removal in 2000). In the future, urbanization may continue. Between 2000 and 2050 the number of people connected to sewage systems may continue to increase, in particular in GO. By 2050, approximately two-thirds of the Bohai Gulf population, half of the Yellow Sea population and one-third of the South China Sea population may have access to sewage systems in GO (Table 2.2). For individual river basins these percentages range from 0% to 92% (Table 2.2). Between 2000 and 2050, TN and TP inputs to rivers from human waste and detergents may double (in the South China Sea) to triple (in the Bohai Gulf and Yellow Sea) as a result of an increasing population with sewage connections and trends in sewage treatment. By 2050, N and P removal in sewage treatment is projected to be 40–50% in GO and 20–30% in AM. These are conservative assumptions, however, because removal rates may be higher considering the current progress in technology.

Because of the increasing demand for food, the use of N and P synthetic fertilizers in agriculture has been increasing since the 1970s–1980s (Gong et al. 2012; Guo et al. 2010; Ma et al. 2012; Seitzinger et al. 2010; Sutton et al. 2013). Our results show that between 1970 and 2000, N and P fertilizer use increased by 500% in the Bohai Gulf and South China Sea to almost 600% in the Yellow Sea (Table 2.2). Other studies (Kahrl et al. 2010; Ma et al. 2012; Ma et al. 2013b) also demonstrate a large increase in fertilizer consumption in China beginning in the 1960s. The Bohai Gulf drainage basin covers approximately two-thirds of the agricultural areas (including arable land for crop production and grasslands for livestock production). For the other two seas, agricultural areas covered approximately one-third of their drainage basins in 1970, and by 2000 the areas were expanded over two-fold (Table 2.2). The increase in fertilizer use may be associated with government initiatives (subsidies) (Ma et al. 2012; Sutton et al. 2013). As a result, farmers tend to prefer synthetic fertilizers to animal manure, decreasing the use of manure as a fertilizer between the 1980s and 2005 even though manure excretion increased. Part of the available animal manure is discharged directly to water bodies or landfilled as waste (Li et al. 2011a; Ma et al. 2010; Ma et al. 2012). Our results indicate that TN and TP inputs to land from animal manure have at least doubled since 1970 (Table 2.2). This, however, may be an underestimation because our model only considers animal manure as a diffuse source of N and P in rivers, ignoring the direct inputs as a point source (Bouwman et al. 2009). In the future, nutrient inputs to land from fertilizers (GO) and manure (AM, GO) may further increase for the three seas, whereas agricultural areas may decrease (Table 2.2), indicating the intensification of agricultural practices in the future. For instance, TN and TP inputs to land from animal manure may increase on average 62% for the Bohai Gulf, 52% for the Yellow Sea and 46% for the South China Sea between 2000 and 2050 in AM (Table 2.2).

Climatic (e.g., precipitation and runoff) and hydrological (e.g., dam retention and water consumption) drivers vary among the Bohai Gulf, Yellow Sea and South China Sea. Annual precipitation and runoff in the Bohai Gulf river basins are generally low (29 mm of runoff in 1970) compared to the Yellow Sea (525 mm of runoff in 1970) and South China Sea (585 mm of runoff in 1970) (Table 2.3). The low precipitation and runoff in the Bohai Gulf basins are consistent with the dry conditions in northern China (Zhai et al. 2005). In dry environments, the inputs of N, P and Si from land to rivers are generally low. Between 1970 and 2000, Global *NEWS*-2 simulated increases in the annual precipitation (7%) and runoff (40%) for the Bohai Gulf basins, whereas decreases were simulated for the Yellow Sea and South China Sea basins (5–10%) (Table 2.3). Between 2000 and 2050, the annual precipitation of the Bohai Gulf may further increase (10%), however annual runoff may decrease (approximately 20%) in AM and GO. For the Yellow Sea and South China Sea, a 7–16% increase in annual precipitation and runoff is projected for this period (Table 2.3). Our modeling results are generally in agreement with existing studies (Milliman & Ren 1995; Tang et al. 2013; Yan-Chun 1998), indicating that the basins of the Bohai Gulf are located in drier areas than the basins of the other seas. The annual precipitation is in the range of 300 to 700 mm per year for the Yellow basin (Tang et al. 2013), which is in agreement with our results. Several studies (Fu et al. 2004; Tang et al. 2013; Wang et al. 2007; Wang et al. 2013b; Yan-Chun 1998) indicate a large spatial and temporal variation in runoff and precipitation in the large river basins (e.g., Yellow, Yangtze) from the 1960s to the 1990s. For example, Yan-Chun (1998) showed a slight increase in annual runoff for the Yellow from 1970 to 1989 and a decrease after the 1990s. Fu et al. (2004) indicated increasing trends in runoff of the Yellow during the winter and spring and decreasing trends during the summer. Wang et al. (2013b) reported that large decreases in runoff of the Yangtze occurred after the 1990s in the lower reach due to human activities. In this study, we calculated the annual average runoff and precipitation for the river basins of the Bohai Gulf, Yellow Sea and South China Sea. This explains the differences between our modeling results and the results of other studies.

Table 2.2. *Anthropogenic drivers of the nitrogen and phosphorus inputs to the watersheds of the 16 rivers draining into the Bohai Gulf, Yellow Sea and South China Sea. "n" is the number of river basins. The averaged values of river basins are presented outside of the brackets. The ranges among river basins are presented inside of brackets when available. These data are based on Global NEWS-2 (Mayorga et al. 2010)*

Drivers	Year	Chinese seas		
		The Bohai Gulf (n=6)	The Yellow Sea (n=6)	The South China Sea (n=4)
Income, GDP at purchase power parity (1995 US$ 1000 inh^{-1})**(a)**	1970:	0.5	0.5	0.5
	2000:	3.6	3.6	3.5
	2050 (AM):	19.5	19.4	19.3
	2050 (GO):	38.5	38.4	38.1
Total Population (inh. km^{-2})**(b)**	1970:	131 (91-479)	183 (82-369)	140 (135-180)
	2000:	202 (139-736)	281 (126-566)	216 (207-276)
	2050 (AM):	231 (159-842)	322 (142-648)	249 (239-316)
	2050 (GO):	203 (139-738)	282 (126-568)	218 (209-277)
Population connected to sewage systems (fraction)**(c)**	1970:	0.08 (0-0.13)	0.04 (0-0.05)	0.07 (0-0.08)
	2000:	0.26 (0-0.32)	0.18 (0-0.29)	0.17 (0-0.32)
	2050 (AM):	0.53 (0-0.70)	0.37 (0-0.69)	0.24 (0.23-0.36)
	2050 (GO):	0.67 (0.12-0.92)	0.50 (0-0.88)	0.34 (0.28-0.50)
TN and TP inputs from sewage systems (ton km^{-2})**(d)**	1970:	0.04 (0-0.1)	0.03 (0-0.03)	0.03 (0-0.04)
	2000:	0.2 (0-0.9)	0.2 (0-0.6)	0.2 (0-0.4)
	2050 (AM):	0.7 (0-3.7)	0.7 (0-2.5)	0.3 (0.3-0.6)
	2050 (GO):	0.7 (0.1-3.4)	0.7 (0-2.5)	0.4 (0.3-0.7)
Agricultural area (fraction of basin area)**(e)**	1970:	0.72 (0.30-0.82)	0.35 (0-0.75)	0.26 (0-0.29)
	2000:	0.83 (0.65-0.88)	0.70 (0.27-0.91)	0.70 (0.54-0.89)
	2050 (AM):	0.78 (0.30-0.89)	0.66 (0.18-0.91)	0.67 (0.64-0.98)
	2050 (GO):	0.77 (0.30-0.86)	0.65 (0.05-0.86)	0.60 (0.38-0.61)
TN and TP inputs from fertilizer (ton km^{-2})**(f)**	1970:	0.7 (0.2-1.4)	0.8 (0-2.5)	0.6 (0-1.1)
	2000:	4.2 (2.6-8.6)	5.5 (2.0-16.8)	3.6 (2.8-9.2)
	2050 (AM):	4.0 (3.4-10.5)	5.0 (1.5-12.6)	2.9 (2.5-8.7)
	2050 (GO):	7.3 (4.5-23.5)	8.7 (1.6-21.2)	5.5 (5.1-7.4)
TN and TP inputs from manure (ton km^{-2})**(f)**	1970:	1.3 (0.7-4.6)	1.2 (0-3.1)	1.3 (0-1.5)
	2000:	2.6 (1.9-7.6)	2.9 (0.6-6.9)	2.8 (2.2-5.7)
	2050 (AM):	4.2 (2.1-10.5)	4.4 (1.4-10.2)	4.1 (3.2-6.6)
	2050 (GO):	5.0 (2.2-12.0)	5.3 (0.5-10.8)	5.0 (3.2-5.1)

(a) No range. **(b)** The average population per km^{-2} for the whole region is estimated as follows: the total population of this region (people) divided by the total basin area of this region. **(c)** The average population with a sewage connection (fraction) for each region is estimated as follows: the population of the region that is connected to sewage systems (people) divided by the total population of the region (people). **(d)** The average inputs of TN (total nitrogen) and TP (total phosphorus) to the surface waters for each region is estimated as follows: the sum of TN inputs from human waste (ton $year^{-1}$) and the TP inputs from the human waste and detergents (ton $year^{-1}$) divided by the total area of the region. **(e)** The average agricultural area (fraction) for each region is estimated as follows: the agricultural area (km^2) of the region divided by the total basin area of the region (km^2). **(f)** The average for the region is the sum of the TN and TP inputs from fertilizers to the watersheds (land) of the region (ton $year^{-1}$) divided by the total area of this region. The same holds for the TN and TP inputs to the watersheds from animal manure.

Construction of dams increases nutrient retention within the river systems of the three seas. For the Bohai Gulf, the retention fraction of DIN in rivers is higher (0.20 on average for its river basins) than for the Yellow Sea (0.11 on average for its river basins) and the South China Sea (0.03–0.04 on average for its river basins) in past years (Table 2.3). Approximately one-third of the annual DIP and sediment inputs to rivers of the Bohai Gulf were retained within dammed reservoirs in the past. This is similar for the Yellow Sea. For the South China Sea, approximately 25% of DIP and sediment inputs to rivers were retained within reservoirs. Between 2000 and 2050, nutrient retentions may further increase in particular for the Yellow Sea and the South China Sea under GO. For example, dam retention for DIP may increase by approximately 90% in the Yellow Sea's rivers and by approximately 135% in the South China Sea's rivers between 2000 and 2050. In addition, the fraction of nutrient removal through consumptive water use in the Bohai Gulf is much higher (0.54–0.56) than in the Yellow Sea (0.21–0.27) and South China Sea (0.13–0.18) in the past and projected to be in the future (Table 2.3).

2.3.2 Trends in river export of nutrients

River export of nitrogen and phosphorus is dominated by dissolved inorganic forms (Figure 2.2). For the Bohai Gulf, the share of DIN in the total N in rivers is 50%, that of DON is approximately 40% and of PN is approximately 10%. The DIP accounts for 65% of the total P export to the Bohai Gulf, the DOP accounts for approximately 30%, and the remainder is PP. For the Yellow Sea and South China Sea, the DIN accounts for over two-thirds of the total N river export, whereas the remainder is dissolved organic and particulate N, with DON dominating. The DIP and PP each account for approximately half of the total P river export. The DOP has a minor share (approximately 10%) in the total P export to the Yellow Sea and South China Sea (see Figure 2.2). We calculate, in agreement with Qu and Kroeze (2010), that approximately two-thirds of the total N is exported as DIN and over half of the total P as DIP to the coastal waters of three seas in China. More nitrogen and phosphorus (approximately 70%) is exported by rivers draining into the Yellow Sea compared to rivers draining into the Bohai Gulf (5–10%) and the South China Sea (25–30%). This is in line with the size of the drainage basins of these three seas (see Figure 2.1).

Table 2.3. *Same as Table 2.2 but for climatic and hydrological drivers of nutrient export. These data are based on Global NEWS-2 (Mayorga et al. 2010)*

Drivers	Year	Chinese seas		
		The Bohai Gulf (n=6)	The Yellow Sea (n=6)	The South China Sea (n=4)
Annual precipitation (mm year^{-1})	1970:	484[(a)] (404-562)[(b)]	1301 (795-1807)	1642 (1423-1851)
	2000:	519 (435-696)	1245 (895-1631)	1556 (1420-1773)
	2050 (AM):	561 (484-739)	1335 (990-1743)	1709 (1593-1934)
	2050 (GO):	566 (490-745)	1346 (1001-1757)	1728 (1615-1954)
Annual runoff from land to surface water (mm year^{-1})	1970:	29 (18-41)	525 (65-957)	585 (435-735)
	2000:	40 (22-62)	465 (114-707)	508 (417-650)
	2050 (AM):	32 (20-43)	498 (130-751)	574 (489-714)
	2050 (GO):	33 (23-42)	507 (136-768)	589 (501-728)
Basin-wide dam retention fraction for DIN (0-1)[(c)]	1970:	0.20 (0-0.77)	0.11 (0-0.24)	0.03 (0-0.09)
	2000:	0.20 (0-0.70)	0.11 (0-0.23)	0.04 (0-0.10)
	2050 (AM):	0.26 (0-1)	0.11 (0-0.23)	0.04 (0-0.12)
	2050 (GO):	0.26 (0-1)	0.18 (0.05-0.30)	0.08 (0-0.17)
Basin-wide dam retention fraction for DIP (0-1)[(d)]	1970:	0.31 (0-1)	0.35 (0-0.79)	0.14 (0-0.38)
	2000:	0.33 (0-1)	0.35 (0-0.79)	0.14 (0-0.40)
	2050 (AM):	0.32 (0-1)	0.37 (0-0.78)	0.17 (0-0.51)
	2050 (GO):	0.32 (0-1)	0.68 (0.13-1)	0.33 (0-0.71)
Basin-wide dam retention fraction for sediments (0-1)[(d)]	1970:	0.32 (0-1)	0.41 (0-0.86)	0.23 (0-0.74)
	2000:	0.34 (0-1)	0.42 (0-0.85)	0.24 (0-0.76)
	2050 (AM):	0.33 (0-1)	0.43 (0-0.86)	0.26 (0-0.81)
	2050 (GO):	0.34 (0-1)	0.63 (0-0.90)	0.33 (0-0.86)
Fraction of nutrients removed through consumptive water use (0-1)	1970:	0.54 (0.39-0.76)	0.21 (0.04-0.67)	0.13 (0.11-0.17)
	2000:	0.55 (0.48-0.72)	0.25 (0.04-0.69)	0.19 (0.16-0.25)
	2050 (AM):	0.56 (0.31-0.78)	0.27 (0.05-0.74)	0.19 (0.16-0.26)
	2050 (GO):	0.55 (0.33-0.77)	0.25 (0.04-0.73)	0.18 (0.15-0.24)

(a) Average values for the river basins (e.g., the sum of annual precipitation of the river basins divided by the number of river basins in the region). This holds for all drivers presented in this Table. **(b)** Values represent the range among river basins. **(c)** This value cannot exceed 0.965 in Global *NEWS*-2. **(d)** This value cannot exceed 0.85 in Global *NEWS*-2.

Between 1970 and 2000, the river export of DIN increased five-fold for the Bohai Gulf and by over two-fold for the Yellow Sea and the South China Sea (Figure 2.3). Approximately 25% of the DIN in the Bohai Gulf rivers was from sewage in 2000, and approximately 40% was from agriculture. For the Yellow Sea and South China Sea's rivers, fertilizers and manure are dominant sources of DIN. Our results are generally in agreement with existing studies (Liu et al. 2009b; Liu et al. 2008b; Ning et al. 2010; Tao et al. 2010; Zhang et al. 2004). Liu et al. (2009b) indicated that DIN loads from the Yangtze increased by a factor of approximately 6–10 during the last 30 years due to agriculture. This study also discusses the dominant contribution of agriculture to the DIN in rivers of the South China Sea as well. Ning et al. (2010), Liu et al. (2008b), Tao et al. (2010) and Zhang et al. (2004) show increasing trends in DIN loads for the rivers draining into the Bohai Gulf over the period 1960–2000, resulting from human activities on land. Between 2000 and 2050, the DIN river export to the Bohai Gulf may further increase by approximately one-third and export to the Yellow Sea and South China Sea by approximately 50% in GO. For AM, we calculate a stabilizing DIN export at the level that existed in 2000 to all three seas. For the Bohai Gulf, the relative share of agriculture to the DIN export may decrease, in particular under AM, whereas the relative share of sewage may increase in the future. For the Yellow Sea and South China Sea, we calculate that agriculture is the largest source of riverine DIN (Figure 2.3).

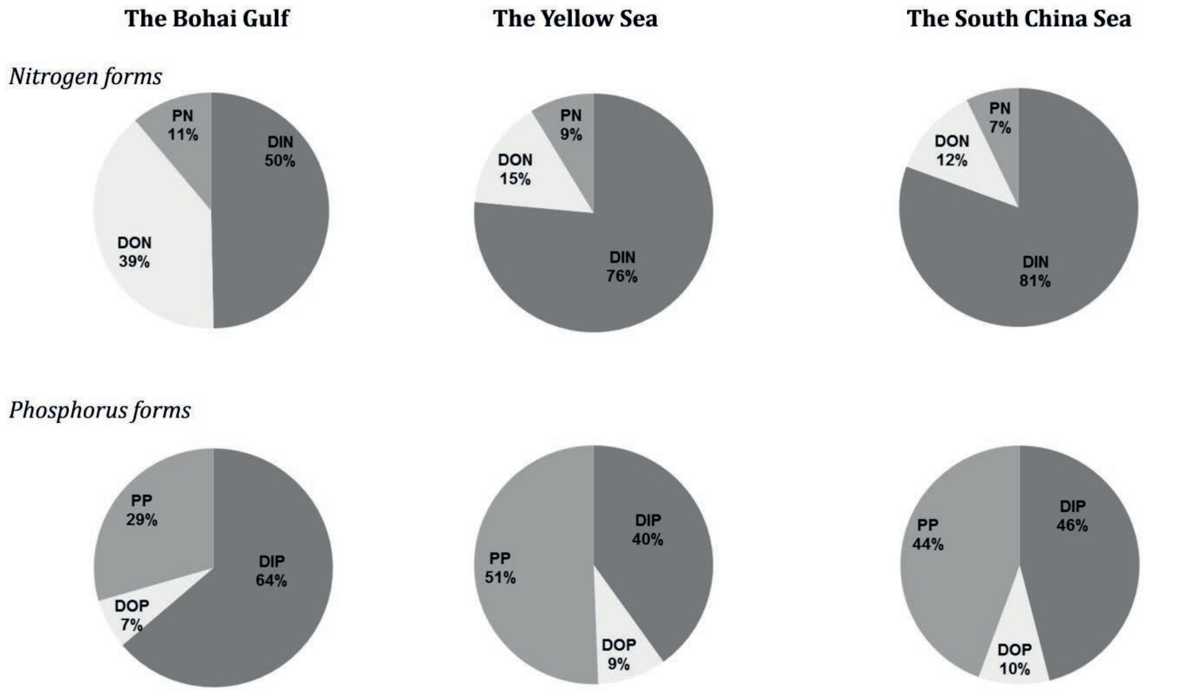

Figure 2.2. *Shares of the dissolved inorganic (DIN, DIP), dissolved organic (DON, DOP) and particulate (PN, PP) nitrogen (N) and phosphorus (P) in the total N and P river exports to the Bohai Gulf, Yellow Sea and South China Sea (%) in 2000. Based on Global NEWS-2 (Mayorga et al. 2010; Seitzinger et al. 2010).*

<u>River export of DIP</u> increased five-fold to the Bohai Gulf, over four-fold to the Yellow Sea and three-fold to the South China Sea between 1970 and 2000 (Figure 2.3). In the future, the DIP export to the three seas may continue to increase. AM projects the largest increases in DIP export to occur between 2000 and 2050, whereas DIP river export triples for the Bohai Gulf and Yellow Sea and doubles for the South China Sea. Between 2030 and 2050, DIP export decreases in GO for the Yellow Sea (by 24%) and South China Sea (by 8%) and stabilizes at this level from 2030 onwards for the Bohai Gulf (Figure 2.3). These decreases can be explained by the assumed improvement in wastewater treatment in GO. The increases in DIP export are associated with sewage sources.

Sewage effluents are calculated to be the only sources of DIP in the rivers of the Bohai Gulf. This is in line with the specific climate and hydrology in this region. The DIP inputs from diffuse sources hardly reach coastal waters as a result of low annual runoff and high P retention within the watersheds and river systems of the Bohai Gulf (see Section 2.3.1). Our results are in agreement with Wang et al. (2007) who reported on a decrease in water discharge and a high retention in reservoirs of the Yellow (the Bohai Gulf region) over the period 1950–2000, which decreases nutrient export from land-based sources to the sea. In contrast, sewage systems and agriculture are major sources of DIP in the rivers of the Yellow Sea and South China Sea (Figure 2.3). Our source contributions are in agreement with Liu et al. (2009b) for the Yangtze basin (Yellow Sea region) and with Huang et al. (2003) for the Pearl River basin (South China Sea region).

In 2000, the DON and DOP export by rivers of the Bohai Gulf was approximately three times and two times higher than in 1970, respectively (Figure 2.3). This is because of increased sewage inputs in 2000 compared to 1970. For the Yellow Sea, a 30% increase in dissolved organic N and P is calculated for this period. For the South China Sea, this increase was approximately 10%. These increases are mainly a result of the increased contribution of sewage sources to the river export of DON and agricultural sources to the river export of DOP between 1970 and 2000. In the future, the river export of dissolved organic N and P may continue to increase, in particular in GO. For the Bohai Gulf, sewage contributes approximately 80% of the calculated future DON inputs and approximately 50% of DOP inputs to the coastal waters. For the Yellow Sea and South China Sea, sewage systems dominate trends in DON export and agriculture (fertilizers and manure) trends in DOP export by rivers.

In contrast to dissolved N and P, the river export of PN and PP to the Bohai Gulf decreased by 20% between 1970 and 2000 (Figure 2.3). Particulate N and P inputs to the Yellow Sea stabilized during this period and increased slightly to the South China Sea. These small changes are the net effect of increased inputs of particulate N and P in rivers as a result of climate change (increased water discharge), land use change, and an increased retention behind dams in the rivers (see Section 2.3.1). Some studies (Dai & Lu 2013; Liu et al. 2012; Wang et al. 2007) indicate decreasing trends in sediment loads from large rivers such as the Yangtze and Yellow to Chinese seas over the last 30 years due to the increased constructions of dams. This is in agreement with our study, where we calculated an increase in nutrient retention in reservoirs (see Section 2.3.1) because of river damming since 1970. Higher nutrient retentions in reservoirs lead to lower nutrient export. In the future, the river export of particulate nutrients may slightly increase or decrease depending on the scenario and region (changes of <10%) (Figure 2.3). In the GO scenario more dam construction is assumed than in AM (Seitzinger et al.

2010). Therefore, we calculated a lower river export of particulate N and P in GO than in AM.

River export of DSi shows similar trends as the particulate N and P (Figure 2.4). Between 1970 and 2000, the DSi export by rivers decreased by approximately 5%. This decrease is associated with the damming of rivers since 1970. Decreasing trends in DSi loads to the seas were also reported in various studies (Chai et al. 2006; Huang et al. 2003; Liu et al. 2009b; Liu et al. 2008b). Future trends depend on the scenario and region. DSi export to the Bohai Gulf is calculated to increase slightly between 2000 and 2050 (Figure 2.4). DSi export to the Yellow Sea is projected to increase (approximately 5%) in AM and decrease (approximately 15%) in GO between 2000 and 2050. Decreasing trends in DSi export are calculated for the South China Sea's rivers in GO and a stabilization in AM. Differences in DSi trends among regions and years can be explained by changes in hydrology associated with damming and climate change.

2.3.3 Potential coastal eutrophication

Next, we calculate whether rivers of the Bohai Gulf, Yellow Sea and South China Sea are N or P limited (Figure 2.5). In 1970 and 2000, rivers of the Bohai Gulf were N limited (TN:TP<16) except for the Yellow (1970, 2000) and the Dalinghe (2000). Rivers of the Yellow Sea and South China Sea were P limited (TN:TP>16) except for the Fuchunjiang (1970), the Huai River of the Yellow Sea and the Jiulong He of the South China Sea (1970). Our results are comparable with other studies. For example, Liu et al. (2012) and Liu et al. (2008b) also calculated P limitation for the Yellow. Huang et al. (2003) and Yin et al. (2004) indicated P limitation for the Pearl River (the largest river in the South China Sea region). They concluded that P limitation is one of the factors causing high concentrations of chlorophyll a in the Pearl River estuary. For the Yellow Sea, Wang (2006) indicated an increasing trend in N:P ratios for the Yangtze, indicating P limitation. Similar conclusions were drawn by Chai et al. (2006) for the Yangtze. In the future, the rivers of the Bohai Gulf are projected to remain N limited in GO and AM. For the Yellow Sea, N limitation is calculated for the Huai and Fuchunjiang rivers and P limitation is calculated for the Yangtze, Menjiang, Yalu and Qujiang (except for AM) in both scenarios. Rivers of the South China Sea will stay P limited except for the Jiulong He (Figure 2.5). These differences in N and P limitations among rivers are associated with dominant sources of nutrients in rivers. For instance, the P-limited rivers of the South China Sea receive more N from agricultural sources than P from sewage and as a result their TN:TP ratios exceed the Redfield value of 16 (see Section 2.3.2).

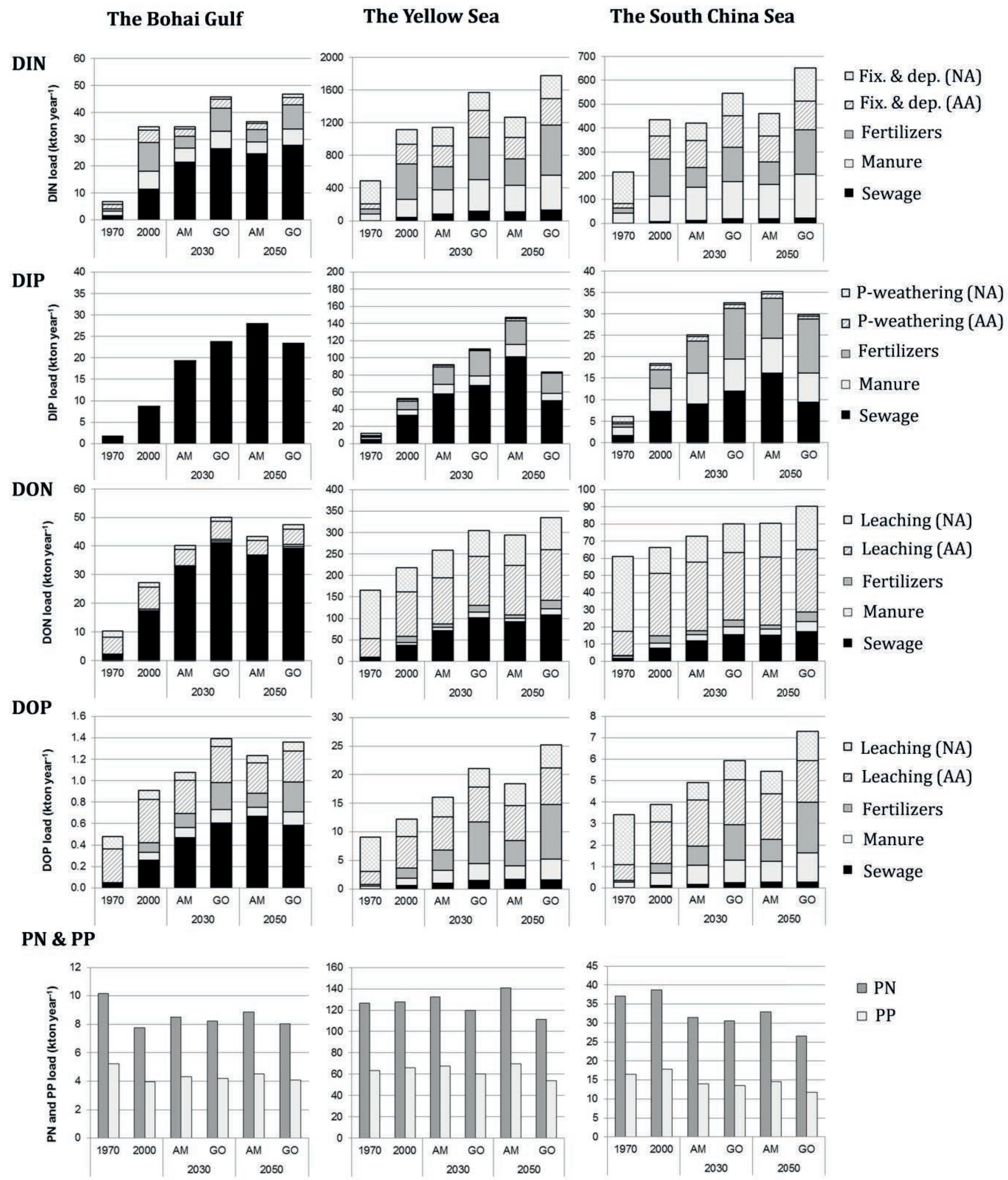

Figure 2.3. *Modeled export of the dissolved inorganic (DIN, DIP), dissolved organic (DON, DOP) and particulate (PN, PP) nitrogen (N) and phosphorus (P) by the 16 rivers draining into the three Chinese seas: the Bohai Gulf, the Yellow Sea and the South China Sea during past (1970, 2000) and future (2030, 2050) years. GO and AM are the Global Orchestration and Adapting Mosaic scenarios from the Millennium Ecosystem Assessment, respectively. The river export of dissolved nutrients is presented by source. NA and AA are non-agricultural and agricultural areas, respectively. Fix is nitrogen N fixation, and dep is deposition. These charts are based on Global NEWS-2 (Mayorga et al. 2010; Seitzinger et al. 2010).*

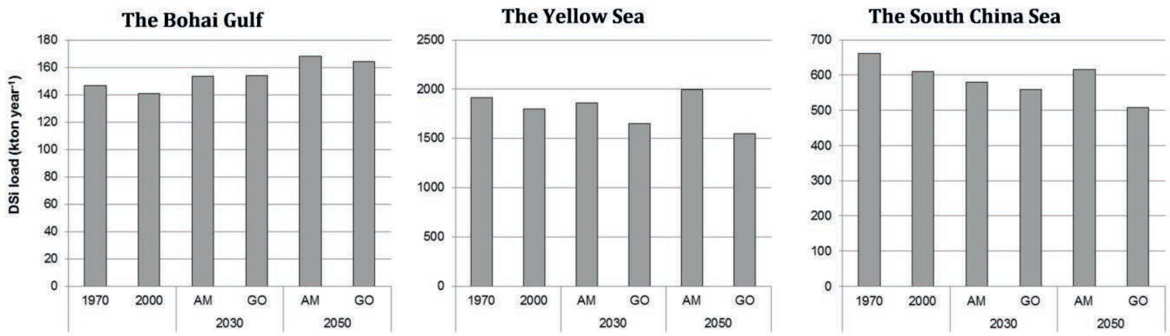

Figure 2.4. Modeled export of the dissolved silica (DSi) by the 16 rivers draining into the three Chinese seas: the Bohai Gulf, Yellow Sea and South China Sea during past (1970, 2000) and future (2030, 2050) years. GO and AM are the Global Orchestration and Adapting Mosaic scenarios of the Millennium Ecosystem Assessment, respectively. These charts are based on Global NEWS-2 (Mayorga et al. 2010; Seitzinger et al. 2010).

Our results indicate that in most rivers there was no shortage of DSi over TN and TP fluxes in the past. However, this may change in the future (Figure 2.5). In 1970, DSi:TN ratios were below the Redfield value of 1.25 for the P-limited rivers Yangtze, Menjiang and Yalu of the Yellow Sea and the Zhujiang of the South China Sea (left column of Figure 2.5), indicating an excess of TN over DSi. DSi:TP ratios are below the Redfield value of 20 for the N-limited rivers, such as the Hai and Xiaoqinghe of the Bohai Gulf (right column of Figure 2.5), indicating that DSi levels are too low to maintain growth of siliceous phytoplankton sustained by TP fluxes. Between 1970 and 2000, the number of river basins with DSi deficiencies over TN and TP fluxes increased (Figure 2.5). For the future scenarios (2030 and 2050), DSi:TN and DSi:TP ratios decrease for almost all river basins. For some small rivers, DSi fluxes are in excess over TN and TP inputs. These rivers are, for example, the Luan of the Bohai Gulf (in GO, AM) and the Oujiang of the Yellow Sea (in AM and in GO with respect to TP fluxes) (see Figure 2.5). Decreases in DSi:TN and DSi:TP ratios are associated with increased DSi retentions in the river system primarily due to the construction of dams (Billen & Garnier 2007). For example, the Yellow of the Bohai Gulf is a highly dam-regulated basin (see Table 2.3 of Section 2.3.1) (Liu et al. 2012). This decreases DSi export to coastal waters. Liu et al. (2009b) reviewed literature regarding ratios of nutrients and their effects on phytoplankton compositions in estuaries of the Bohai Gulf, Yellow Sea and South China Sea. They reported decreasing trends in ratios of DSi to DIN and DIP for the three seas. This is in agreement with our findings. They also indicate that changes in the nutrient ratios over time have caused a shift from siliceous phytoplankton to non-siliceous phytoplankton harmful species (e.g., dinoflagellates and cyanophytes) in estuaries.

<u>The ICEP indicator</u> summarizes the aforementioned results for N and P limitations and DSi deficiencies (Figure 2.6). For 1970, negative ICEP values (ICEP<0) were estimated for all river basins except for the largest river of the Bohai Gulf (Yellow), indicating a relatively low risk for growth of non-siliceous harmful phytoplankton (e.g., cyanobacteria). The Yellow hardly exports DSi fluxes to its coastal zones because of dam retention and relatively low runoff from land to surface waters (Section 2.3.1, Table 2.3). As a result, TN and TP fluxes are in excess over DSi fluxes, indicating a potential for harmful algal blooms. We calculated positive ICEP values (ICEP>0) for 2000 for the Yellow and Hai, draining into the Bohai Gulf, as well as for many rivers of the Yellow Sea and South China Sea (Figure 2.6). In general, our results correspond with the findings of Sutton et al. (2013) and Wang et al. (2008). Sutton et al. (2013) reported intensive growth of harmful algae in the Taihu Lake (located close to the Yellow Sea's coastal waters). Wang et al. (2008) reported increases in the distribution of harmful algal blooms along the coastal zones of the Pearl River (the Zhujiang and Dongjiang of the South China Sea) between 1980 and 2001. Xiao et al. (2007) indicated that the number of harmful algal blooms increased from approximately five events in 1972 to approximately 120 events in 2004 in Chinese seas. A majority of these events occurred along the coastal waters of the South China Sea and Yellow Sea. Diaz and Rosenberg (2008) also reported many eutrophic and hypoxic areas along the coastal waters of Chinese seas.

For future years (2030, 2050) we calculated positive ICEP values for most river basins in GO and AM (Figure 2.6). Rivers covering approximately 80% of the Bohai Gulf basin are associated with eutrophication risk (ICEP>0) in 2030 and 2050 (AM, GO). For the Yellow Sea and South China Sea, this percentage is 90% (Figure 2.6). Our findings are, generally, in agreement with Garnier et al. (2010), who concluded that the potential for coastal eutrophication may increase in the North and South Pacific Ocean in the future. Thus, the potentials for coastal eutrophication in Asia may be increasing unless environmental policies are implemented as in many European countries (Garnier et al. 2010). We calculated differences between the AM and GO scenario. For instance, the Dongjiang of the South China Sea, the Dalinghe of the Bohai Gulf, and the Menjiang and Fuchunjiang of the Yellow Sea are projected to have negative ICEP values in AM and positive ICEP values in GO (Figure 2.6).

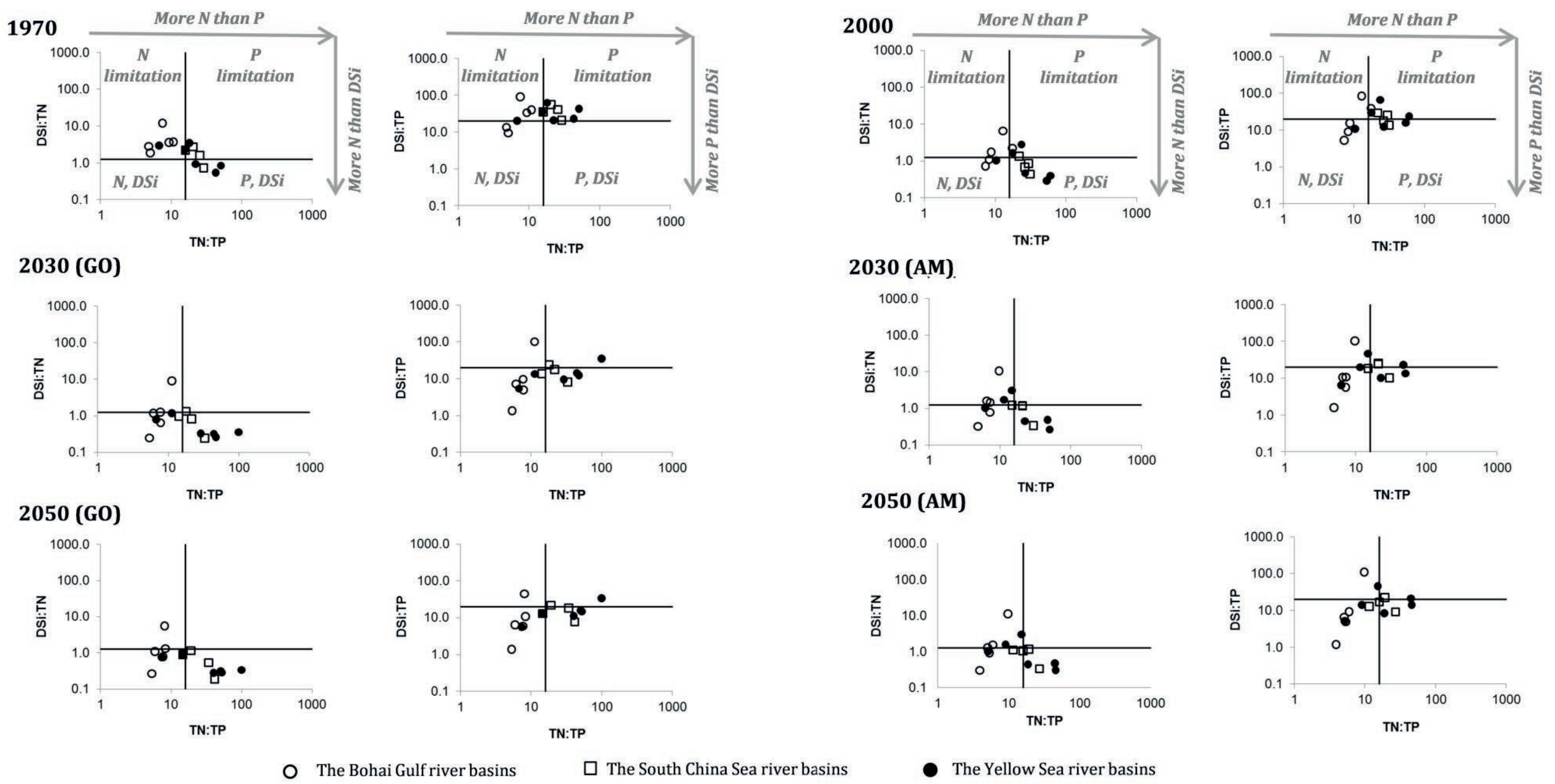

Figure 2.5. *The molar ratios (log scale) of the dissolved silica to the total nitrogen (DSi:TN) and the total nitrogen to the total phosphorus (TN:TP) on the left-hand side, and the dissolved silica to the total phosphorus (DSi:TP) and the total nitrogen to the total phosphorus (TN:TP) on the right-hand side for the 16 rivers draining into the Bohai Gulf, Yellow Sea and South China Sea during past (1970, 2000) and future (2030, 2050) years. GO and AM are the Global Orchestration and Adapting Mosaic scenarios of the Millennium Ecosystem Assessment, respectively. The Redfield ratios for Si:N (20:16=1.25, mol:mol) and Si:P (20:1, mol:mol) are indicated by horizontal lines, and N:P (16:1, mol:mol) is marked by vertical lines, following Turner et al. (2003) (see Section 2.2.3 for a full description). These charts are based on Global NEWS-2 (Mayorga et al. 2010; Seitzinger et al. 2010).*

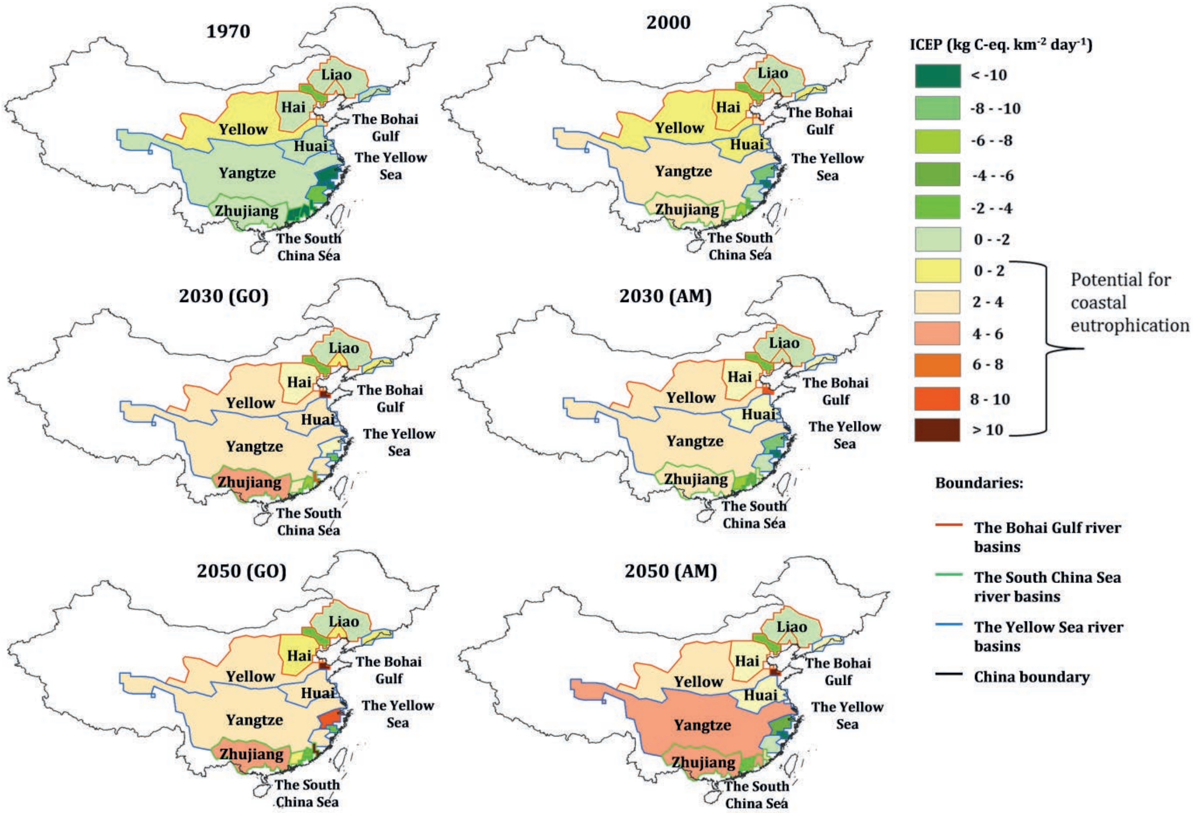

Figure 2.6. *Calculated ICEP values (kg C-eq. km^{-2} day^{-1}) for the river basins draining into the Bohai Gulf, Yellow Sea and South China Sea in China during past (1970, 2000) and future (2030, 2050) years. GO and AM are the Global Orchestration and Adapting Mosaic scenarios of the Millennium Ecosystem Assessment, respectively. These data are based on Global NEWS-2 (Mayorga et al. 2010; Seitzinger et al. 2010).*

2.3.4 Management of future eutrophication

We demonstrated in previous sections that river export of N and P is projected to further increase in the coming years, whereas river export of Si may stabilize or decrease (slight increase for the Bohai Gulf rivers). As a result, eutrophication may be increasingly observed along the coastal waters of China. We explored possibilities to reduce nutrient inputs from human activities in the future (2050). We formulated sea-specific management options to reduce nutrients in Chinese rivers (see Table 2.1). The options are combined in three alternative scenarios (S1, S2, S3), using GO 2050 as a basis. The scenarios address agricultural management (S1), sewage management (S2) and a combination of both options (S3) (see Table 2.1, Section 2.2.4).

Our results indicate that management options to reduce TN and TP export are more effective in some regions than in others (Figure 2.7). Sewage management (S2) seems most effective at reducing TN and TP inputs to the coastal waters of the Bohai Gulf between 2000 and 2050 (Figure 2.7). River export of TN and TP increases by approximately 20–50% during this period in S2 instead of 70–200% in GO. However, variations among river basins are large. For instance, we calculated that the Xiaoqinghe exports considerable amounts of TN and the Dalinghe exports considerable amounts of TP in 2050. For the Yellow Sea, river export of TN and TP increases 3–5% in S3 instead of 30% in GO between 2000 and 2050 (Figure 2.7). For TN export, the difference between S1 (agricultural management) and S3 (combination of agriculture and sewage) is small (<5%), indicating the usefulness of applying agricultural management options (S1) to reduce TN inputs to the sea. For TP export, this difference is 15%, indicating that management of not only agriculture but also sewage is effective in reducing TP export to the sea. For the South China Sea, river export of TN stabilizes from 2000 onwards in S1 instead of a 30% increase in GO (Figure 2.7). TP export to the sea increases by approximately 15% in S1 instead of 50% in GO between 2000 and 2050. For this region, the reduction effects between S1 and S3 on TN and TP export are comparable, showing the effectiveness of agricultural management options (S1). Variations among river basins of the Yellow Sea and South China Sea are not as large as for river basins of the Bohai Gulf (Figure 2.7).

Reducing TN and TP inputs from human activities may decrease the potential for coastal eutrophication, but not for all river basins (Figure 2.7). In general, ICEP values are lower when assuming increased removal of N and P during treatment (S2) for the Bohai Gulf and reduced N and P inputs to lands from agriculture (S1) for the Yellow Sea and South China Sea. For some individual river basins, we calculated negative instead of positive values of ICEP as a result of nutrient management. For the Bohai Gulf, these were for the Dalinghe and Hai river basins in S2 and S3. For the Yellow Sea, this was the Menjiang basin in S3. For the South China Sea, these included the Hanjiang and Dongjinag basins in S1 and S3.

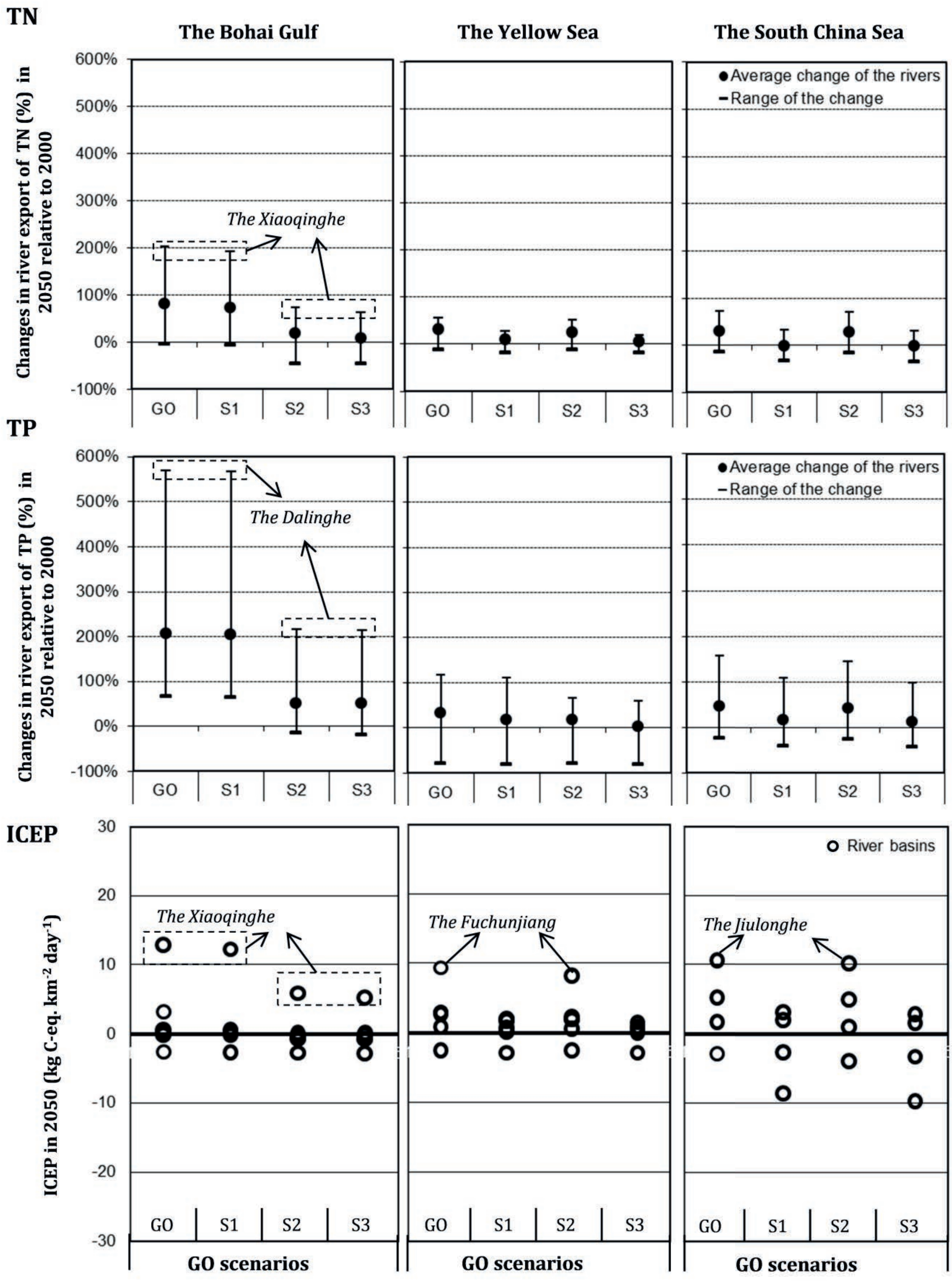

__Figure 2.7.__ Changes (%) in the total nitrogen (TN=DIN+DON+PP) and total phosphorus (TP=DIP+DOP+PP) river exports (loads, ton year^{-1}) between 2000 and 2050 and the ICEP (Indicator for coastal eutrophication potential, kg C-eq. km^{-2} day^{-1}) calculated for the rivers draining into the Bohai Gulf, Yellow Sea and South China Sea. The Global Orchestration (GO) scenario is the baseline. S1, S2 and S3 are three alternative scenarios developed relative to GO (see Table 2.1 for scenario descriptions).

2

We also explored the effects of increased urbanization on nutrient export by selected rivers of the Bohai Gulf, Yellow Sea and South China Sea. We developed two hypothetical alternative scenarios relative to AM (baseline) as an illustrative example of how urban management may influence coastal eutrophication in a regionalized future. Alternative scenario 1 (Alt-1) assumes that at least 50% of the population is connected to sewage systems, and at least 80% of N and P are removed by sewage treatment. Alternative scenario 2 (Alt-2) assumes that at least 70% of the population has a sewage connection and at least 50% of N and P are removed during treatment. The potential for coastal eutrophication is lower in 2050 AM (Alt-1) than in the baseline AM scenario (Figure 2.8). On the other hand, it is higher in the 2050 AM (Alt-2) scenario. These results indicate that reducing or stabilizing nutrient loads will be difficult without managing the speed of urbanization and improving nutrient removal during treatment in the future. Fast urbanization without appropriate urban management may enhance point source inputs of N and P to rivers and, as a result, coastal eutrophication. For instance, we calculated high potentials for eutrophication in the coastal waters of the Yangtze (the Yellow Sea) and Zhujiang (the South China Sea) (ICEP>4 kg C-eq. km^{-2} day^{-1}) under AM, Alt-1 and Alt-2 (Figure 2.8).

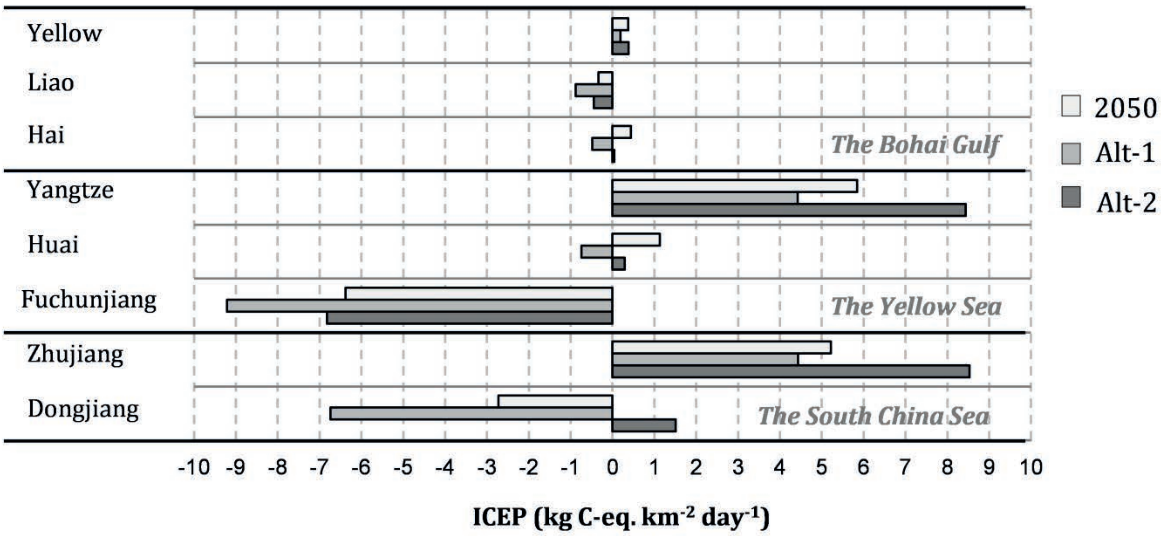

Figure 2.8. *ICEP (Indicator for Coastal Eutrophication Potential) values for the individual rivers draining into the Bohai Gulf, Yellow Sea and South China Sea in 2050 (kg C-eq. km^{-2} day^{-1}). The Adapting Mosaic (AM) scenario of the Millennium Ecosystem Assessment is used as the baseline (2050). Alternative scenario 1 (Alt-1) assumes that at least 50% of the population is connected to sewage systems and that at least 80% of nutrients are removed during sewage treatment. Alternative scenario 2 (Alt-2) assumes that at least 70% of population is connected to sewage systems and that at least 50% of the nutrients are removed during treatment.*

2.4 Conclusion

We analyzed past and future trends of potential eutrophication in the Bohai Gulf, Yellow Sea and South China Sea as influenced by river export of N, P and Si. We used the Global *NEWS*-2 model to analyze the river export of nutrients, i.e., inputs to coastal seas, and their main sources. We applied the ICEP approach to evaluate the associated eutrophication potentials. We used Global Orchestration (GO) and Adapting Mosaic (AM) scenarios of the Millennium Ecosystems Assessment for future analyses.

Coastal eutrophication in the Bohai Gulf is increasing fast because of nutrient inputs from sewage. The Bohai Gulf region includes six river basins covering approximately one-third of the study area. The rivers are N limited except for the Yellow and Dalinghe. The N and P river export from approximately two-thirds of the region (e.g., the Yellow and Haiho) that drain into the coastal waters contributed to the threats to coastal waters in 2000 (ICEP>0). This is because dissolved N and P inputs to the sea increased by a factor of two to five, whereas dissolved Si inputs decreased by approximately 6% between 1970 and 2000. The increasing inputs of N and P are primarily from sewage systems. Agriculture is a relatively small source of N and P in these rivers because of high nutrient retention within the region's watersheds. The decreasing inputs of dissolved Si resulted from high nutrient retention in dammed reservoirs. The particulate N and P decreased by approximately 20% between 1970 and 2000 primarily due to damming. In the future, the ratio of dissolved Si to total N and total P may decrease for many rivers of the Bohai Gulf, indicating low Si levels to sustain phytoplankton growth, and thus increasing the risk for harmful algal blooms. This risk may be reduced by increasing N and P removal in sewage treatment. For example, with an approximate 40% increase in nutrient removal during wastewater treatment relative to GO limits, the calculated increase in total N and P inputs to the Bohai Gulf is 20–50% instead of 70–200% in GO between 2000 and 2050.

The coastal waters of the Yellow Sea were not largely eutrophied in 1970 (ICEP<0). This is different for 2000 and future years, for which we calculated increasing nutrient inputs from agriculture and sewage. The Yellow Sea region consists of six river basins covering more than half of the study area. Rivers covering approximately half of the region are P limited and the others are N limited. We calculated positive values of ICEP for large rivers (e.g., Yangtze, Huai) indicating a potential for harmful algal blooms; the primary reason is riverine inputs of dissolved inorganic N and P to coastal seas, which increased by a factor of two to three between 1970 and 2000. We calculated minor decreases in riverine inputs for dissolved Si and no considerable changes in particulate N and P during these past decades. Agricultural inputs explain the increasing levels of DIN and DOP in rivers and sewage inputs of DIP and DON. From 2000 onwards, ratios of DSi to total N and P decrease for almost all rivers indicating DSi shortages to maintain growth of siliceous phytoplankton under increased dissolved N and P levels (except for DIN in AM). This implies an increasing risk for harmful algal blooms. It may be possible to reduce this risk through additional management in agriculture and sewage treatment that would reduce future N and P inputs to the Yellow Sea. Our scenarios indicate that such management could limit the increase in riverine inputs of TN and TP to the sea to only 3–5% between 2000 and 2050 instead of 30% in GO. The effect of agricultural management was found to exceed that of improved sewage treatment.

The South China Sea suffered from coastal eutrophication in 2000. Additionally, for the future, we project an increasing risk for harmful algal blooms. This is primarily caused by agriculture. This region includes four river basins that cover approximately 10% of the study area. Most rivers (e.g., the Zhujiang and Dongjiang) are P limited. The eutrophication in the past (ICEP>0 for large rivers) is largely associated with a doubling to tripling of dissolved N and P inputs to coastal seas between 1970 and 2000. Agriculture is the main reason for these increases. Sewage systems also contribute to the DIP and DON in rivers. We calculated small changes in riverine inputs of particulate N and P and DSi between 1970 and 2000. Between 2000 and 2050 the risk for harmful algal blooms may increase under GO and AM. This is indicated by decreased ratios of DSi to total N and P for many rivers as a response to increased riverine inputs of dissolved N and P and decreased riverine inputs of dissolved Si. Reducing nutrient inputs to rivers from agriculture may help to reduce the risk for further eutrophication in the coming years. In our alternative scenarios, the total N input to the sea stabilizes from 2000 onwards, instead of increasing by 30% in GO, whereas total P inputs increase by approximately 20% instead of 50%. These reduced growth rates result from assumed improved management in agriculture relative to the baseline GO.

Our study illustrates the usefulness of applying integrated models such as Global *NEWS*-2 to analyze trends in eutrophication and the subsequent causes for the Bohai Gulf, Yellow Sea and South China Sea. Our modeling study may support the formulation of specific measures to reduce future eutrophication in Chinese seas. We show that better sewage treatment is the most effective way to reduce the eutrophication risk in the Bohai Gulf and efficient agricultural practices is most effective in the other seas.

Acknowledgments

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Chapter 3.

Increasing dissolved nitrogen and phosphorus export by the Pearl River (Zhujiang): a modeling approach at the sub-basin scale to assess effective nutrient management

3

Abstract

The Pearl River (Zhujiang in Chinese) has been exporting excess of nitrogen (N) and phosphorus (P), causing eutrophication in the coastal waters of southern China for decades. However, sources of these nutrients and their locations are not well studied for the Pearl River basin. As a consequence, it is difficult to formulate effective management options to reduce these nutrients in the river and to prevent further eutrophication. We developed a sub-basin model based onto the Global *NEWS*-2 (Nutrient Export from WaterSheds) model for the period 1970-2050 to analyze trends in dissolved inorganic N and P (DIN and DIP) and to identify the main sources of these nutrients and their locations. We validated our model by comparing modeled nutrient fluxes with observed. Future analyses are based on Millennium Ecosystem Assessment scenario that assumes a globalized world with a reactive environmental management. DIN and DIP inputs to the coastal waters are calculated to increase by a factor of 2-2.5 between 1970 and 2050. Over two-thirds of the DIN and DIP inputs to the coastal waters stem from two downstream basins (Zhujiang delta and Dongjiang), where agriculture and sewage are important drivers of this increase. Agriculture accounts for over 40% of DIN inputs to coastal waters. Sewage and agriculture account for over 90% of DIP inputs. Thus nutrient management in agriculture and sewage in downstream areas is more effective in reducing coastal eutrophication than nutrient management in up- and middlestream areas of the Pearl River basin.

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3.1 Introduction

China is a country with a rapidly growing population and economy (Hou et al. 2013; Liu et al. 2012; Ma 2012; Ma et al. 2013b; Vitousek et al. 2009; Zhou et al. 2012). An unwanted side-effect of this is increased environmental pollution. For instance, coastal eutrophication problems with harmful algal blooms have been observed since the 1970s in Chinese seas (Diaz & Rosenberg 2008; Liu & Qiu 2007; Selman et al. 2008; Wang et al. 2008). This is a result of increased nitrogen (N) and phosphorus (P) inputs to rivers from human activities on the land such as agriculture, industry, and sewage (Aregay & Minjuan 2012; Ermolieva et al. 2009; Maimaitiming et al. 2013; Qu & Kroeze 2010; Qu & Kroeze 2012; Vitousek et al. 2009).

In this study we focus on the Pearl River basin, which is the third largest river in China (around 440 thousands km^2; Figure 3.1). The drainage basin of the river covers the Yunnan, Guizhou, Guangxi and Guangdong provinces. It serves as a major water supplier for human consumption, agriculture (e.g., irrigation), hydropower generation and navigation in southern China (Cui et al. 2007; Weng 2007). However, the Pearl River estuary has been eutrophic since the 1970s leading to harmful algal blooms, and environmental and economic losses (Huang et al. 2003; Wang et al. 2008). The contribution of human activities (e.g., agriculture and urbanization) to eutrophication may increase in the coming years because of a rapidly growing population and economy (Hou et al. 2013; Liu et al. 2012; Ma 2012; Ma et al. 2013b; Vitousek et al. 2009; Zhou et al. 2012).

The sources of nutrients and their locations in the basin have not been studied to a large extent because of the large basin size that complicates a quantitative assessment. This holds for both empirical and model-based studies. Empirical studies are scarce and often limited to concentration measurements at specific locations, for selected nutrient forms at specific times (Chen et al. 2008b; Huang et al. 2003; Liu et al. 2009b; Yan et al. 1999; Yan et al. 2011; Zhang 2002; Zhang et al. 1999). Modeling studies (Alexander et al. 2002b; Kroeze et al. 2013; Ongley et al. 2010) exist but their spatial resolution is often ether too coarse (Mayorga et al. 2010; Ti et al. 2012) or too detailed (Arnold et al. 1998; Schwarz et al. 2006; Smith et al. 1997) for such large data-poor basins. Examples are the SWAT (Soil and Water Assessment Tool) model and a coupled physical–biological model applied to the Dongjiang (Wu & Chen 2013; Zhou et al. 2012) and to the Pearl River delta (Hu & Li 2009), respectively. An advantage of the SWAT model is that it can simulate nutrients in surface waters and their sources (e.g., agriculture) on a monthly basis for small watersheds, making it possible to analyze spatial and seasonal variability of nutrient fluxes (Gassman et al. 2007). Implementing this model to the entire Pearl River basin, however, requires detailed input data (e.g., daily hydrology, soil properties of different depths), which are often not available in good quality for such large basins. A coupled physical–biological model of Hu and Li (2009) quantifies nutrient fluxes and their transformations, but only for the Pearl River delta and only for the contemporary period.

Global *NEWS*-2 (Nutrient Export from WaterSheds) (Mayorga et al. 2010; Seitzinger et al. 2010) is a global spatially explicit basin scale model. It quantifies past (1970, 2000) and future (2030, 2050) trends in river export of nutrients and their sources (e.g., agriculture, sewage). The model has been applied at the global scale (Mayorga et al. 2010) and at regional scales such as the Black Sea (Strokal & Kroeze 2013; Strokal et al. 2014c), the Bay of Bengal (Sattar et al. 2014; Zinia & Kroeze 2015), Indonesia (Suwarno et al. 2013; Suwarno et al. 2014a), Africa (Yasin et al. 2010). The model was applied to the Chinese rivers including the Pearl River (Qu & Kroeze 2010; Qu & Kroeze 2012; Strokal et al. 2014b) and the Yangtze (Yan et al. 2010). The main strength of this model is to analyze causes of coastal eutrophication in large data-poor regions such as the Pearl River basin (see Section 3.2 for more details about the model). However, for rivers as large as the Pearl modeling at the basin scale may not help us in prioritizing pollution management strategies at the provincial or county level to manage coastal eutrophication in an efficient way. The Pearl River Water Resources Commission stresses the need to perform sub-basin evaluation of water resources (Cui et al. 2007). So far, sub-basin studies do not exist for the entire Pearl basin.

Our main research objective is to quantify trends in dissolved inorganic N (DIN) and P (DIP) export by the Pearl River to coastal waters by source, and to identify locations of the nutrient sources. These nutrients are the most reactive forms of N and P, and thus important in coastal eutrophication (Dumont et al. 2005; Garnier et al. 2010). To this end, we applied Global *NEWS*-2 at the sub-basin scale for the period 1970-2050. In the following, we describe the sub-basin scale modeling approach applied to Global *NEWS*-2 (Section 3.2), present model results on trends in DIN and DIP export by the Pearl River and their sources (Section 3.3), and conclude main findings (Section 3.4). Our study provides new insights in effective sub-basin management to reduce eutrophication in aquatic systems.

3.2 Methodology

3.2.1 A sub-basin scale modeling approach for the Pearl River

We developed a sub-basin scale approach to quantify dissolved inorganic N and P export by the Pearl River to the coastal waters by source during 1970-2050 (Figure 3.2). We applied this approach to the Global *NEWS*-2 model. The original Global *NEWS*-2 model is described by Mayorga et al. (2010). Below we briefly describe the sub-basin scale approach for DIN and DIP export by the Pearl River. First, we identified sub-basins of the Pearl River and then we quantified nutrient export at the sub-basin scale.

We identified six sub-basins: Yujiang, Liujiang, Xijiang, (forming the West River basin), Beijiang, Dongjiang and Zhujiang delta (Figure 3.1a,b). Their names refer to the names of the rivers at their outlets. The Beijiang, Dongjiang and Zhujiang delta sub-basins are defined on the basis of earlier studies (Cui et al. 2007; Niu & Chen 2010; Weng 2007; Zhang et al. 2007). We added to these the Yujiang and Liujiang rivers, which are two main tributaries of the Xijiang river (the West River) according to Niu and Chen (2010), Cui et al. (2007) and Zhang et al. (2007). We classify these six sub-basins into upstream (Liujiang and Yujiang), middlestream (Xijiang and Beijiang) and downstream (Zhujiang delta and Dongjiang), which is needed for further calculations (Figure 3.2).

Our sub-basin modeling approach for the Pearl River (Figure 3.2) allows for analyzing the contribution of sources at sub-basin scale to DIN and DIP inputs to the coastal waters, for past and future years. This has not been done before for the Pearl River. DIN and DIP export by the Pearl River is modeled as a function of N and P inputs to surface waters in each sub-basin. We account for diffuse and point sources (see below for a description), corrected for nutrient retention on land. Some of these nutrients are lost (e.g., N denitrification, water withdrawal for irrigation) or/and retained (e.g., N and P sedimentation in reservoirs) in surface waters before reaching the outlets of the sub-basins (Figure 3.2). The amount of nutrients exported to the coastal waters of the Pearl River from each sub-basin outlet is determined by transport of nutrients through river systems in the more downstream sub-basins. Thus DIN and DIP exported to the outlet of an upstream sub-basin is either transported to the coastal waters by the rivers in the middle- and downstream sub-basins, or lost or/and retained before reaching thdowne coastal waters. Likewise, middlestream sub-basins transport nutrients that are not lost or retained to more downstream sub-basins. The downstream sub-basins include the mouth of the Pearl River, and discharge into the coastal waters (Figure 3.2).

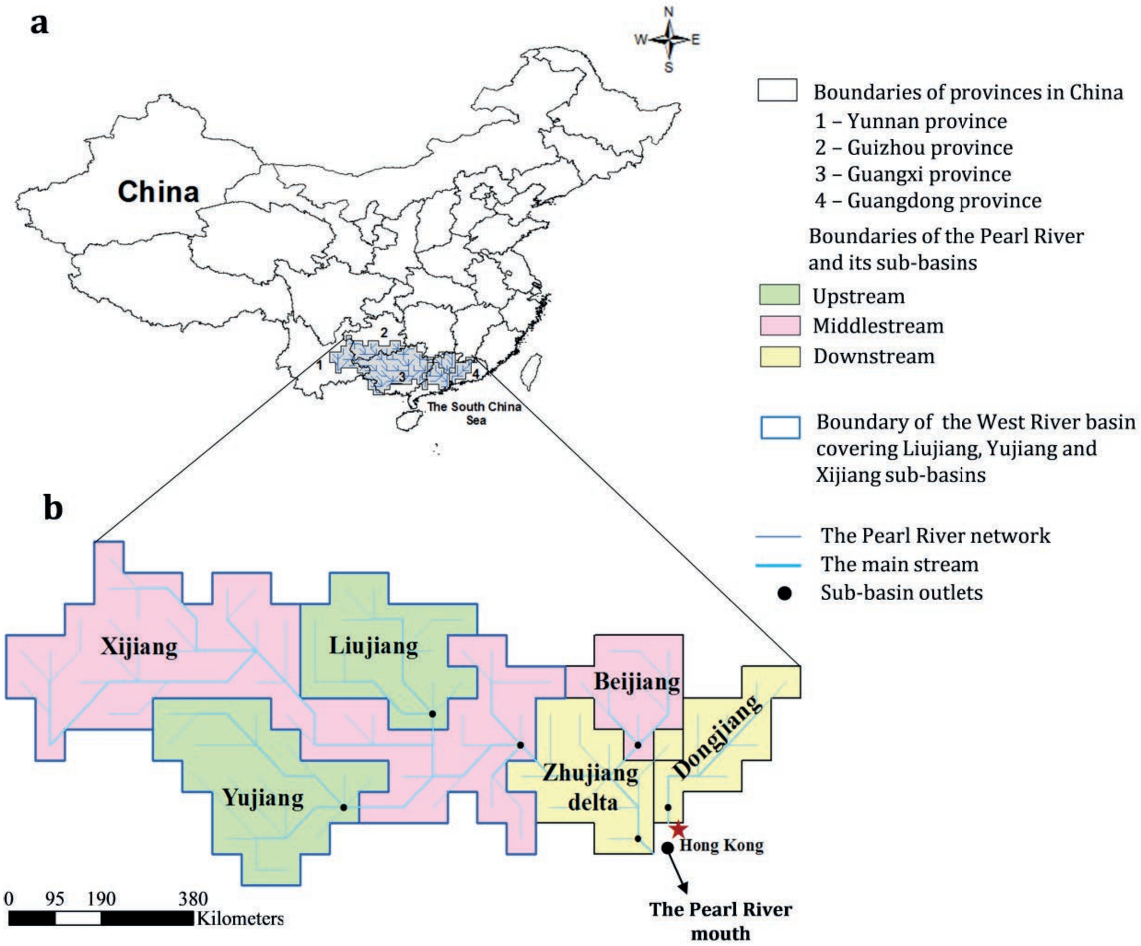

__Figure 3.1.__ (a) The Pearl River basin located in China and (b) the Pearl River sub-basins. Provincial boundaries are from the Global Administrative Areas (GADM 2012). Sub-basins are delineated based on the Topological Simulated Network (STN-30 v6.01) (Mayorga et al. 2010; Vörösmarty et al. 2000b).

Our sub-basin model quantifies DIN and DIP transport to the mouth of the Pearl River (indicated as M) from each sub-basin j (ju, jm and jd for upstream, middlestream and downstream sub-basins, respectively) and from each source y as follows:

$$M_{F.y.ju} = (RS_{F.y.ju} \cdot FE_{riv.F.ju}) \cdot FE_{riv.F.jm} \cdot FE_{riv.F.jd} \quad (3.1)$$

$$M_{F.y.jm} = (RS_{F.y.jm} \cdot FE_{riv.F.jm}) \cdot FE_{riv.F.jd} \quad (3.2)$$

$$M_{F.y.jd} = RS_{F.y.jd} \cdot FE_{riv.F.jd} \quad (3.3)$$

where

$M_{F.y.ju}$, $M_{F.y.jm}$, $M_{F.y.jd}$ are the transport of nutrients by form (F: DIN, DIP) to the mouth of the Pearl River from source y and from upstream (ju), middlestream (jm) and downstream (jd) sub-basins (kg year^{-1}), respectively. $RS_{F.y.ju}$, $RS_{F.y.jm}$, $RS_{F.y.jd}$ are nutrient inputs (F: DIN, DIP) to the river system (RS) from source y in upstream (ju), middlestream (jm) and downstream (jd) sub-basins (kg year^{-1}). $FE_{riv.F.ju}$, $FE_{riv.F.jm}$,

$FE_{riv.F.jd}$ are fractions of nutrient (F: DIN, DIP) inputs to the rivers that are exported at the outlet of upstream (ju), middlestream (jm) and downstream (jd) sub-basins (0-1).

We applied Global *NEWS*-2 (Mayorga et al. 2010) to model $RS_{F.y.ju}$, $RS_{F.y.jm}$, $RS_{F.y.jd}$ and $FE_{riv.F.ju}$, $FE_{riv.F.jm}$, $FE_{riv.F.jd}$. Box S3.1 and Table S3.1 in Supplementary Materials summarize the main equations to quantify these variables. These variables are modeled similarly for each sub-basin and thus they are indicated as $RS_{F.y.j}$ and $FE_{riv.F.j}$ without specifying up-, middle- and downstream sub-basins in Box S3.1.

Nutrient inputs to the river system include diffuse ($RSdif_{F.y.j}$) and point ($RSpnt_{F.y.j}$) source inputs of nutrients from land. *Diffuse sources* include synthetic N and P fertilizer use (for DIN, DIP), animal manure excretion (for DIN, DIP), atmospheric N deposition on agricultural areas and non-agricultural areas, and biological N fixation (for DIN) by agricultural crops and natural vegetation, and weathering of P-contained minerals in agricultural and non-agricultural areas. In this study agricultural areas combine five land use types: grassland in pastoral systems (e.g., dominated by grazing, limited manure storage and application), grassland in mixed systems (e.g., areas close to rivers, manure storage and application can take place), wetland rice, legumes (e.g., soybean and pulses) and cropland (e.g., maize, cereals) (Bouwman et al. 2009). Non-agricultural areas include any other land cover.

Nutrient inputs to the river system in sub-basin y from diffuse source j, except for P weathering, are calculated as (see Box S3.1 in Supplementary Materials) (Bouwman et al. 2009; Mayorga et al. 2010):

$$RSdif_{F.y.j} = WSdif_{E.y.j} \cdot G_{F.j} \cdot FE_{ws.F.j} \tag{3.4}$$

where

$RSdif_{F.y.j}$ is the nutrient input to the river system (RS) from diffuse (dif) source y in sub-basin j (kg year^{-1}). $WSdif_{E.y.j}$ is the inputs of nutrient by element (E: N, P) to land from diffuse source y in sub-basin j (kg year^{-1}). $G_{F.j}$ is the fraction of land-surface diffuse F (F: DIN, DIP) sources remaining after animal grazing and crop harvesting in sub-basin j (0-1), applicable for agricultural areas only. It is calculated as nutrient export from land by animal grazing and crop harvesting divided by the total nutrient inputs to agricultural land from all diffuse sources (see Box S3.1 and Table S3.1 in Supplementary Materials). $FE_{ws.F.j}$ is the fraction of nutrient form (F: DIN, DIP) that is exported from land (watershed) to the river of sub-basin j (0-1). This fraction is calculated as a function of annual runoff from land to surface waters (see Box S3.1 and Table S3.1 in Supplementary Materials).

Weathering of P-containing minerals in soils of agricultural and non-agricultural areas is modeled as a function of annual runoff from land to streams (an export-coefficient approach), following Global *NEWS*-2. Details are presented in Box S3.1 and Table S3.1 in Supplementary Materials and in Mayorga et al. (2010).

Point sources include human waste (for DIN and DIP) and detergents (for DIP). Nutrient inputs to the rivers of each sub-basin from each point source are calculated (see Box S3.1 and Table S3.1) (Mayorga et al. 2010; Van Drecht et al. 2009):

$$RSpnt_{F.y.j} = [(1 - hw_{frem.E.j}) \cdot I_j \cdot WShw_{E.y.j}] \cdot FEpnt_{F.j} \quad (3.5)$$

where

$RSpnt_{F.y.j}$ is the nutrient input to the river system (RS) from point (pnt) source y in sub-basin j (kg year^{-1}). $hw_{frem.E.j}$ is the fraction of nutrient element (E: N, P) removed during wastewater treatment in sewage facilities of sub-basin j (0-1). I_j is the fraction of population connected to sewage facilities (0-1). $WShw_{E.y.j}$ is the input of nutrient element (E: N, P) in human waste (y) and detergents (y) in watersheds (land) of sub-basin j (kg year^{-1}). $FE_{pnt.F.j}$ is the fraction of nutrient element (E: N, P) in sewage effluents that is entering the river of sub-basin j as a form (F: DIN, DIP) (0-1).

3

The river export fractions ($FE_{riv.F.j}$ in equations 3.1-3.3) reflect the retention of nutrient form (F: DIN, DIP) in the river system ($L_{F.j}$, 0-1), in dammed reservoirs ($D_{F.j}$, 0-1), and nutrient losses by water consumption for purposes such as irrigation and hydropower ($FQrem_j$, 0-1) (Mayorga et al. 2010):

$$FE_{riv.F.j} = (1 - L_{F.j}) \cdot (1 - D_{F.j}) \cdot (1 - FQrem_j) \quad (3.6)$$

River retention ($L_{F.j}$, 0-1) is considered for DIN only in Global *NEWS*-2 (Mayorga et al. 2010), and reflects the denitrification. The fraction of DIN retention in rivers is calculated as a function of sub-basin area. In this study we also consider river retentions of DIP (e.g., by sedimentation processes). For DIP we assume a 50% retention for all sub-basins and years based on Strokal and de Vries (2012). This reflects the net effect of different retention processes (e.g., accumulation of phosphorus in sediments due to its binding by iron). We discuss this assumption in Section 3.3.4.

Nutrient retentions in dammed reservoirs of each sub-basin ($D_{F.j}$) are calculated following Global *NEWS*-2 (Mayorga et al. 2010): first, nutrient retention is quantified for each reservoir and then averaged over the sub-basin using actual water discharge (after water is removed for consumption) at the outlet of the sub-basin (see Box S3.1 and Table S3.1 for details).

Nutrient removal from the river system by water consumption ($FQrem_j$, for DIN and DIP) is a function of actual and natural (before water is removed for consumption) water discharges at the outlet of each sub-basin (Box S3.1 and Table S3.1) (Mayorga et al. 2010).

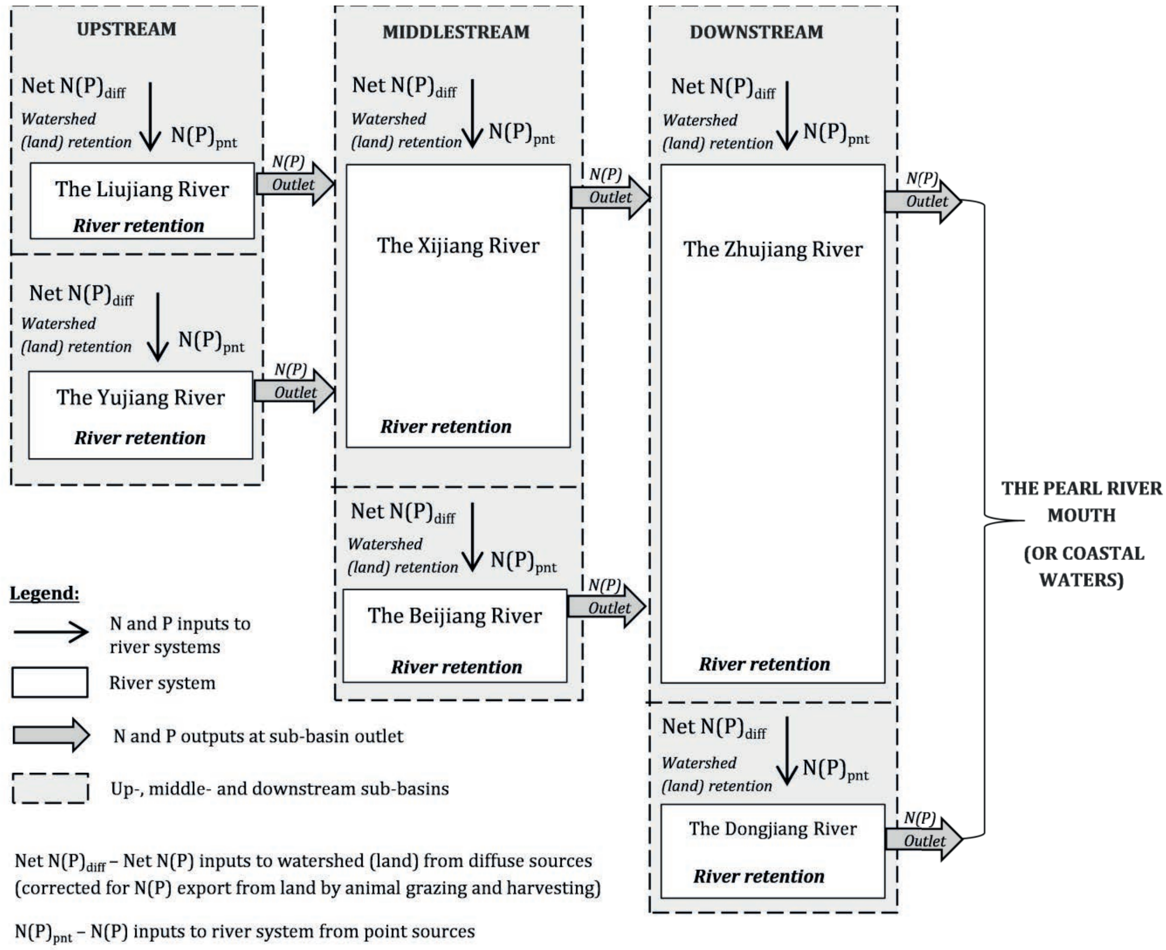

Figure 3.2. *Sub-basin scale modeling framework for the Pearl River.*

3.2.2 Model inputs

In this study, most of the model inputs are from existing gridded global datasets developed for the Global *NEWS*-2 model (0.5 x 0.5 degree longitude by latitude). Model parameters are from Global *NEWS*-2, except for reservoir characteristics, which are from the Global Reservoir and Dam (GRanD) database. Below we describe the sources of model inputs and parameters in more detail.

Gridded datasets of the Global *NEWS*-2 model were developed in earlier studies (Bouwman et al. 2009; Fekete et al. 2010; Van Drecht et al. 2009). Bouwman et al. (2009) and Van Drecht et al. (2009) prepared model inputs for diffuse (e.g., nutrient inputs to land from synthetic fertilizers, animal manure applications) and point (e.g., human waste production) sources of nutrients using the IMAGE (Integrated Model for the Assessment of the Global Environment) model. Fekete et al. (2010) prepared hydrological model inputs (e.g., runoff, water discharge) using the Water Balance Model (WBM) model. These datasets are available for 1970, 2000 and 2050. And, they were implemented to the original Global *NEWS*-2 model (Mayorga et al. 2010; Seitzinger et al.

2010). For the year 2050, the storylines of the four Millennium Ecosystem Assessment (MEA) scenarios (Alcamo et al. 2005) were quantitatively interpreted by the IMAGE model to produce the gridded inputs (Seitzinger et al. 2010). These scenarios are Global Orchestration (GO), Adapting Mosaic (AM), TechnoGarden (TG) and Order from Strength (OS) (Alcamo et al. 2005; Carpenter et al. 2006; Seitzinger et al. 2010). These storylines address future trends in climate and hydrology (Fekete et al. 2010), and in nutrient management for agriculture (Bouwman et al. 2009) and sewage (Van Drecht et al. 2009). Detailed descriptions of the scenarios can be found in various sources (Alcamo et al. 2005; Bouwman et al. 2009; Carpenter et al. 2006; Fekete et al. 2010; Seitzinger et al. 2010; Van Drecht et al. 2009).

We used these existing gridded datasets to derive the following variables for Pearl River sub-basins for 1970, 2000 and 2050 (Box S3.1, Table 3.1): sub-basin areas, land use (agricultural areas and non-agricultural areas), nutrient inputs to watersheds (land) from diffuse ($WSdif_{E.y.j}$) and point sources ($WShw_{E.y.j}$), total population and population with a sewage connection (I_j), annual runoff from land to streams and water discharges at the sub-basin outlets. We applied ArcGIS functions to aggregate gridded information to the sub-basin scale (for details see Table S3.1). For 2050 we used the gridded dataset prepared based on the GO scenario because it assumes a globalized trend in socio-economic development with a reactive approach towards environmental management. Economic growth is driven by globally connected markets. Population growth is moderate because GO assumes low fertility (e.g., 2 births per woman in China) and mortality, as a result of better education (Alcamo et al. 2005). Investments in education and infrastructure are assumed to be high in this world (Alcamo et al. 2005; Carpenter et al. 2006). People can migrate across national borders because of the globally connected society, resulting in higher migration rates (Alcamo et al. 2005). Also the urban population is assumed to increase as a result of migration of people from rural to urban areas for better jobs (Alcamo et al. 2005; Van Drecht et al. 2009). Society will have access to better sanitation and improved sewage wastewater treatment. Thus more people are assumed to be connected to sewage systems (Van Drecht et al. 2009). This may increase nutrient inputs to rivers from sewage unless sewage treatment is effective enough to reduce nutrients. Food production will be diversified to meet the food demand of a growing world population (Bouwman et al. 2009). This is associated with increased nutrient inputs to agriculture. The demand for energy and irrigation will drive construction of dams (Fekete et al. 2010), influencing nutrient retention in the river network. In this GO world, the economy is generally considered more important than the environment. Environmental problems will be addressed only when they occur. Local environmental problems will be difficult to solve due to globalization trends (Alcamo et al. 2005; Carpenter et al. 2006).

Table 3.1. *An overview of model variables and parameters. X indicates whether variables / parameters vary among sub-basins, nutrient elements (N, P) or years. Blanks indicate that model variables / parameters do not vary among sub-basins, nutrient elements (N, P) or years. Sources: A is global datasets at 0.5 by 0.5 degree (Bouwman et al. 2009; Fekete et al. 2010; Van Drecht et al. 2009); B is basin scale information from Global NEWS-2 (Mayorga et al. 2010); C is Global Reservoir and Dam database (GRanD) (Lehner et al. 2011a; Lehner et al. 2011b). Q means calculated as indicated in the text. For details see Table S3.2.*

Variable / parameter	Varying among			Source
	Sub-basins	Nutrients	Years	
Total area	X			A
For diffuse sources:				
Agricultural area	X		X	A
Watershed diffuse sources and watershed export	X	X	X	A
Watershed export constant		X		B
Diffuse export coefficient	X			B
For point sources:				
Watershed point sources	X	X	X	Q
Nutrient removal during sewage treatment	X[(a)]	X	X[(a)]	B
Total population and population with sewage connection	X		X	A
For hydrology and reservoirs				
Runoff, actual and natural water discharges	X		X	A
Data for each reservoirs: volume, depth and water discharge	X	X	X	C
Retention in reservoirs	X	X	X[(b)]	Q
Retention in the river	X	X[(c)]		Q
Runoff shape constants		X		B

(a) Nutrient removal is the same for the Zhujiang River sub-basins (Liujiang, Yujiang, Xijiang, Beijiang and Zhujiang delta). Values for 1970 are from Global *NEWS*-2 (Mayorga et al. 2010). Values for the Dongjiang sub-basin are assumed for N to be 0.8 in 2000 and 2050, and for P to be 0.8 in 2000 and 0.9 in 2050 based on expert knowledge (see the text). **(b)** Nutrient retention in reservoirs of each sub-basin ($D_{F.j}$) is calculated for each reservoir based on input data from GRanD database, and then averaged for sub-basins using actual water discharge at the outlet of the sub-basin for 1970 and 2000. For 2050, reservoir-specific input data are not available. We assumed a fixed ratio between 2000 and 2050 for D_{DIN} and D_{DIP} for sub-basins derived from Global *NEWS*-2 (Mayorga et al. 2010) for the Zhujiang and Dongjiang basins (see Figure 3.1). The factor was calculated for these basins as: (value of 2050 minus value of 2000) / value of 2000. This way, we calculated a factor of 1.7 increase for the sub-basins of the Zhujiang basin (the Liujiang, Yujiang, Xijiang, Beijiang and Zhujiang delta) and 3.0 for the Dongjiang sub-basin. **(c)** Nutrient retention in the river (e.g., via denitrification for DIN, sedimentation processes for DIP) is calculated as a function of sub-basin area (see Box S3.1) for DIN. For DIP, we assumed 0.5 based on Strokal and de Vries (2012).

Basin scale information from Global *NEWS*-2 (Mayorga et al. 2010) was taken for the following model parameters: watershed export constant (needed to calculate $FE_{ws.F}$), diffuse export coefficient (needed to calculate DIP from weathering), constants determining runoff curve (needed for $FE_{ws.F}$) and nutrient removal during sewage treatment ($hw_{frem.F}$, needed to calculate $RSpnt_{F.y.j}$) (see equations 3.4, 3.5, Table 3.1 and Box S3.1). These are basin scale parameters. Global *NEWS*-2 (Mayorga et al. 2010) divides the Pearl River into two basins: the Zhujiang (covering the Liujiang, Yujiang, Xijiang, Beijiang and Zhujiang delta sub-basins in our study) and Dongjiang (Figure 3.1b). We used the model parameters for the Zhujiang basin in Mayorga et al. (2010) for the Liujiang, Yujiang, Xijiang, Beijiang and Zhujiang delta sub-basins for 1970, 2000 and 2050. For the Dongjiang sub-basin we also adopted values from Mayorga et al. (2010), except for N and P removal during waste water treatment for 2000 and 2050. For 2000 we assumed an N and P removal fraction of 0.80, and for 2050 this fraction is assumed 0.80 for N and 0.90 for P. These removal fractions are considerably higher than in the other sub-basins (all <0.4 for N and <0.5 for P), reflecting the technologies currently implemented in the Hong Kong area (based on expert judgment).

The GRanD database (Lehner et al. 2011a; Lehner et al. 2011b) was used for model inputs for dams and reservoirs. It includes 64 dams constructed during the period of 1957-2000 within the Pearl River sub-basins (Table S3.6). From the GRanD database we derived the following model inputs for each reservoir: volume, depth and water discharge (see Box S3.1 and Table S3.6). These inputs are needed to quantify DIN and DIP retentions in dammed reservoirs.

Model inputs for reservoirs for 2050 are not available at the sub-basin scale from the GRanD database. We, therefore, estimated the DIN and DIP retentions in reservoirs for each sub-basin ($D_{DIN.j}$, $D_{DIP.j}$, see equation 3.6) by assuming that the change between 2000 and 2050 at the sub-basin scale is the same as at the basin scale. To this end, we calculated the change in $D_{DIN.j}$ and $D_{DIP.j}$ between 2000 and 2050 from Mayorga et al. (2010) for the Zhujiang and Dongjiang basins (see Table 3.1).

3.2.3 Model validation

Global *NEWS*-2 was validated for world rivers (Mayorga et al. 2010) and for continents (Strokal & Kroeze 2013; Suwarno et al. 2013; Thieu et al. 2010; Van der Struijk & Kroeze 2010; Yasin et al. 2010) including the Chinese rivers (Qu & Kroeze 2010). Their results confirmed an acceptable performance of the model for DIN and DIP. For example, the Nash-Sutcliffe efficiency (R_{NSE}^2) is 0.54 for DIN and 0.51 for DIP export by large world rivers according to validation results of Mayorga et al. (2010).

In a recent study (Strokal et al. 2014b) we evaluated further the model for DIN and DIP export by Chinese rivers (including the Pearl River). We used the Pearson's coefficient of determination (R_P^2), R_{NSE}^2 and Model error (ME) (see Moriasi et al. (2007) for detailed descriptions). We calculated an R_P^2 of 0.96, R_{NSE}^2 of 0.42 and ME of 18% for DIN and DIP export by the Chinese rivers. We concluded that the model performance is good for the Chinese rivers. However, the model seems to overestimate yields of dissolved inorganic nutrients for China to some extent (Qu & Kroeze 2010) or/and underestimates retention of nutrients in river systems of large basins like the Pearl.

Here we evaluated the model at the sub-basin scale for the Pearl River. We compared modeled fluxes of DIN and DIP for 2000 with observations. Observations are available for DIN and DIP fluxes at the mouth of the Pearl River and at the outlets of the Dongjiang, Yujiang and Xijiang sub-basins. The observed fluxes (see Table S3.3 for the literature sources) were calculated from nutrient concentrations using reported water discharges and areas (see Table S3.4 for the literature sources).

Our modeled values are generally in line with the observed DIN and DIP fluxes (Figure 3.3 and Table S3.3). We calculate about 430 kg km^{-2} $year^{-1}$ of DIN and 10 kg km^{-2} $year^{-1}$ of DIP inputs exported from all sub-basins of the Pearl River to the coastal waters. These values are in reasonable agreement with literature: 523 to 1148 kg km^{-2} $year^{-1}$ for DIN and 6.3 to 29 kg km^{-2} $year^{-1}$ for DIP (Figure 3.3 and Table S3.3). For the Dongjiang sub-basin the modeled DIN and DIP fluxes at its outlet fall in the range of the observations (Figure 3.3). The observations for DIN, however, vary greatly in the literature that may be associated with differences in the locations of measurement, selected the nutrient forms and time period considered. For example, we calculate around 2000 kg DIN km^{-2} $year^{-1}$, while this value varies between observed 438 and 2864 kg DIN km^{-2} $year^{-1}$ (Table S3.3). For the Xijiang and Yujiang outlets only concentrations of ammonium (NH_4-N) were available (Table S3.4). We, thus, calculated modeled NH_4-N fluxes for them (see Table S3.3). Modeled values for these sub-basins are within the range of observed values (Figure 3.3). For the Xijiang outlet we model 58 kg km^{-2} $year^{-1}$ while observed values are in the range of 41-108 kg km^{-2} $year^{-1}$. For the Yujiang outlet we model fluxes (69 kg km^{-2} $year^{-1}$) within the observed range (4 and 87 kg km^{-2} $year^{-1}$) (Table S3.3 and Figure 3.3).

Our validation results (Figure 3.3) also indicate that the sub-basin model intends to underestimate DIN and DIP export by the Pearl River to the coastal waters. This may be caused by missing parameters in the model representing other sources of nutrients in rivers like direct discharges of animal manure to rivers (see discussion in Section 3.3.4). In addition to model validation, we also verified some of our model inputs by comparing them with an independent Chinese county dataset (Figure S3.1). This comparison shows a good agreement between two datasets: our gridded dataset and county dataset (R_P^2 is in the range of 0.82-0.99 for compared model inputs). We discuss this comparison in Section 3.3.4.

Based on the validation results we consider the model applicable to the Pearl River. We realize that we rely on a limited number of observations. Some of these observations vary among literature sources (e.g., DIN observed yields for the Dongjiang, see Table S3.3). This indicates that more measurements are needed to test the accuracy of the model. Despite these limitations, the comparison of modeled DIN and DIP values with the few available observations gives us an indication that the model performs in a satisfactory way. This is in line with earlier model validations at the scale of China, with a similar model at the basin scale (Qu & Kroeze 2010; Qu & Kroeze 2012; Strokal et al. 2014b).

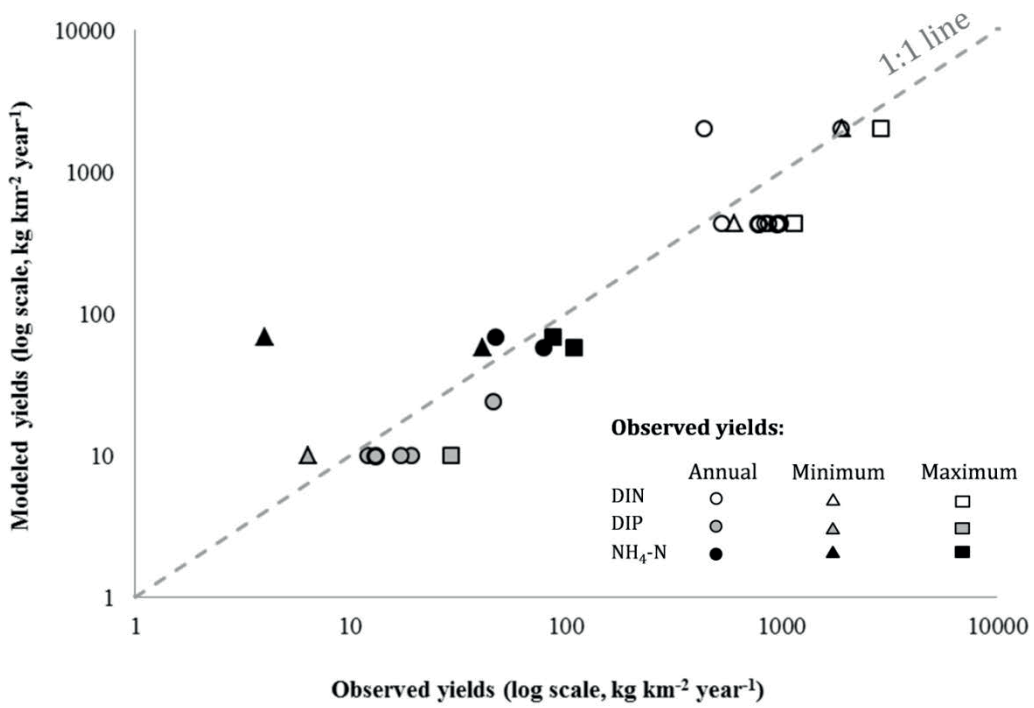

Figure 3.3. *Modeled versus observed yields of dissolved inorganic nitrogen (DIN) and phosphorus (DIP) export by the Pearl River at the sub-basin scale (log scale, kg km^{-2} year^{-1}). Modeled ammonium (NH4-N) yields were calculated assuming that 15% of the total DIN is in ammonium form according to (Meybeck 1982). Tables S3.3 and S3.4 in Supplementary Materials provide detailed information on the sources of observed yields.*

3.3 Results and discussion

3.3.1 Characteristics of the sub-basins

The middlestream sub-basins include the Xijiang and Beijiang, and cover half of the drainage area of the Pearl River. The remainder of the drainage area includes the upstream sub-basins Yujiang and Liujiang (30%) and the downstream sub-basins Zhujiang delta and Dongjiang (20%) (see Figure 3.1, Table 3.2).

Agriculture is one of the main suppliers of N and P inputs to land (Figure 3.4). In 1970 agriculture was less intensive than in 2000. Agricultural areas for crop and livestock production covered less than 10% of the sub-basins except for the Xijiang where about half of the basin is covered by agricultural areas (Figure 3.4). By 2000 these areas had expanded considerably in all sub-basins especially in the downstream Dongjiang where almost all land was agricultural. This expansion of agriculture implied increased fertilizer use and manure excretion. The N inputs to land from synthetic N fertilizer use and animal manure increased by a factor of 12 in the Liujiang, Xijiang and Zhujiang delta sub-basins, and by a factor of 50-55 in the Yujiang and Dongjiang sub-basins between 1970 and 2000 (Figure 3.4). Likewise, P inputs to land doubled in the Liujiang, Xijiang and Zhujiang delta sub-basins and increased about seven-fold in the Yujiang and Dongjiang sub-basins.

By 2050 the calculated agricultural areas are smaller than in 2000 in most sub-basins. This may seem surprising given the increasing population, but it can be explained by the assumed expansion of urban areas (Qu & Kroeze 2010; Qu & Kroeze 2012). Nevertheless, N and P inputs to land may still increase (Figure 3.4), indicating the intensity of agricultural practices in the future.

Inputs of N and P to rivers from agricultural sources (fertilizers and manure) depend on nutrient retentions in watersheds (land). In this study the watershed export fractions ($FE_{ws.F}$ in Table 3.2) account for these retentions. This fraction is lower for the up- and middlestream sub-basins than for the downstream sub-basins (Table 3.2). In other words, lower fractions of N and P inputs to land enter rivers in upstream and middlestream sub-basins. This can in part be explained by lower annual runoff from land to streams in those sub-basins (for details on hydrology see Table S3.5). We do not calculate large differences in watershed export fractions between past and future years. These export fractions, however, are much higher for DIN than for DIP. This is because P has generally a stronger ability for accumulation is soils than N (e.g., Bouwman et al. 2009; Schoumans & Groenendijk 2000).

Sewage is another important source of N and P in the sub-basins (Figure 3.4). In this study sewage sources include wastewater from human excreta (for N and P) and detergents (for P: from laundry and dishwashers). In 1970 the sewage production of N and P in the watersheds of the Zhujiang delta sub-basin was higher (0.9 ton km^{-2} for N, 0.2 ton km^{-2} for P) than in the other sub-basins (about 0-0.4 ton km^{-2} for N, 0-0.08 ton km^{-2} for P). Between 1970 and 2000 N and P production doubled in all sub-basins. From 2000 onwards nutrient production from sewage may increase further under a globalized word with reactive environmental management (Figure 3.4). An important reason of these increases is the growing population and economy (Qu & Kroeze 2010; Qu & Kroeze 2012). The Zhujiang delta sub-basin was more populated (around 300 inh. km^{-2}) than the upstream (about 90-95 inh. km^{-2}), middlestream (120-130 inh. km^{-2}) and Dongjiang (about 170 inh. km^{-2}) sub-basins in 1970 (Table 3.2). Between 1970 and 2000, the population in the Pearl River basin increased by around 55% and may continue increasing in the future (Table 3.2).

Table 3.2. *Characteristics of the sub-basins of the Pearl River: sub-basin area, population density, watershed ($FE_{ws.F}$) and river ($FE_{riv.F}$) export fractions for dissolved inorganic nitrogen (DIN) and phosphorus (DIP) in 1970, 2000 and 2050. See Section 3.2.2 for information on model inputs.*

Sub-basins	Area (1000 km^2)	Year	Population (inh km^{-2})[(a)]	Watershed export fraction ($FE_{ws.F}$, 0-1)[(b)]		River export fraction ($FE_{riv.F}$, 0-1)[(b)]	
				$FE_{ws.DIN}$	$FE_{ws.DIP}$	$FE_{riv.DIN}$	$FE_{riv.DIP}$
Liujiang	58	1970	89	0.38	0.05	0.32	0.42
		2000	136	0.38	0.05	0.30	0.22
		2050	137	0.47	0.07	0.28	0.07
Yujiang	77	1970	95	0.32	0.04	0.32	0.42
		2000	148	0.24	0.02	0.29	0.37
		2050	154	0.27	0.03	0.29	0.35
Xijiang	194	1970	121	0.38	0.05	0.30	0.24
		2000	186	0.38	0.05	0.23	0.06
		2050	187	0.47	0.07	0.18	0.07
Beijiang	31	1970	133	0.55	0.09	0.38	0.45
		2000	205	0.54	0.09	0.28	0.07
		2050	205	0.69	0.12	0.22	0.07
Zhujiang delta	48	1970	308	0.62	0.11	0.31	0.39
		2000	473	0.59	0.10	0.28	0.29
		2050	474	0.67	0.12	0.28	0.25
Dongjiang	34	1970	167	0.69	0.12	0.34	0.35
		2000	257	0.61	0.11	0.30	0.22
		2050	258	0.69	0.12	0.18	0.06

(a) Average values were calculated as the total population of the sub-basin divided by the area of the sub-basin. **(b)** Values were calculated based on equations in Box S3.1.

The amount of N and P entering rivers from sewage sources depends on the number of people connected to sewage systems and the effectiveness of nutrient treatment in these sewage systems (Van Drecht et al. 2009). In 1970 the number of people connected to sewage systems was very low, except for the Zhujiang delta sub-basin (Figure 3.4). As a result N and P inputs from sewage systems were not discharged to rivers of those sub-basins. These nutrients were assumed to be lost or retained in the terrestrial system in line with Van Drecht et al. (2009). In contrast, about 15% of the population in the Zhujiang delta sub-basin was with sewage connection in 1970 (Figure 3.4), however, sewage treatment was absent. By 2000 more people became connected to sewage systems especially in the downstream sub-basins (Figure 3.4). N and P removal via sewage treatment was about 10% for the sub-basins except for the Dongjiang (80%). By 2050 almost two-thirds of the Zhujiang delta population and half of the Dongjiang population might be connected to sewage systems (Figure 3.4). In other sub-basins, however, sewage connection is lower, especially in the Beijiang sub-basin (Table 3.2,

Figure 3.4). The efficiency of nutrient treatment is expected to increase in the coming years (see Section 3.2 for model inputs).

The river network can retain considerable amounts of N and P, and this is influencing nutrient export to coastal waters. In this study the river export fraction ($FE_{riv.F}$ in Table 3.2) accounts for nutrient retentions within (via dam construction and in-river retentions) and losses from (via water consumption) the river network (see Tables S3.5 and S3.6 for details). This fraction ranges from 0.25 to 0.45 in 1970, depending on the sub-basin (Table 3.2). Between 1970 and 2000 this fraction decreased slightly for DIN for all sub-basins and decreased largely for DIP with larger decreases for the Liujiang, Xijiang and Beijiang (Table 3.2). The main reason for these decreases is the increasing number of dams along the river network between 1970 and 2000 (Table S3.6). Between 2000 and 2050 the river export fraction may decrease further for the majority sub-basins because of envisaged damming of rivers (Table 3.2).

Another important factor influencing nutrient export to coastal waters is the distance that nutrients travel through a sub-basin to the coastal waters. For instance, DIN and DIP inputs to rivers of the upstream sub-basins have to travel across middle- and downstream sub-basins. The longer the transport takes, the more nutrients are retained within the river systems before reaching the coastal waters. Ye et al. (2012) studied the effects of nuttrient retentions in rivers and their transport to the outlet of the Vermilion Basin in east-central Illinois, USA. They also reported that the traveling distance of dissolved nutrients from upstream to downstream areas can affect their transport to the outlets.

3.3.2 Riverine inputs of dissolved inorganic N from sub-basins to coastal waters 1970-2050

We calculate that about 120 kton of DIN was exported by the Pearl River to coastal waters in 1970 (Figure 3.5, left column). By 2000 the total DIN inputs (from all sources) to coastal waters had increased by about 60%. Between 2000 and 2050 the DIN inputs to coastal waters may further increase, reaching almost 250 kton in 2050 (Figure 3.5, left column). Agricultural activities such as synthetic N fertilizer use and animal manure excretion are the main causes of these increases. The downstream sub-basins, Zhujiang delta and Dongjiang, are dominant contributors of DIN inputs to the coastal waters (Figure 3.5, right column). However, their contribution changes between past and future years (Figure 3.5).

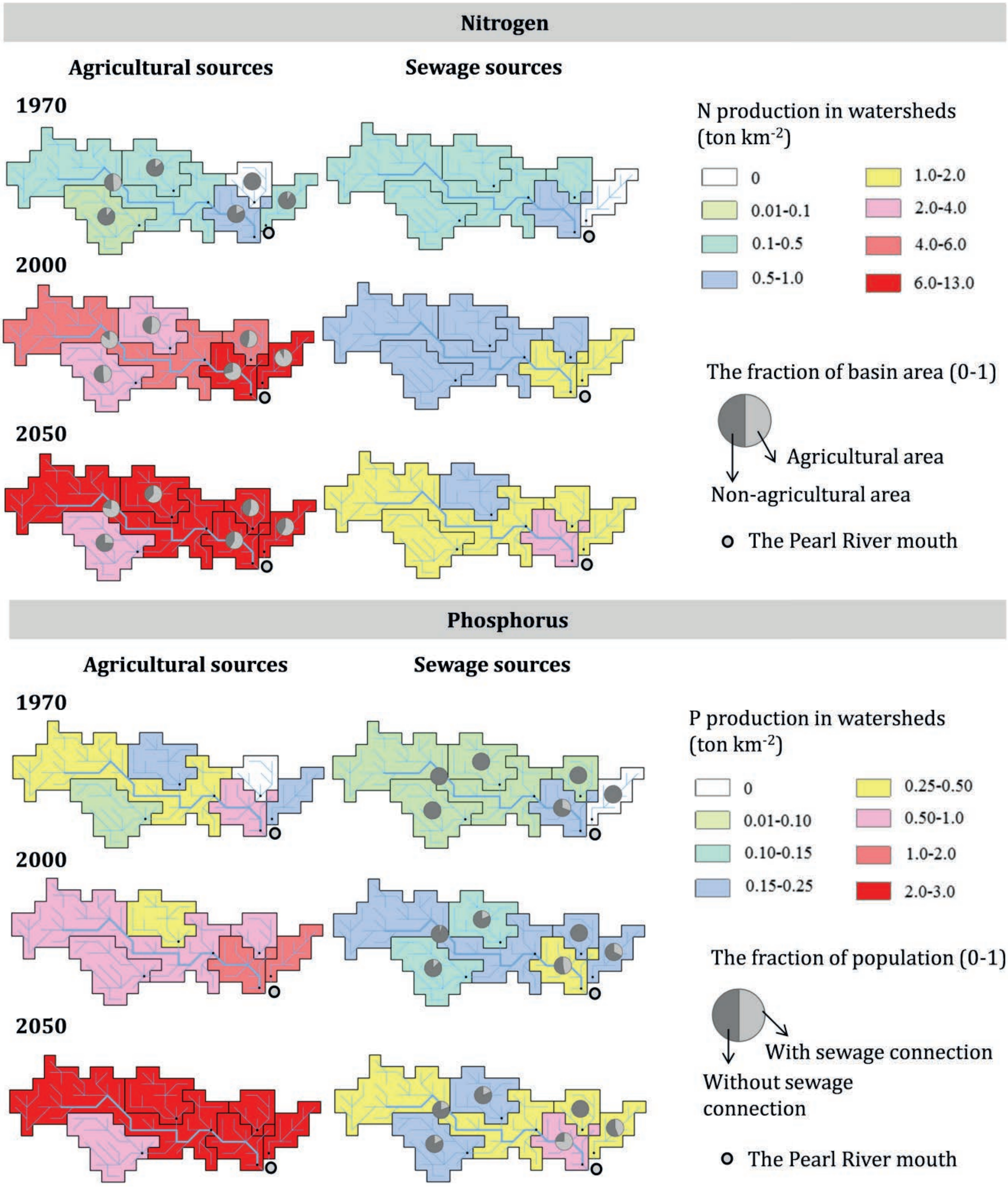

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Figure 3.4. *Nitrogen (N) and phosphorus (P) production (as element, ton km^{-2} year^{-1}) in watersheds (land) of the Pearl River sub-basins from agricultural ($WSdif_{E.y.j}$ in equation 3.4) and sewage ($WShw_{E.y.j}$ in equation 3.5) sources in 1970, 2000 and 2050. Agricultural sources include animal manure excretion and synthetic fertilizer use. Sewage sources include wastewater from human excreta (for N and P) and detergents (for P). Pie charts represent the fractions of basin areas that are agricultural and non-agricultural (left hand side), and the fractions of population with (I_j in equation 3.5) and without sewage connection (right hand side). Nutrient inputs for 2050 are based on the Global Orchestration scenario of the Millennium Ecosystem Assessment that assumes a globalized world with reactive management towards the environment. Section 3.2.2 and Table S3.2 provide information on model inputs.*

In 1970 agricultural sources contributed by 40% to the total DIN inputs to the coastal waters. About 50-70% of these agricultural DIN inputs originated from the Zhujiang delta sub-basin, about 20% from the Dongjiang sub-basin and 10-20% from the Xijiang sub-basin in 1970 (Figure 3.5, middle column). By 2000 agricultural sources were already responsible for almost two-thirds of the riverine inputs of DIN (from all sub-basins) to the coastal waters (Figure 3.5, left column). Between 1970 and 2000 the contributions of the Zhujiang delta to the agricultural DIN export decreased (45%) while the contribution of the Dongjiang sub-basins increased (35-40%). A decreased contribution of the Zhujaing delta sub-basin resulted from the net effect of increased N inputs to land from agriculture and increased N retentions in land as well as in rivers of this sub-basin (Section 3.3.1). The main reason of the increased Dongjiang contribution is the expanded agricultural areas during the period of 1970 – 2000 for crop and livestock production that increased N inputs to land (Section 3.3.1).

In 2050 agriculture is calculated to remain the dominant source of DIN in coastal waters (Figure 3.5, left column). The Zhujiang delta sub-basin may transport more agricultural DIN to the coastal waters than in 2000 (Figure 3.5) because of the projected further increases in N inputs to land from fertilizers and manure (Section 3.3.1). Inversely, the Dongjiang may export less agricultural DIN in coastal waters in 2050 than in 2000. An important reason for this is the projected decrease in N inputs to agricultural land and increase in DIN retentions in reservoirs of the sub-basins (Section 3.3.1).

Sewage was a relatively small anthropogenic source of DIN in the coastal waters of the Pearl River in 1970. Between 2000 and 2050 the share of sewage to the riverine inputs of DIN to the coastal waters is calculated to increase. The Zhujiang delta sub-basin is a major contributor to these DIN inputs (Figure 3.5). This might be explained by the growing urban population and higher number of sewage connections. As a result, DIN export to rivers can increase from sewage systems unless the efficiency of N removal in public wastewater treatment plants is high enough to reduce DIN discharges to rivers of the Zhujiang sub-basin (Section 3.3.1).

In 1970 non-anthropogenic sources such as atmospheric N deposition on non-agricultural areas and biological N fixation by natural vegetation were responsible for almost half of the total DIN inputs to the coastal waters. Between 1970 and 2000, and from 2000 onwards, their share is calculated to decrease because of increased contributions of agricultural sources (see Figure 3.5).

Our results indicate a minor contribution of the upstream sub-basins and a relatively small contribution of the middlestream sub-basins. This is the net effect of (i) spatial variations in land-based N sources, (ii) N retentions in watersheds (land) and in rivers, and (ii) traveling distances of N from sub-basins to coastal waters. This holds for riverine inputs of DIP to coastal waters as well (see Section 3.3.3). Details on sub-basin characteristics can be found in Section 3.3.1.

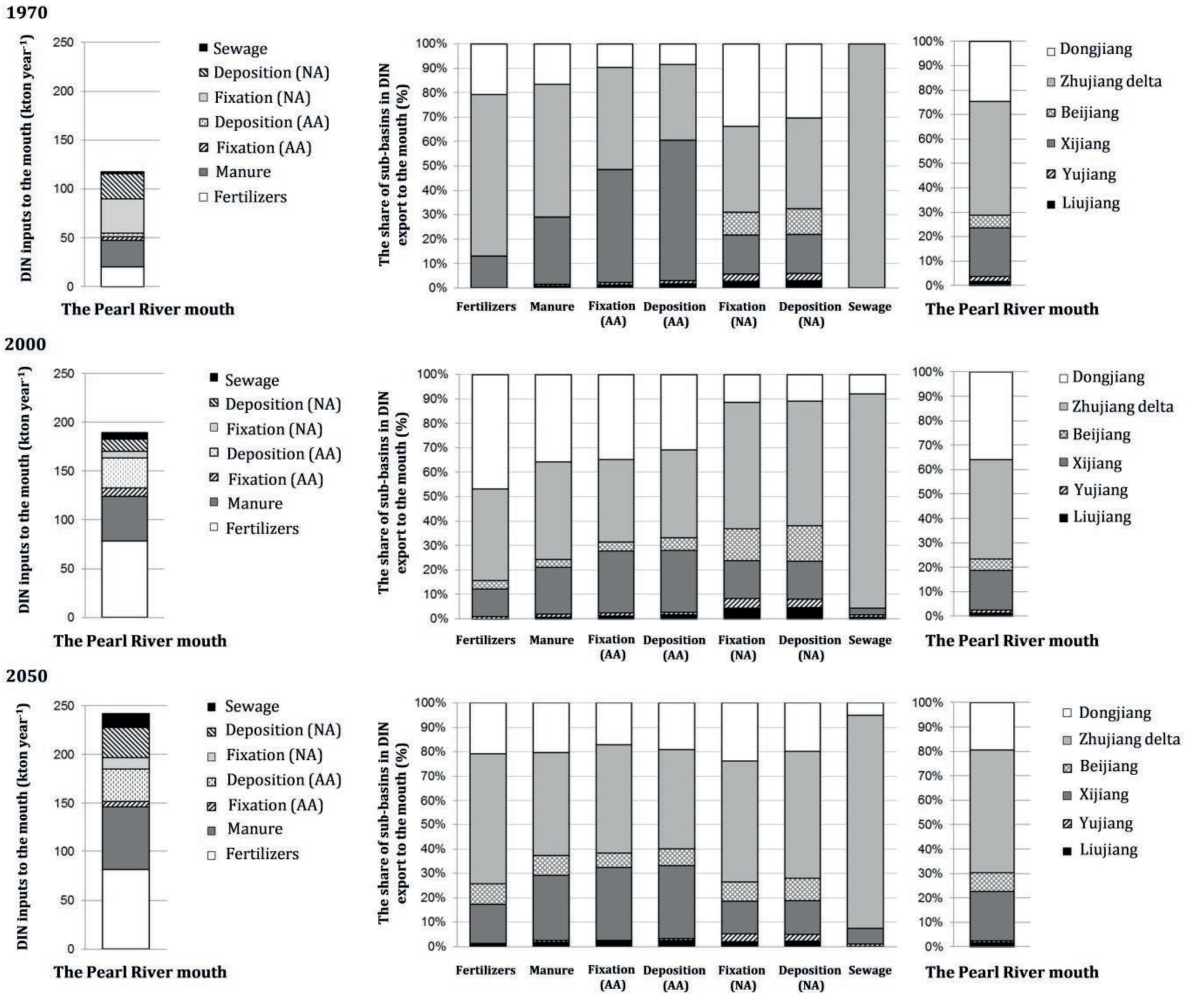

Figure 3.5. *Modeled inputs of dissolved inorganic nitrogen (DIN) to the coastal waters of the Pearl River (kton year^{-1}) by source categories (left column) and the share of Pearl River sub-basins (%) in DIN export from each source category (middle column) and in the total DIN export (right column) in 1970, 2000 and 2050. Future export is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment that assumes a globalized world with reactive management towards environment. AA is agricultural area and NA is non-agricultural area.*

3

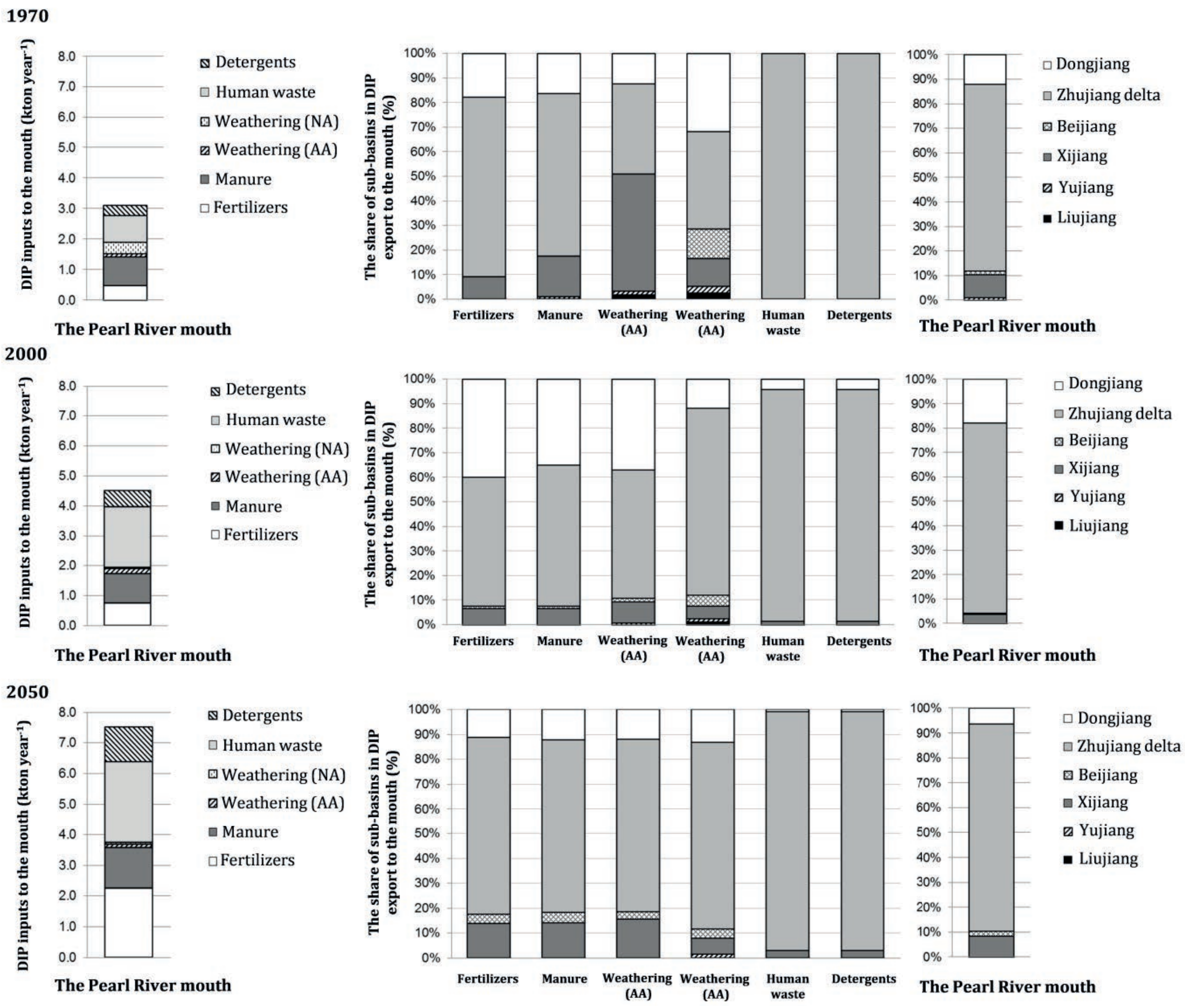

Figure 3.6. *Modeled inputs of dissolved inorganic phosphorus (DIP) to the coastal waters of the Pearl River (kton year-1) by source categories (left column) and the share of Pearl River sub-basins (%) in DIN export from each source category (middle column) and in the total DIN export (right column) in 1970, 2000 and 2050. Future export is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment that assumes a globalized world with reactive management towards environment. AA is agricultural area and NA is non-agricultural area.*

3.3.3 Riverine inputs of dissolved inorganic P from sub-basins to coastal waters 1970-2050

About three kton of total DIP inputs were exported to the coastal waters of the Pearl River from all sources and sub-basins in 1970. Between 1970 and 2000, these DIP inputs increased by one-third (4.5 kton in 2000) and may continue to increase from 2000 onwards (see Figure 3.6, left column). These increases are the result of anthropogenic sources such as agriculture and sewage. Similar to DIN (Section 0), the Zhujiang delta sub-basin is the major source of DIP in coastal waters, and this holds for both agriculture and sewage (Figure 3.6, right column). In contrast to DIN, about half of the total DIP inputs are from agriculture and another half from sewage (Figure 3.6, left column).

In 1970 about 70% of agricultural DIP inputs to the coastal waters were exported from the Zhujiang delta sub-basin, 20% from the Dongjiang and 10% from the Xijiang (Figure 3.6, middle column). The share of these sub-basins is different in 2000. The contribution of the Zhujiang delta sub-basin decreased from 70% (1970) to 50% (2000) and of the Dongjiang sub-basin increased from 20% (1970) to 40% (2000) (Figure 3.6, middle column). An important reason for the decreased contribution of the Zhujiang delta sub-basin is an increase in DIP retentions in the river system of this sub-basin due to the number of dams being doubled from 1970-2000 (Section 3.3.1). An explanation for the increased contribution of the Dongjiang sub-basin is that P inputs to agriculture increased between 1970 and 2000 as a result of increased agricultural area (see Section 3.3.1) and this increased the DIP export from land to rivers and coastal waters.

In 2050, two-thirds of the agricultural DIP inputs to the coastal waters are calculated to be from the Zhujiang delta sub-basin and the remainder from the Dongjiang (15%) and Xijiang (15%) sub-basins (Figure 3.6, middle column). These results for the Dongjiang sub-basin can be explained by the net effect of decreased agricultural areas and increased P retentions in the river system due to damming of the river (see Section 3.3.1). For the Zhujiang delta sub-basin P inputs to agricultural land from P fertilizers and animal manure applications are projected to increase and this increases P export to rivers of this sub-basin and consequently to coastal waters (Section 3.3.1).

Sewage DIP inputs to the coastal waters are mainly from the Zhujiang delta sub-basin for all years (Figure 3.6). The main reasons are: (i) this sub-basin is the most populated with a higher number of sewage connections than the others and thus (ii) the P inputs to sewage systems of this sub-basin are higher while the effectiveness of treatment is low compared to the Dongjiang (Section 3.3.1).

3.3.4 Strengths and weaknesses of the modeling approach at the sub-basin scale

As with any other model, our sub-basin model has its weaknesses. An important source of uncertainty in the model is associated with model parameters and inputs that are based on assumptions and simplifications. For instance, the watershed export constants (eDIN, eDIP in Box S3.1 and Table S3.1) and runoff shape constants (aDIN, aDIP, bDIP in Box S3.1 and Table S3.1) do not change over time, and are assumed the same for each sub-basin (see Table 3.1). These constants are used to quantify watershed export fractions ($FE_{ws.F.j}$, equation 3.4) that reflect nutrient retentions in the watershed. We realize that this simplification affects the final result. However, we assume that the associated model error is relatively small. It should be noted that these constants were taken from Global *NEWS*-2 that was calibrated for large world rivers (Mayorga et al. 2010). No data exist for a calibration for our sub-basins. Therefore, we adopted these global values. Likewise, we used global values for the diffuse export coefficient (EC_{DIP} in Box S3.1 and Table S3.1) used to quantify DIP weathering in agricultural and non-agricultural areas.

We used most of the model inputs from the gridded global datasets of Global *NEWS*-2 to quantify nutrient export from sub-basins (Bouwman et al. 2009; Fekete et al. 2010; Van Drecht et al. 2009) (see Section 3.2.2). We, however, realize that these datasets were derived from large-scale (e.g., national) assessments (such as from FAO (2008) and Bouwman et al. (2009)) and thus may contribute to uncertainties in the final results. This may be illustrated by the study by McCrackin et al. (2013), who compared predictions of Global *NEWS*-2 with the SPARROW model (SPAtially Referenced Regressions On Watersheds) for the United States basins. Some disagreements in model predictions were found in particular for smaller basins because of differences in model inputs (e.g., regional US data in SPARROW, and global datasets in Global *NEWS*-2) and modeling approaches. They concluded that model inputs can be one of the uncertainty sources in the model predictions. We compared some of our sub-basin inputs to an independent Chinese county dataset ((RESDC 2014), Wang M. pers. comm.) for the year 2000. We compared data on synthetic fertilizers, animal excretion, biological N fixation by agricultural crops, and total human population. Figure S3.1 indicates that there is a good agreement between our sub-basin inputs and sub-basin data derived from the county dataset. For example, R_P^2 is 0.82 (N) and 0.93 (P) for synthetic fertilizers, 0.97 (N) and 0.96 (P) for animal manure. For biological N fixation R_P^2 is 0.98. For the total human population and sub-basin areas this parameter is calculated at 0.91 and 0.99, respectively (Figure S3.1). Furthermore, the drainage areas of the sub-basins delineated based on the global STN-30 network (Vörösmarty et al. 2000a) are in line with various hydrological studies for the Pearl River (Cui et al. 2007; Niu & Chen 2010; Zhang et al. 2007; Zhang et al. 2008), and with the county dataset (Figure S3.1). We, thus, consider the gridded datasets acceptable for modeling nutrient export by the Pearl River at the sub-basin scale.

We realize that we used little information from independent regional datasets in our sub-basin model for the Pearl River compared to some other existing studies in the field of nutrient flows in China (e.g., Yan et al. (2010), Ma et al. (2012), Ti et al. (2012)). In contrast to our study, these existing studies analyze nutrient fluxes at a basin or provincial scale, using provincial information (e.g., from China Statistical Yearbooks) as one of the sources for their analyses. For our sub-basin analyses provincial data might not be a good choice because it should be first downscaled to sub-basins under certain assumptions and simplifications (two provinces cover about 80% of the Pearl basin, see Figure 3.1), and the data do not give enough information for 1970, 2000 and 2050. The county-based dataset, which was used for verifying some of our model inputs (see Figure S3.1), may be suitable for sub-scale modeling, but this dataset is not complete yet. Hence, in this study we used the gridded dataset because it provides the required inputs for our sub-basin model for the Pearl River.

Another source of uncertainty is associated with the very fast socio-economic development in parts of our study area, making it difficult to project changes over time. Future projections of nutrient export by the Pearl River depend on the storylines of scenarios. In this study we used the globalized GO scenario with reactive environmental management. Details on the storylines of GO can be found in Fekete et al. (2010) for hydrology, Van Drecht et al. (2009) for urbanization, and in Bouwman et al. (2009) for agriculture (see also Section 3.2.2). For the Pearl River this GO scenario assumes a moderate population growth (Table 3.2) with high economic development (e.g., a 10-fold increase in GDP at purchase power parity). Agricultural areas (except for the Beijiang and Liujiang sub-basins) may decrease (up to 46%) between 2000 and 2050 because of urbanization that requires more land for cities. Nevertheless, agriculture is projected to be intensive for food security reasons. More people are assumed to be connected to sewage systems in 2050 with relatively low efficiencies of nutrient removal during treatment. All these are reflected in the final results, showing increasing trends in nutrient export by the Pearl River (Sections 0 and 3.3.3).

It should be noted that the storylines of GO were developed from large-scale assessments (e.g., national). This means that this scenario may not account for the fast economic development in large cities of the Pearl basin like Shenzhen (located in the Zhujiang delta sub-basin) and Hong Kong. An example of this is the assumed N and P removal in sewage treatment in the original GO scenario. In most of the Pearl River basin, nutrient removal in sewage treatment is relatively low. However, some urban areas develop relatively fast, for instance in the Dongjiang sub-basin draining into Hong Kong. GO projects relatively low efficiencies of N and P removal for the Dongjiang sub-basin. This is not in line with the current developments in technology in, for example, Hong Kong. We, therefore, modified the original GO scenario, and assume a removal fraction for N and P for this sub-basin of 80-90% in sewage treatment for 2050. This implies that the latest technology in sewage treatment is implemented in our study, based on our knowledge of the fast technical developments in this sub-basin. However, data for these parameters are not available in the literature.

Another model assumption that is relatively uncertain is the retention of DIP in rivers ($L_{DIP.j}$, equation 3.6). Basin scale models often ignore retention of DIP in rivers, for example Mayorga et al. (2010) and Harrison et al. (2005b). In reality, however, not all DIP is exported to the river mouth. We assumed that 50% of DIP inputs to rivers is retained in rivers (e.g., via accumulation processes between sediment and water). This is based on Strokal and de Vries (2012) who showed that including river retention for DIP in Global *NEWS*-2 improves the model performance for large river basins in the world. They showed that 50% is a reasonable estimate, and that the model is not so sensitive to variation in this parameter. We realize that in reality this fraction differs among water bodies and nutrient forms (Chen et al. 2010; Doyle et al. 2003; Reinhardt et al. 2005; Schulz & Köhler 2006; Ti et al. 2012).

In our model, human waste is considered a point source of nutrients in rivers, and animal manure a diffuse source. In reality, however, human wastes can also be a diffuse source (e.g., toilets that are not connected to sewage systems in rural areas) while animal manure can be a point source (e.g., direct discharge into surface waters) in some areas of the Pearl River such as the Beijiang sub-basin (expert judgment). Van Drecht et al. (2009) confirm that human waste can be a diffuse source in areas without sewage systems in China. Recently, Morée et al. (2013) quantified N and P inputs to agriculture from human wastes (as a recycling) on a country scale globally. Their analysis illustrates the importance of human waste as diffuse sources of nutrients in agriculture. Other studies (Bai et al. 2013; Hou et al. 2013; Ma et al. 2010; Ma et al. 2012; Ma et al. 2013b) indicated considerable losses of nutrients to water bodies from animal manure systems in Chinese agriculture. Ti et al. (2012) indicated that 25% of livestock and human waste are directly discharged to Chinese rivers. Even though human waste is a relatively small source of nutrients in rivers and direct inputs of manure to rivers may be exceptions, we suggest to include these sources in our model in the future.

We modeled DIN and DIP export for six sub-basins. We consider this level of detail arbitrary on the one hand, but appropriate on the other, given the scarcity of empirical data needed for validation, and our wish to identify the most important causes of coastal eutrophication in the past and the future. We argue that the chosen scale is probably optimal for the Pearl River, providing results of acceptable reliability that can be used to address the most important environmental issues. Larger scales like river basin scale applied in the original Global *NEWS*-2 model are not sufficient to identify the sources of polluting nutrients in coastal waters for large river basins (see Section 3.1). More detailed models such as the SWAT model, are powerful tools, but only when they can be accurately applied. For the Pearl River upscaling of such detailed models may result in complex systems that are not transparent and with relatively large uncertainties.

Despite of the abovementioned weaknesses and limitations, we believe that our sub-basin model fits the purpose of our study. We developed the new sub-basin modeling approach that can be applied to large basins to assess effective management of coastal nutrient pollution. In this study we incorporated this approach to Global *NEWS*-2 and applied it to the Pearl River for the first time (see Section 3.2 on the methodology). There are four main strengths of the model.

First, our model is a spatially explicit model. A unique feature of the model is that it can quantify from where nutrients have been transported to the coastal waters (at sub-basin scale) and which human activities (e.g., agriculture, sewage) in these sub-basins are the sources of the nutrients. In other words, the model allows to quantify (1) the contribution of the main sources of DIN and DIP in rivers and coastal waters, and (2) the contribution of sub-basins to nutrient inputs to coastal waters. This was not done before for the Pearl River nor for other large world rivers. Various existing studies (Harrison et al. 2010; Moore et al. 2011; Yan et al. 2010; Yasin et al. 2010) quantify source attributions for nutrient export by large rivers on the basin scale. However, they do not give insights into locations of these nutrient sources within basins (see also Section 3.1). Our model fills this gap. This information is essential for effective nutrient management. For example, this study provides insights that management of human activities in downstream areas of the Pearl River will likely be more effective to reduce coastal nutrient pollution than management in upstream areas (see conclusions). In addition, the model also addresses past and future trends in DIN and DIP export. This opens the opportunity to explore options to manage nutrients in the coming decades (e.g., via sensitivity and scenario analyses) that will enable to identify effective options.

Second, our sub-basin scale model is transparent and user-friendly. The model is developed for large basins such as the Pearl River. Thus, it is easy to apply this model to other large river basins in China (e.g., the Yangtze and Yellow rivers) and worldwide. This is because many large basins are data poor and their large scale makes difficult to conduct empirical studies (see also Section 3.1). Our model, therefore, offers the possibility to quantify the nutrient export while considering sub-basin characteristics and offers also the opportunity for up scaling to all of China.

Third, the model does not require a lot of input data. Most required inputs can be derived from the existing sources that we mentioned in Section 3.2.2 In addition, we verified some of the model inputs, and the results give us a confidence of using these sources in sub-basin modeling for the Pearl River.

Fourth, taking into account all the above mentioned strengths and weaknesses, we argue that our model is well balanced in terms of the accuracy of model results and uncertainties associated with poor data availability.

From our experience we may draw lessons for modeling nutrient export at the sub-basin scale:

1. Modeling nutrient export at the sub-basin scale for large poorly documented basins is possible;
2. The size of the sub-basins may seem arbitrary, however, it is appropriate for large data-poor river basins;
3. The model has its weaknesses, but model strengths overcome these weaknesses, resulting in a proper balance between the accuracy and uncertainty;
4. The model is a useful tool for sub-basin analyses of nutrient flows.

3.4 Conclusions

Coastal eutrophication has been increasing fast in the South China Sea since the 1970s. The underlying causes are human activities that increase exports of dissolved inorganic nitrogen (DIN) and phosphorus (DIP) by the Pearl River. The objective of our study was to quantify dissolved inorganic N (DIN) and P (DIP) export from sub-basins of the Pearl River to coastal waters by source. To this end, we developed a sub-basin scale modeling approach for 1970-2050 to assess the relative shares of sources and sub-basins in the nutrient export at the river mouth. We applied this approach to the Global *NEWS*-2 (Nutrient Export from Watersheds) model. We used datasets of the Global *NEWS*-2 model to derive most of the model inputs (gridded 0.5 by 0.5 degree cell datasets), and parameters (river basin information) except for inputs needed to model nutrient retentions in reservoirs. For these retentions we used Global Reservoir and Dam databases. Future scenarios for 2050 are based on the Global Orchestration (GO) scenario of the Millennium Ecosystem Assessment (MEA), assuming a globalized world with reactive environmental management. The Pearl River basin consists of six sub-basins including upstream sub-basins (Liujiang and Xijiang), middlestream sub-basins (Xijiang and Bejiang) and downstream sub-basins (Zhujiang delta and Dongjiang). Our study illustrates the importance of applying nutrient management in agriculture and sewage within downstream sub-basins because of their large contribution to coastal eutrophication.

Dissolved inorganic N inputs to the Pearl River mouth are calculated to double between 1970 and 2050 mainly because of agricultural activities in two downstream sub-basins. In 1970 the downstream Zhujiang delta contributed 50-70% and the downstream Dongjiang about 20% to the river export of agricultural DIN. By 2000 this percentage increased to about 35-50% for the Dongjiang because of expanding agricultural activities to sustain the growing population. We calculate that from 2000 onwards the Dongjiang sub-basin may contribute less to DIN in the Pearl River because of decreasing N inputs to land from agriculture and increasing DIN retentions in reservoirs. This is different for the Zhujiang delta sub-basin with a decreasing contribution between 1970 and 2000, and an increase again by 2050 due to increasing N inputs to agricultural land. The share of the middlestream Beijiang and Xijiang sub-basins to the total DIN inputs to the river mouth is projected to increase slightly from 2000 to 2050 mainly due to agricultural activities.

Dissolved inorganic P inputs to the Pearl River mouth are calculated to increase 2.5-fold between 1970 and 2050. The main cause of this is sewage and agriculture in the downstream sub-basins. About half of the DIP in the river mouth originates from sewage (human waste and detergents) and the other half from agriculture. The downstream Zhujiang sub-basin is a major contributor to DIP inputs from sewage because of urbanization. Agriculture is a dominant source of DIP in the Zhujiang and Dongjiang sub-basins in 2000. In 1970 the Zhujiang contributed by about two-thirds to the agricultural DIP inputs to the mouth. The Dongjiang contributed about 20%. In 2000 the contribution of the Dongjiang was higher than in 1970 while the contribution of the Zhujiang was lower. This is because the Dongjiang sub-basin is dominated by agriculture (90% of the land was agricultural area in 2000) with an increasing use of P fertilizers and manure. Another reason is that the number of dams in the Zhujiang sub-basin doubled between 1970 and 2000, increasing DIP retention in rivers and reservoirs and thus decreasing DIP export to the river mouth. In 2050 the Zhujiang may contribute about two-thirds of the total DIP inputs at the river mouth originating from agriculture. The share of the Dongjiang is projected to decrease due to decreasing agricultural areas and high DIP retentions in reservoirs of this sub-basin. The relative share of the middlestream Xijiang sub-basin in DIP export by the Pearl River may increase in the future as a result of large increases in P inputs to land from agricultural activities.

The large contribution of downstream sub-basins and very low (or zero) contribution of up- and middlestream sub-basins to coastal eutrophication can be explained as follows. The first reason is the spatial variability of land-based sources of DIN and DIP. For instance, N and P production in watersheds from agricultural and sewage sources are generally higher in the downstream sub-basins compared to up- and middlestream onces. Furthermore, downstream sub-basins are densely populated areas with a higher number of people connected to sewage. The number of people connected to sewage systems is another important variable in our calculations. In up- and middlestream sub-basins the percentage of people connected to sewage systems does not exceed 15% in 2000 and 2050. This explains partly their low (or zero) contribution to the total DIP export to the Pearl River mouth. The second reason is the retention of nutrients in river systems of the sub-basins and their hydrology. For instance, the upstream sub-basins are characterized by lower annual runoff than downstream sub-basins, leading to lower nutrient export from land to rivers. Considerable amounts of nutrients remain in rivers of the sub-basins due to dams, retention processes in waters and water withdrawal for different purposes. This implies that the fractions of nutrient inputs to rivers that are actually transported to coastal seas decreases with distance to the river mouth. Or, in other words, a larger fraction of nutrients entering rivers close to the river mouth reaches the coastal waters than of nutrients entering river more upstream.

Our study is a first attempt to quantify DIN and DIP export from sub-basins of the Pearl River to the coastal waters for 1970, 2000 and 2050. The chosen sub-basin scale is appropriate for the large Pearl River basin to identify the main sources of nutrient pollution and their locations (sub-basins). We identified the most polluting sub-basins that contribute largely to DIN and DIP in coastal waters of the Pearl River. Reduction strategies are most effective in these basins. Clearly, nutrient management in agriculture and sewage in downstream sub-basins may reduce coastal eutrophication more than management in upstream sub-basins of the Pearl River. Our modeling study can, therefore, support decision making on strategies to reduce DIN and DIP inputs to rivers and thus to avoid further eutrophication in the coastal waters of the Pearl River. This study can serve as an example for other river basins, where allocation of nutrient management areas is required.

Acknowledgments

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Chapter 4.

Alarming nutrient pollution of Chinese rivers as a result of agricultural transitions

Abstract

Transitions in Chinese agriculture resulted in industrial animal production systems, disconnected from crop production. We analyzed side-effects of these transitions on total dissolved nitrogen (TDN) and phosphorus (TDP) inputs to rivers. In 2000, when transitions were ongoing, 30-70% of the manure was directly discharged to rivers (range for sub-basins). Before the transition (1970) this was only 5%. Meanwhile, animal numbers more than doubled. As a result, TDN and TDP inputs to rivers increased 2 to 45-fold (range for sub-basins) from 1970-2000. Direct manure discharge accounts for over two-thirds of nutrients in the northern rivers and for 20-95% of nutrients in the central and southern rivers. Environmental concern is growing in China. However, in the future, direct manure inputs may increase. Animal production is the largest cause of aquatic eutrophication. Our study is a warning signal and an urgent call for action to recycle animal manure in arable farming.

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4.1 Introduction

Many aquatic systems in China are polluted with nitrogen (N) and phosphorus (P), causing eutrophication and harmful algal blooms (Conley et al. 2009; Cui et al. 2013; Liu & Diamond 2005; Liu et al. 2013). Over half of the Chinese lakes are eutrophic today (Le et al. 2010). For example, the water quality in Thaihu Lake, the third largest lake in China, declined from Class I/II (oligotrophic clean water: Class I suitable for drinking and Class II suitable for fishing and bathing) in the 1960s to Class IV (eutrophic polluted water by nutrients, not suitable for drinking or bathing) (Le et al. 2010). Many Chinese rivers show similar trends. Northern rivers, such as the Yellow (Huanghe), Hai, Liao and Huai, and river deltas are polluted by nutrients to the extent that their water is not suitable for human contact (Class VI) (Xu et al. 2014). Leaching of nutrients from fertilized soils is generally considered the major cause (Cui et al. 2013; Liu et al. 2008a; Ongley et al. 2010; Tao et al. 2010; Xu et al. 2014). Nutrient leaching from land is increased by inputs from synthetic and organic fertilizers, atmospheric N deposition, and biological N fixation by crops. This leaching of N and P from land is a diffuse source of N and P in rivers.

Here, we argue that nutrient pollution of Chinese rivers is largely associated with manure discharges, i.e., point sources. Chinese agricultural transitions, in particular the recent industrialization of animal production and the disconnection of crop and animal production, result in direct discharges of animal manure to surface waters (Figure 4.1). Existing studies (Cui et al. 2013; Liu et al. 2008a; Ongley et al. 2010; Tao et al. 2010; Xu et al. 2014) generally do not account for these point sources and thus may underestimate actual nutrient loads to rivers (see Section 4.3 below). Some links between agricultural transitions and water quality were published in studies (e.g., Bai et al. 2014; Ma et al. 2010; MEP et al. 2010; Schneider 2011; Steinfeld et al. 2006) focusing largely at national analyses. China's pollution report (MEP et al. 2010) indicates that animal production is today responsible for around 20% of nitrogen and 40% of phosphorus pollution in aquatic systems. However, none of these studies explicitly account for direct discharges of animal manure to rivers. Clearly, the implications of agricultural transitions on N and P inputs to Chinese rivers are not well known, making it difficult to formulate effective environmental policies. Our analyses fill these gaps.

In our study, we quantify the nutrient pollution of large Chinese rivers associated with direct point discharges of animal manure to rivers as a result of agricultural transitions. Our analysis includes the following rivers in China: the Yellow (Huanghe), Yangtze (Changjiang), Pearl (Zhujiang), Huai, Hai and Liao. The drainage basins of the largest rivers (Yellow, Yangtze, Pearl) were divided into sub-basins (Figure S4.1). Although a few sub-basin scale analyses of the Yangtze River (Bao et al. 2006; Liu et al. 2008a) exist, ours is the first to assess nutrient pollution in Chinese rivers by source while accounting for direct discharges of manure by sub-basin.

4.2 Methodology

4.2.1 Model description and inputs

Our study is based on two models: Global *NEWS*-2 (Nutrient Export from WaterSheds) (Mayorga et al. 2010; Strokal et al. 2014b) and NUFER (NUtrient flows in Food chains, Environment and Resources use) (Ma et al. 2012). Global *NEWS*-2 is a spatially explicit model that has been applied to analyze nutrient-related problems worldwide (Sattar et al. 2014; Strokal & de Vries 2012; Strokal & Kroeze 2013; Strokal & Kroeze 2014; Strokal et al. 2014c; Suwarno et al. 2013; Suwarno et al. 2014a; Suwarno et al. 2014b; Thieu et al. 2010; Van der Struijk & Kroeze 2010; Yasin et al. 2010; Zinia & Kroeze 2015) including China (Qu & Kroeze 2010; Qu & Kroeze 2012; Strokal et al. 2015; Strokal et al. 2014b). This model quantifies river export of different nutrients (nitrogen, phosphorus, carbon, silica) in different forms (dissolved inorganic, dissolved organic, particulate) as a function of human activities on land (e.g., agriculture, sewage) and basin characteristics (e.g., hydrology, land use). This basin scale model has been used to analyze past (1970, 2000) and future (2030, 2050) trends (Mayorga et al. 2010; Seitzinger et al. 2010). NUFER was developed for China to quantify efficiencies of nutrients in the food chain at national (Bai et al. 2013; Hou et al. 2013; Ma et al. 2010; Ma et al. 2011) and provincial (Ma et al. 2012) scales over time (e.g., the period of 1980-2010 (Hou et al. 2013; Ma et al. 2012)). This model is also used to assess management options for efficient nutrient management in the food chain (Ma et al. 2013a; Ma et al. 2013b).

We developed a sub-basin version of the Global *NEWS*-2 model in which we included information from NUFER (see Text S4.1 for extended Methods in Supplementary Materials). Novel aspects of our modeling approach include (i) the sub-basin scale at which we run Global *NEWS*-2 to quantify total dissolved nitrogen (TDN) and phosphorus (TDP) inputs to Chinese rivers by source, and (ii) the coupling of this sub-basin version of Global *NEWS*-2 with NUFER. The latter we did to include direct discharge of animal manure as a point source of nutrients in rivers in our modeling to analyze the consequences of agriculture transitions on river quality in China. This has not been done before. TDN and TDP include both dissolved inorganic and organic forms of N and P.

To assess the consequences of agricultural transition on water quality, we modeled TDN and TDP inputs to rivers for 1970 and 2000. We selected 1970 to reflect the pre-transition period and 2000 to reflect the ongoing transitions in Chinese agriculture. We also modeled TDN and TDP inputs to rivers for 2050 as an illustrative example to show the risks for future river pollution.

Our model includes point ($RSpnt_{F.total.j}$, kg, see equation 4.1) and diffuse ($RSdif_{F.total.j}$, kg, see equation 4.2) sources of nutrients in rivers. Point sources of the nutrients (F: TDN and TDP) include direct discharges of animal manure to rivers ($RSpnt_{F.ma.j}$, kg) and human sewage ($RSpnt_{F.sew.j}$, kg). Diffuse sources of the nutrients (F: TDN and TDP) include manure ($RSdif_{F.ma.j}$, kg) and synthetic fertilizers ($RSdif_{F.fe.j}$, kg) used in croplands, atmospheric N deposition (only for TDN; $RSdif_{F.dep.j}$, kg), biological N fixation (only for TDN; $RSdif_{F.fix.j}$, kg), leaching of organic matter (for dissolved organic N and P as a function of runoff) ($RSdif_{F.lch.j}$, kg), and P-weathering (only for TDP; $RSdif_{F.wth.j}$, kg).

$$RSpnt_{F.total.j} = RSpnt_{F.ma.j} + RSpnt_{F.sew.j} \tag{4.1}$$

$$RSdif_{F.total.j} = RSdif_{F.ma.j} + RSdif_{F.fe.j} + RSdif_{F.dep.j} + RSdif_{F.fix.j} + RSdif_{F.lch.j} + RSdif_{F.wth.j} \tag{4.2}$$

TDN ($RSpnt_{TDN.ma.j,}$ kg) and TDP ($RSpnt_{TDP.ma.j,}$ kg) inputs to rivers of sub-basin j from the manure point source were calculated as:

$$RSpnt_{TDN.ma.j} = Nexc_j \cdot frN_{sw.j} \tag{4.3}$$

$$RSpnt_{TDP.ma.j} = Pexc_j \cdot frP_{sw.j} \tag{4.4}$$

Here $Nexc_j$ and $Pexc_j$ are the N and P animal excretion in each sub-basin (j) (kg), respectively. We used gridded Global *NEWS*-2 information: 0.5 longitude by 0.5 latitude; i.e. cell areas ranging from 2135 km^2 to 2868 km^2 for the study area. Gridded information includes animal manure excretion corrected for losses of N during storage and housing (see Figure S4.2) (Mayorga et al. 2010). These values were originally derived from the IMAGE (Integrated Model to Assess the Global Environment) model (Bouwman et al. 2005). Bouwman et al. (2009) prepared these inputs for Global *NEWS*-2. IMAGE first calculated country-based manure production based on animal stocks and excretion rates, and then spatially allocated this manure over grids using land use maps (see details in (Bouwman et al. 2005; Bouwman et al. 2009)). We used this gridded information to calculate animal manure for sub-basins (see Figure S4.3 for values) using ArcGIS functions. We used these values for P excretion ($Pexc_j$). We calculated N excretion ($Nexc_j$) from the available N animal manure from Global *NEWS*-2 while accounting for N losses to the air (these losses were derived from NUFER, see Supplementary Materials).

$frN_{sw.j}$ and $frP_{sw.j}$ are the fractions (0-1) of N and P in animal excretion that are discharged directly to surface waters (sw) of the sub-basin (j), respectively. These fractions were calculated for each sub-basin from provincial data by NUFER (Ma et al. 2012) (Figure S4.4) using ArcGIS (area-weighted averages). Thus the fraction of manure that is directly discharged to surface waters of a sub-basin is $\sum_i$ (fraction of area covered by province i · fraction of manure directly discharged for province i). Fractions of manure discharged for provinces were taken from NUFER for 1980 and 2005 because

NUFER includes those years. We assume that 1980 is close to 1970 and 2005 is close to 2000 in our study. In NUFER, discharges of N and P from animal manure to surface waters were calculated from manure excretion by correcting for losses during storage and housing, and for applications to cropland and to grassland (see details in Ma et al. (2012)). Manure excretion was calculated based on animal feed from crop and kitchen residues, animal stocks (e.g., pigs, layer and broiler poultry, milk and beef cattle, sheep and goat), and excretion rates for N and P (see Text S4.1 for sources of information in the Supplementary Materials). In NUFER, animal production (see details in Ma et al. (2012)) includes two intensities of animal breeding, reflecting traditional and industrial farming systems. The high intensity types of animal breeding are defined for pig systems (> 50 heads), dairy cattle (> 5 heads), beef cattle (> 50 heads), layer poultry (>500 heads), and meat poultry (> 2000 heads) (Figure S4.5 as an example).

Nutrient inputs to rivers from sewage and diffuse sources were quantified following the modeling approaches of Global *NEWS*-2 (Mayorga et al. 2010) (summarized in Text S4.1 in the Supplementary Materials). Most model inputs were derived from gridded global datasets (0.5 longitude by 0.5 latitude) developed for Global *NEWS*-2 (Bouwman et al. 2009; Fekete et al. 2010; Van Drecht et al. 2009) for 1970, 2000 and 2050, and most of the model parameters are also from this model (Mayorga et al. 2010) (Figure S4.2). Inputs for 2050 were based on earlier studies (Bouwman et al. 2009; Fekete et al. 2010; Van Drecht et al. 2009) which are based on interpretations of the storylines of the MEA scenarios (Alcamo et al. 2005; Seitzinger et al. 2010). In this study we used model inputs that reflect a Global Orchestration (GO) scenario assuming globalized trends towards socio-economic development and reactive management of environmental problems. This scenario was implemented in the sub-basin Global *NEWS*-2 model as a starting point (Mayorga et al. 2010; Strokal et al. 2014a). The total population is assumed to have increased slightly by 2050 because of better education (e.g., low mortality and fertility rates, and high migration). However, urban population is assumed to have increased quickly. This reflects migration of people to cities to find better jobs (Van Drecht et al. 2009). The demand for animal production will have increased by 2050 and thus agriculture is projected to intensify for food security reasons (Bouwman et al. 2009). This existing scenario assumes that all available manure will be applied on land. However, this may not be likely. Considering the development of industrial farms today and the poor manure management it is more likely that direct discharges of animal manure to rivers will occur in the coming years without efficient manure management. Thus, in this study we assumed that direct discharges of manure to surface waters will still occur in 2050. Therefore, we used for 2050 the same fractions of manure directly discharged to rivers as for 2000 (see details in the Supplementary Materials).

4.2.2 Model performance

Model uncertainties are associated with model inputs, parameters and approaches. The original Global *NEWS*-2 was validated and calibrated for world rivers (Mayorga et al. 2010), indicating an acceptable performance according to the Nash-Sutcliffe model efficiency (R_{NSE}^2: 0.54, 0.51, 0.71, 0.90 for DIN, DIP, DON, and DOP, respectively). Strokal et al. (2014b) indicated a satisfactory performance of the model for DIN and DIP export by large Chinese rivers for 2000 (Pearson's coefficient of determination (R_P^2): 0.96; R_{NSE}^2: 0.42; model error (ME): 18%). Yan et al. (2010) validated the model for DIN export by the Yangtze River for 1970-2002 ($R_P^2 = 0.93$). Similar conclusions were drawn for the application of Global *NEWS*-2 to other world regions (see Text S4.1 for references). NUFER was developed based on validated statistical data, field surveys, literature, and has been widely applied in many studies (Bai et al. 2013; Hou et al. 2013; Ma et al. 2010; Ma et al. 2012; Ma et al. 2011). See details in Text S4.1 in the Supplementary Materials on evaluating the original Global *NEWS*-2 and NUFER.

We consider our model appropriate for modeling N and P inputs to the Chinese rivers at the sub-basin scale for three main reasons (see details in Text S4.1 for references). *First*, our modeled nutrient pollution levels in rivers are generally in line with observations. Our model captures the increasing trends in dissolved N and P inputs to the Chinese rivers since 1970, which is in agreement with other studies (e.g., (Li et al. 2007; Müller et al. 2008; Tao et al. 2010; Wang 2006)). *Second*, the comparison of model inputs with independent county data (Figure S4.6) convincingly shows that our model inputs for sub-basins are of good quality (e.g., R_P^2: 0.73-0.98 for studied rivers for N and P synthetic fertilizers and animal manure). *Third*, we verified relevant model parameters with experts and local information (e.g., nutrient removal during treatment in the Dongjiang basin, fractions of direct manure discharges from NUFER; see Supplementary Materials).

4.3 Results and discussion

Here we first discuss agricultural transitions in China. Next, we show consequences of agricultural transitions on river quality and discuss the risk for future river pollution by nutrients in China. We finish this section by comparing our results with existing studies.

4.3.1 Understanding agricultural transitions in China

Chinese agriculture has been in transitions since the 1980s (Herrero et al. 2013; Schneider 2011; Steinfeld et al. 2006). These transitions include changes in animal and crop production systems. Pre-1980, Chinese agriculture was dominated by small traditional farms with combined crop and animal production (Bai et al. 2013; Ju et al. 2005; Schneider 2011) (Figure 4.1 and Figure S4.7). Synthetic fertilizer was not widely used. Instead, farmers used most animal manure collected in confinements or from other places as a fertilizer (Ju et al. 2005). Thus direct discharge of manure to surface waters was minimal (Ju et al. 2005).

Since the 1980s Chinese agriculture has been industrializing to ensure food security (Figure 4.1) (Bai et al. 2013; Herrero et al. 2013; Ju et al. 2005; Schneider 2011). Crop production gradually separated from animal production in large parts of the country. This happened when the centrally planned economy shifted to a market-oriented economy (Ju et al. 2005; Schneider 2011). The human population increased over this period (Figure S4.9, Table S4.1). With the increasing prosperity and urbanization, the human diet shifted towards more meat consumption. This all led to an increasing demand for agricultural products (Ma et al. 2012). Synthetic fertilizers are now preferred over animal manure, because of their relatively low prices and low labor demand (Ju et al. 2005; Li et al. 2013) (Figure 4.1).

Animal production has thus shifted from traditional to industrial, land-less farming systems (Figure 4.1, Figure S4.7 and Figure S4.5). This allows for a higher production needed to meet the demand for meat in China, but also in the world (e.g., around half of the pork, and 18% of the poultry meat produced worldwide were from China in 2005) (Scanes 2007; Schneider 2011; Steinfeld et al. 2006). Between 1970 and 2000 the total number of pigs, poultry, cattle, sheep and goats increased between 1.5 to 8-fold (range for the different animal categories) in China. By 2000 10-40% of these animals, and by 2010 over two-thirds of the pigs and poultry were grown in industrial farms in China (Figure S4.7) (Bai et al. 2013; Ma et al. 2012). Pigs and poultry production accounts for over half of the total manure produced (Figure S4.5). Most of the produced manure is considered waste because industrial farms are largely disconnected from crop production, and often lack facilities to treat or recycle animal manure (Bai et al. 2013; Ju et al. 2005; Ma et al. 2012). Treatment is considered as separation of animal manure into solid and liquid fractions. The solid fraction after composting can be transported and applied to vegetable and fruit farms. The liquid fraction is further treated and then discharged ultimately to water systems. However, the separation and treatment of the liquid fraction is not very effective, and therefore large amounts of N and P are discharged to water systems (Figure 4.1, Ju et al. (2005)). For example, treatment ratios for animal waste in industrial farms are reported as (Ju et al. 2005): 3% for dairy cows, 10% for chicken and 43% for pigs. Both the treated and untreated manure from these systems are point sources rather than diffuse sources of nutrients in Chinese rivers.

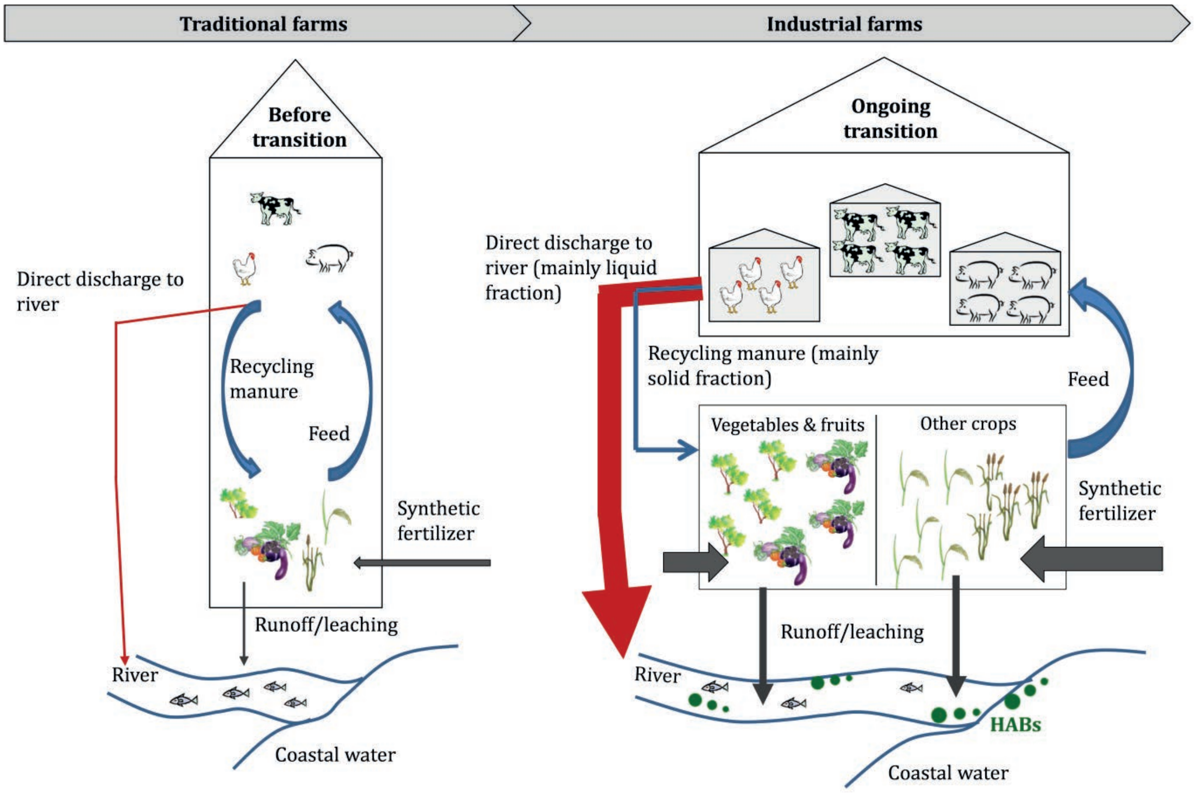

Figure 4.1. *A simplified historical overview of farming systems in China. This overview is shown in relation to direct nutrient losses to rivers, and their environmental impacts (Bai et al. 2013; Conley et al. 2009; Li et al. 2013; Ma et al. 2013b) including harmful algal blooms (HABs). The transition in Chinese agriculture started in the early-1980s.*

4.3.2 Consequences of agricultural transitions on river quality in China

Our results indicate that manure point sources have a much larger share in total TDN and TDP inputs to rivers than other nutrient sources (Figure 4.2 and 4.3, Figure S4.10). In 1970 less than 5% of the animal manure was directly discharged to rivers of the sub-basins as waste. In 2000 this had increased to 30-70% (Figure 4.2). As a result, the calculated basin area-weighted averaged TDN inputs to rivers from only animal manure (diffuse and point) increased 8-fold, from 156 kg per km^2 of basin area in 1970 to 1316 kg km^{-2} in 2000. The TDP inputs increased 22-fold, from 11 kg km^{-2} in 1970 to 243 kg km^{-2} in 2000. These inputs are mainly from direct discharges of manure to rivers (point source). These manure point sources are responsible for around half of the total TDN and 80% of TDP inputs to rivers in 2000, which is much higher than in 1970 (around 6% for TDN and 37% for TDP) (Table 4.1).

Table 4.1. *Area-weighted averaged total dissolved nitrogen (TDN) and phosphorus (TDP) inputs to Chinese rivers in 1970 and 2000 (kg km^{-2} of basin year^{-1}, and share of the animal manure point source in %). The range in brackets is the minimum and maximum values among the sub-basins. See the Supplementary Materials for model description and inputs. Other diffuse sources include biological N fixation, atmospheric N deposition, weathering of P-contained minerals, and N and P leaching from organic matter.*

Nutrient sources in rivers	TDN		TDP	
	1970	2000	1970	2000
Animal manure (point)	43 (0-173)	1148 (355-4338)	9 (0-34)	240 (59-952)
Animal manure (diffuse)	113 (0-941)	168 (1-781)	2 (0-33)	3 (0-15)
Synthetic fertilizers (diffuse)	77 (0-903)	547 (1-3529)	1 (0-19)	6 (0-30)
Other diffuse sources for agricultural areas	87 (0-345)	381 (4-1357)	1 (0-5)	4 (0-17)
Other diffuse sources for non-agricultural areas	379 (1-1891)	227 (0.2-1222)	4 (0-21)	2 (0-8)
Sewage effluents (point)	16 (0-172)	121 (0-837)	6 (0-64)	39 (0-272)
Total	715 (10-3921)	2593 (391-7737)	23 (1-156)	294 (68-1113)
The share of point animal manure (%)	6 (0-86)	44 (17-91)	37 (0-97)	82 (52-97)

The increases in TDN and TDP inputs to rivers and the shares of manure point sources differ largely among rivers and their sub-basins (Figure 4.3, Table 4.1). Sources of TDN and TDP in rivers include animal manure, synthetic fertilizers, atmospheric N deposition, biological N fixation, leaching of organic matter, weathering of P-minerals and human waste. Nutrient inputs to rivers have increased by a factor 2 to 45 for sub-basins between 1970 and 2000 (Figure 4.3). These increases are particularly large for the northern rivers, including the Yellow, Huai, Hai and Liao (Figure S4.10). For example, the total inputs of TDN to these northern rivers increased 11-fold, from 280 to 3000 kton between 1970 and 2000. TDP inputs increased 20-fold, from 27 to 546 kton (Figure S4.10). The sub-basin areas of these rivers are characterized by low precipitation and runoff (Tang et al. 2013). As a result, nutrient inputs from diffuse sources are typically low, and direct discharge of manure thus accounts for at least two-thirds of the TDN and over 80% of TDP inputs to these northern rivers (Figure 4.3). The remainder is from synthetic fertilizers or sewage. Animal manure use in cropland accounts for less than 4% of the nutrients in the northern rivers (Figure 4.3, Figure S4.10). More than half of the total nutrients are concentrated in rivers of the downstream Yellow delta, Huai and Hai basins (Figure 4.3).

For the central (Yangtze) and southern (Zhujaing) rivers we calculate an increase in TDN inputs from 2500 to about 7000 kton (a 2.8-fold increase), and in TDP from 62 to almost 600 kton (a 9.5-fold increase) between 1970 and 2000 (Figure S4.10). Manure point sources are responsible for 20-60% of these TDN and 50-97% of TDP inputs to rivers in 2000 (range for sub-basins; Figure 4.3). The remainder is mainly from synthetic fertilizers (sub-basin range: 12-46%), and atmospheric N deposition on land (sub-basin range: 12-40%). Animal manure as a diffuse source contributes less than 10% to TDN and TDP inputs to rivers. Sewage effluents are important sources of both TDN and TDP inputs particularly in river deltas.

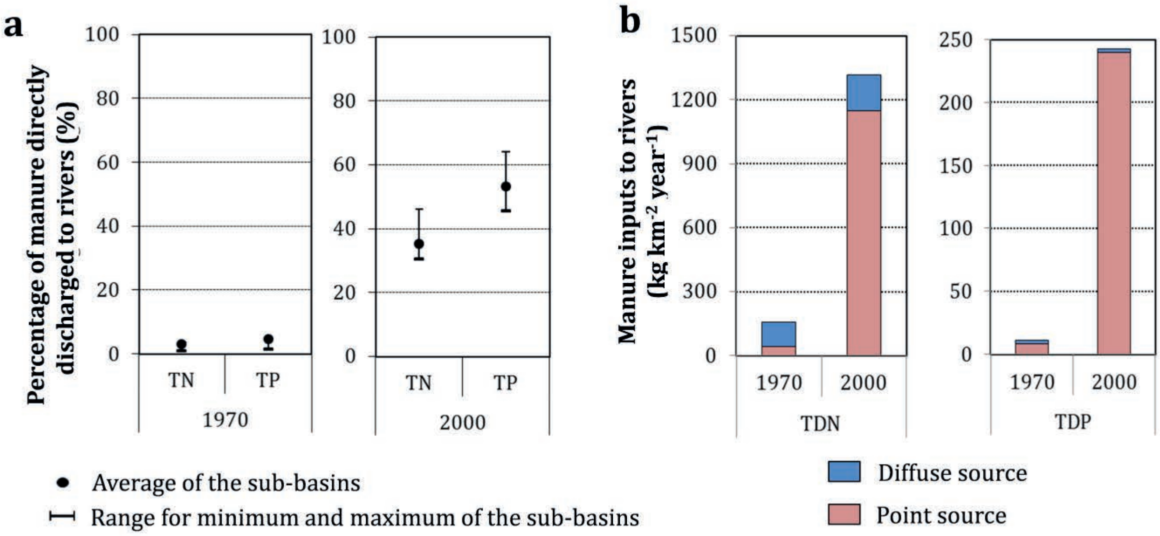

Figure 4.2. *Losses of nitrogen (N) and phosphorus (P) from animal manure to Chinese rivers in 1970 and 2000. (a) Percentage of the total nitrogen (TN) and phosphorus (TP) in animal manure excretion that is discharged directly to rivers in 1970 (the pre-transition period) and in 2000 (reflecting the ongoing transitions in farming systems) (%). (b) The area-weighted averaged inputs of total dissolved nitrogen (TDN) and phosphorus (TDP) from manure to rivers (kg km^{-2} year^{-1}) in China. Total areas were used to calculate area-weighted averaged inputs. Percentages of direct manure discharges to rivers are calculated from the provincial information of NUFER (Ma et al. 2012; Ma et al. 2013b). Animal manure production is from Global NEWS-2 (Mayorga et al. 2010) (see Figure S4.4 for sub-basin information).*

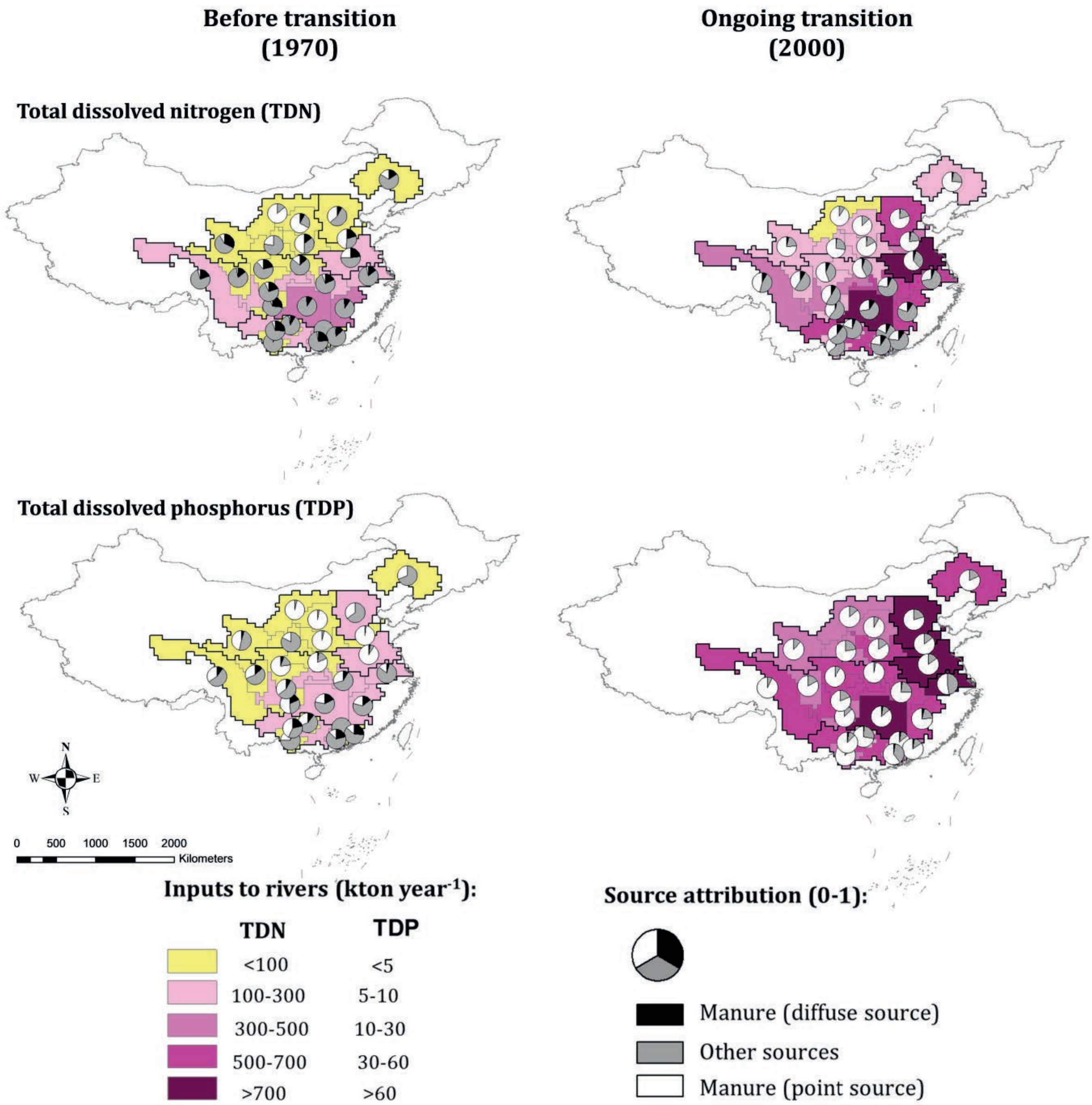

Figure 4.3. *Total dissolved nitrogen (TDN) and phosphorus (TDP) inputs to rivers by sub-basin in China in 1970 and 2000 (kton year^{-1}). The relative share of sources to these TDN and TDP inputs (source attributions in fractions) are shown in pie charts. Other sources include sewage effluents (TDN, TDP), synthetic fertilizer use (TDN, TDP), biological N fixation by agricultural crops and by natural vegetation, atmospheric N deposition (TDN), leaching of organic matter (TDN, TDP), and weathering of P-contained minerals (TDP) from agricultural and non-agricultural soils (see the Supplementary Materials for model descriptions and sources of model inputs, and Figure S4.10 for details on source attribution).*

4.3.3 Risks for future river pollution by nutrients

Nutrient pollution in Chinese rivers may increase in the coming years if future manure management stays as it is during the ongoing transition. Furthermore, the demand for animal products in China will remain high causing further development of industrial farms. We analyzed river pollution in 2050 as an example to illustrate the environmental consequences of future trends without improved manure management. Figure 4.4 illustrates to what extent TDN and TDP inputs to Chinese rivers may increase in 2050 compared to 2000 (reflect the ongoing transition). In 2050 the total inputs of TDN and TDP to Chinese rivers are calculated to be 8-325% higher (range for sub-basins) than in 2000, except for one sub-basin in the Zhujaing River (Figure 4.4). Manure point sources are projected to remain the main contributor to river pollution by nutrients in 2050 (22-91% of the nutrients in rivers are from this source, Table S4.2). In our scenario with a rapid economic development, the total area used for agriculture is projected to decrease between 2000 and 2050 in some sub-basins (up to 46%, Figure S4.11); this results from rapid urbanization (Table S4.1). Rivers may also receive nutrients from other sources. These are, for example, use of synthetic fertilizers to grow more crops, and sewage effluents from urbanized areas (Strokal et al. 2014b; Van Drecht et al. 2009). However, we calculate a lower contribution of these sources to nutrients in Chinese rivers (up to 40%) compared to manure point source (Table S4.2). Therefore, the risk for future river pollution by nutrients will largely depend on how food production will develop, in particular for animal production because of the large contribution of manure to water pollution currently.

4.3.4 Comparison with other studies

Some studies have already quantified N and P transport to waterbodies of the large Chinese rivers (Xing & Zhu 2002), in particular of the Yangtze River (Bao et al. 2006; Ding et al. 2014; Li et al. 2011b; Liu et al. 2008a; Xiao et al. 2007; Yan et al. 2010). A few studies exist on sub-basin analyses of the Yangtze River (Bao et al. 2006; Liu et al. 2008a; Xiao et al. 2007). However, these studies ignore direct discharges of manure to rivers. Therefore, the current nutrient pollution of rivers may be underestimated. For example, our estimates of the total TDN inputs (from all sub-basins) to the Yangtze are comparable with the results of Liu et al. (2008a) and Bao et al. (2006) for 1970 when direct discharges of animal manure were small, but higher than the results of Liu et al. (2008a) and Xing and Zhu (2002) for 2000 when direct discharges of manure were considerable. Liu et al. (2008a) and Bao et al. (2006) quantified around 2 Tg N transported to waterbodies of the Yangtze River in 1980, which is close to our estimate of 1.7 Tg TDN in 1970. For 2000 Liu et al. (2008a) quantified approximately 4.5 Tg of N, of which around 0.4 Tg N is from animal manure used in agriculture.

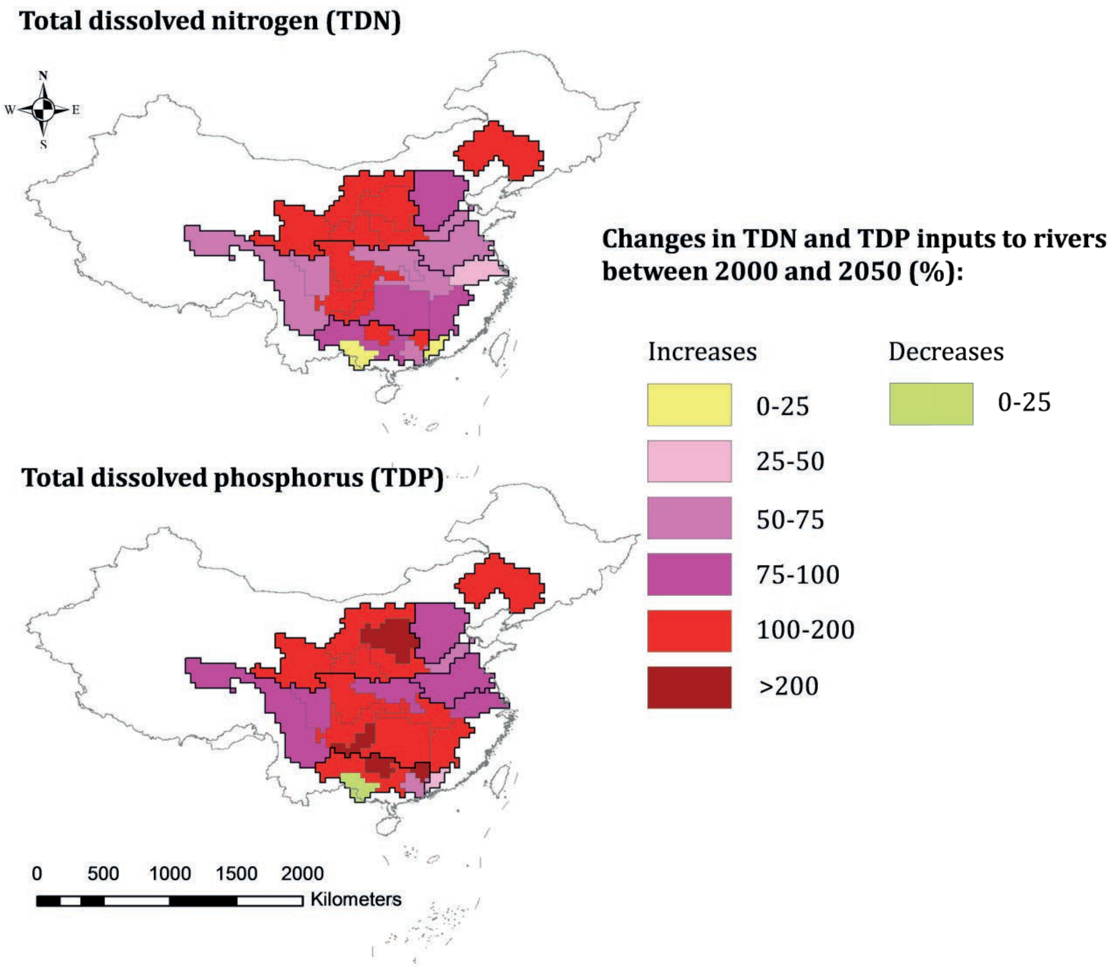

Figure 4.4. Risk for future river pollution by nutrients in China. The maps show changes in total dissolved nitrogen (TDN) and phosphorus (TDP) inputs to Chinese rivers between 2000 and 2050 (%). High risks for river pollution are indicated by increasing inputs of TDN and TDP to rivers in the future. See the Supplementary Materials on model description, inputs and scenario description for 2050.

We, however, calculated 5.3 Tg TDN, of which 2.2 Tg TDN from direct discharges of animal manure and 0.4 Tg N from animal manure use (Figure S4.10). We argue that the impact of direct discharges of animal manure from livestock production on river quality is much larger than the impact of over-fertilization of soils by synthetic fertilizers.

Few studies consider direct discharges of manure to rivers, but not at the sub-basin scale (Ma et al. 2010; Ma et al. 2012; Ti et al. 2012). For example, Ma et al. (2010) quantified that 30% of N and 45% of P from manure were discharged to surface waters in 2005 at the national scale. Ti et al. (2012) adopted a 25% value for livestock waste discharged to waterbodies to quantify N export by the Yangtze, Zhujiang and Yellow at the basin scale for the 2000s. We, however, demonstrate that this percentage is much higher for 2000 and ranges between 30% and 45% for N, and 46% and 64% for P among sub-basins (Figure 4.2).

Most existing modeling studies on river export of N and P emphasize the importance of managing diffuse agricultural sources of N (e.g., synthetic fertilizer use) and point sewage sources of P to reduce water pollution in China (e.g., Qu & Kroeze 2012; Strokal et al. 2014b; Yan et al. 2010). This is because these studies do not account for manure point sources. We show the importance of managing point manure sources for both N and P in the future. Studies on nutrient inputs to surface waters at the national (e.g., Bai et al. 2015; Ma et al. 2010) and provincial scales (e.g., Ma et al. 2012) acknowledge the importance of reducing losses of manure. Our study can contribute to such analyses by providing a better spatially explicit understanding of water pollution at the sub-basin scale.

4.4 Concluding remarks and future outlook

We consider our study a warning signal because nutrient loads in Chinese rivers are now much higher than previously and will continue to increase. Direct discharge of manure is likely the most important source of unexpected serious nutrient pollution in surface waters of China. This was not recognized in previous studies (Bao et al. 2006; Liu et al. 2008a; Ongley et al. 2010; Tao et al. 2010; Xu et al. 2014; Zhang et al. 2013). However, more experimental research is needed to reduce uncertainties (see Text S4.1 in Supplementary Materials). Industrialization of livestock production is common in other world regions such as Europe and North America (e.g., most of pork and poultry production are from industrial farms) (Herrero et al. 2013; Steinfeld et al. 2006). However, animal manure management in those regions is more effective than in China. Examples are permits to discharge manure to water bodies in the United States (Hribar 2010) and regulations to recycle manure in grassland and crop fields in Europe (Herrero et al. 2013; Oenema et al. 2007).

The Chinese government started to recognize the side-effects of livestock production, and introduced regulations such as the Discharge Standard of Pollution for Livestock Production, and the Law of Water Pollution Prevention (Zheng 2013). So far, however, these regulations have not been effective (Zheng 2013). Thus, the actual level of water pollution by nutrients from direct discharges of animal manure is today likely underestimated in modeling studies. The new “Environmental Protection Law”, “Zero-growth in Synthetic Fertilizer after 2020 Policy” and the new regulation for livestock production have been recently announced in 2015 with strict regulations for industrial animal production to reduce pollution (http://www.gov.cn). These regulations emphasize in particular recycling of animal manure on land to substitute synthetic fertilizers. However, the effect of these regulations on reducing water pollution will depend on the effectiveness of their implementation.

Meeting the demand for food will remain a challenge in the coming decades. Thus, industrial animal farms will likely continue to grow and increase in numbers. Nutrient pollution of aquatic systems will likely become more serious (see Figure 4.4 as an example), unless manure management is strongly improved. Besides environmental policies, providing better technologies for manure recycling and treatment, for fertilizer application, and educating farmers and providing more services (scientific and advisory) to help farmers improve their practices, are urgently needed. Future studies of sustainable food production (more food with less environmental impacts) should account for recycling animal manure as fertilizers. This will reduce direct discharges of manure to rivers. Recent experiments (Chen et al. 2011; Chen et al. 2014; Ju et al. 2009; Miao et al. 2010) on integrated soil-crop system management (ISSM: higher crop yields with lower nutrient losses to the environment, Chen et al. (2011)) focused on synthetic fertilizers. The efficient use of animal manure may be a prerequisite for ISSM to effectively reduce nutrient losses to the environment while increasing crop yields (Chen et al. 2011). Such changes, however, imply a major shift in farming practices, and until this shift is made the high nutrient loads to Chinese rivers from animal production remain unprecedented in the world.

Acknowledgments

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Chapter 5.

The *MARINA* model (Model to Assess River Inputs of Nutrients to seAs): model description and results for China

Abstract

Chinese agriculture has been developing quickly towards industrial food production systems that discharge nutrient-rich wastewater into rivers. As a result, nutrient export by rivers has been increasing, resulting in coastal water pollution. We developed a Model to Assess River Inputs of Nutrients to seAs (*MARINA*) for China. The *MARINA* Nutrient Model quantifies river export of nutrients by source at the sub-basin scale as a function of human activities on land. *MARINA* is a downscaled version for China of the Global *NEWS*-2 (Nutrient Export from WaterSheds) model with an improved approach for nutrient losses from animal production and population. We use the model to quantify dissolved inorganic and organic nitrogen (N) and phosphorus (P) export by six large rivers draining into the Bohai Gulf (Yellow, Hai, Liao), Yellow Sea (Yangtze, Huai) and South China Sea (Pearl) in 1970, 2000 and 2050. We addressed uncertainties in the *MARINA* Nutrient Model. Between 1970 and 2000 river export of dissolved N and P increased by a factor of 2-8 depending on sea and nutrient form. Thus, the risk for coastal eutrophication increased. Direct losses of manure to rivers contributed to 60 - 78% of nutrient inputs to the Bohai Gulf and 20-74% of nutrient inputs to the other seas in 2000. Sewage is an important source of dissolved inorganic P, and synthetic fertilizers of dissolved inorganic N. Over half of the nutrients exported by the Yangtze and Pearl rivers originated from human activities in downstream and middlestream sub-basins. The Yellow River exported up to 70% of dissolved inorganic N and P from downstream sub-basins and of dissolved organic N and P from middlestream sub-basins. Rivers draining into the Bohai Gulf are drier, and thus transport fewer nutrients. For the future we calculate further increases in river export of nutrients. The *MARINA* Nutrient Model quantifies the main sources of coastal water pollution for sub-basins. This information can contribute to formulation of effective management options to reduce nutrient pollution of Chinese seas in the future.

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5.1 Introduction

China has been becoming more urbanized with more industrial farms for animal production (Bai et al. 2013; Bai et al. 2014; Liu & Diamond 2005). However, these activities lead to discharges of nutrients, such as nitrogen (N) and phosphorus (P) to rivers. In particular, direct discharges of animal manure to rivers contribute largely to the nutrient pollution of rivers. This is a result of poor manure management, and of industrial animal production being disconnected from crop production (see Section 5.3.1, Strokal et al. (2016b)). In China, industrial animal farms started to emerge since the 1990s as a response to the demand for animal products, especially in urban areas (Liu & Diamond 2005). In 2000, 20-40% of all pigs, chickens and dairy cows in China were grown in industrial farms. By 2010 this number increased up to 20-80% (Bai et al. 2013; Bai et al. 2014; FAO 2014; MOA 2011b). However, animal manure is often ignored as a fertilizer in crop production (Bai et al. 2015; Ma et al. 2012; Strokal et al. 2016b). Meanwhile, farmers use large amounts of synthetic fertilizers to grow crops (Li et al. 2013). As a result, considerably amounts of animal manure enters rivers, resulting in nutrient pollution (Strokal et al. 2016b). Urbanization may also pollute aquatic systems (Ju et al. 2005; Ma et al. 2012; Miao et al. 2010; Xing & Yan 1999). The extent to which depends on the number of people connected to sewage systems and on the effectiveness of nutrient removal during treatment. Chinese seas, the Bohai Gulf, Yellow Sea and South China Sea, receive a lot of N and P from rivers. The large Chinese rivers contribute to the nutrient loads to the seas (Tong et al. 2015). This may result in coastal eutrophication and thus in blooms of harmful algae. Several studies report high measured concentrations of N and P in the seas (Liu et al. 2009b; Müller et al. 2008; Sumei et al. 2008; Xu et al. 2014; Yu et al. 2011). Xiao et al. (2007) indicated that 120 events of algae blooms were reported in the 2000s while less than five events occurred in the 1970s. Some species of harmful algae can be toxic for people (e.g., cyanobacteria), making water unsuitable for, for example, recreation. Blooms of harmful algae can also be a threat for living organisms such as fish because of reduced oxygen concentrations.

The environmental impact of the trends in the industrialization of agriculture and in the urbanization on Chinese seas is, however, not well studied in a quantitative, spatially explicit way. Existing studies acknowledge the adverse-effects of animal production and urbanization on aquatic pollution, but most of the time at the national scale (Bai et al. 2015; Bai et al. 2014; Hou et al. 2013; Ju et al. 2005; Liu & Diamond 2005; Ma et al. 2010; Ma et al. 2013b; Maimaitiming et al. 2013; MEP et al. 2010; Schneider 2011; Steinfeld et al. 2006). Provincial analyses exist for different years using the NUFER model (NUtrient flows in Food chains, Environment and Resources use) (e.g., Ma et al. 2012). However, these provincial analyses are limited to N and P losses from agriculture to surface waters. For example, Ma et al. (2012) quantified 30-48% of N (36% for China) and 43 - 65% of P (53% for China) discharged directly from animal manure to surface waters among provinces in 2005. They indicate that these manure discharges resulted from poor manure management and increasing numbers of industrial farms that do not often recycle manure on land and have poor treatment facilities. However, these provincial studies do not quantify impacts of the direct manure discharges on Chinese seas.

Various studies exist on basin-specific analyses of N and P export by Chinese rivers for the past and future using the Global *NEWS*-2 model (Nutrient Export from WaterSheds) (Li et al. 2011b; Qu & Kroeze 2010; Qu & Kroeze 2012; Strokal et al. 2015; Strokal et al. 2014b; Yan et al. 2010). These studies consider impacts of human activities on coastal water pollution by nutrients. These human activities are, for example, use of fertilizers (synthetic and organic) in crop production and urbanization. However, industrialization of animal production is not accounted for in those studies. Thus, direct discharges of animal manure to Chinese rivers are not considered. Likewise, human waste from unconnected sewage population is not considered either. This can also be a considerable source of nutrients in Chinese rivers (Ju et al. 2005; Ma et al. 2012; Miao et al. 2010; Xing & Yan 1999). As a result, these basin-specific studies may underestimate the nutrient pollution of Chinese seas and thus overestimate the effectiveness of the proposed management options to reduce this pollution in the future.

We believe that sub-basin scale analyses can help to identify areas with human activities that contribute largely to the nutrient pollution of Chinese seas. This is especially important for Chinese rivers with large drainage areas (e.g., Yangtze, Yellow and Pearl) because they may discharge large amounts of nutrients (e.g., Tong et al. 2015). A few sub-basin studies exist for the Yangtze river (Bao et al. 2006; Liu et al. 2008a; Xiao et al. 2007) and for the Pearl River (Strokal et al. 2015), but they do not account for direct manure discharges. Strokal et al. (2016b) analyzed recently the impact of direct manure discharges on inputs of nutrients to Chinese rivers at the sub-basin scale. However, this impact was not addressed on the Chinese seas.

Thus, we need a model that integrates existing approaches and information to quantify N and P export by large rivers to the Chinese seas while accounting for important sources of N and P in rivers. Furthermore, this model is needed to identify locations of sources contributing largely to sea pollution in particular for the largest basins in China (Section 5.2). Such information is important to identify and allocate effective nutrient management to reduce nutrients in the Chinese rivers, and thus coastal eutrophication.

In this study we integrated existing approaches and information into a Model to Assess River Inputs of Nutrients to seAs (*MARINA*) for China. The *MARINA* Nutrient Model quantifies river export of dissolved N and P by source at the sub-basin scale for the past and future. We used modeling approaches of Global *NEWS*-2 as the basis because this model quantifies nutrient export from land to sea in China. We used our previous study (Strokal et al. 2016b) as a basis for our modeling of direct manure discharges to Chinese rivers, but extended them with other missing sources (e.g., human waste from unconnected populations) by integrating approaches and information from Ma et al. (2012) and Morée et al. (2013). To account for locations of nutrient sources within the largest basins, we improved and implemented our previously developed sub-basin modeling approach, which was tested for the Pearl River basin (Strokal et al. 2015). Our model provides quantitative information on the main sources contributing to pollution of the Chinese seas and the locations of these sources over time. This has not been done before for large basins in China. We also evaluate the coastal eutrophication associated with the river export of nutrients for past and future. Below we introduce the study area, following by model description and evaluation (Section 5.2). Next, we present and discuss the model results (Section 5.3) and conclude main findings (Section 5.4).

5.2 Methodology

5.2.1 The study area

We studied six large Chinese rivers draining into the Bohai Gulf, Yellow Sea and South China Sea (Figure 5.1). The Yellow (Huanghe in Chinese), Liao and Hai rivers are located in northern China and drain into the Bohai Gulf. The Yangtze (Changjiang in Chinese) and Huai rivers are located in the middle of China and drain into the Yellow Sea. And the Pearl River (Zhujiang in Chinese) discharges to the South China Sea. The total drainage area of these river basins covers around 4 million km^2 (approximately 40% of China). The Yangtze, Yellow and Pearl are the largest rivers in China with drainage areas of 1.79, 0.89 and 0.44 million km^2, respectively. The Liao, Hai and Huai basins cover 0.27, 0.25 and 0.24 million km^2, respectively.

We divided the drainage basins of the Yellow, Yangtze and Pearl rivers in sub-basins (Figure 5.2). These sub-basins were classified as upstream, middlestream and downstream. This was done to better understand the spatial variations in human activities within the largest basins in China, and the impact of human activities on coastal pollution by nutrients. The Yellow River basin was divided into six sub-basins (Huang et al. 2009a; Wang et al. 2010). These sub-basins were named after monitoring (or hydrological) stations or after rivers located at their outlets. Lanzhou and Toudaogual are upstream sub-basins covering half of the Yellow basin. Longmen, Wehe and Huayuankou are middlestream and cover around 40% of the Yellow basin. The Delta sub-basin is located downstream and occupies less than 10% of the basin.

The Yangtze basin was divided into 10 sub-basins that were named following Yang and Lu (2014b) and Zhou et al. (2013) (Figure 5.2). Jinsha, Min, Wu and Jialing are sub-basins with tributaries draining into the Upper stem sub-basin. These upstream sub-basins cover around half of the Yangtze basin. Han, Dongting and Poyang are middlestream sub-basins with tributaries that drain into the Middle stem sub-basin. These cover around 40% of the basin area. The remainder (<10%) is covered by the Delta sub-basin located in downstream areas of the Yangtze.

The Pearl basin has six sub-basins (Cui et al. 2007; Niu & Chen 2010) that were divided into up-, middle- and downstream according to Strokal et al. (2015) (Figure 5.2). Liujiang and Yujiang are upstream tributaries and their drainage areas cover 30% of the Pearl basin. Xijiang and Beijiang are middlestream sub-basins covering half of the basin. Delta and Dongjiang are downstream sub-basins and their size equals 20% of the total basin area.

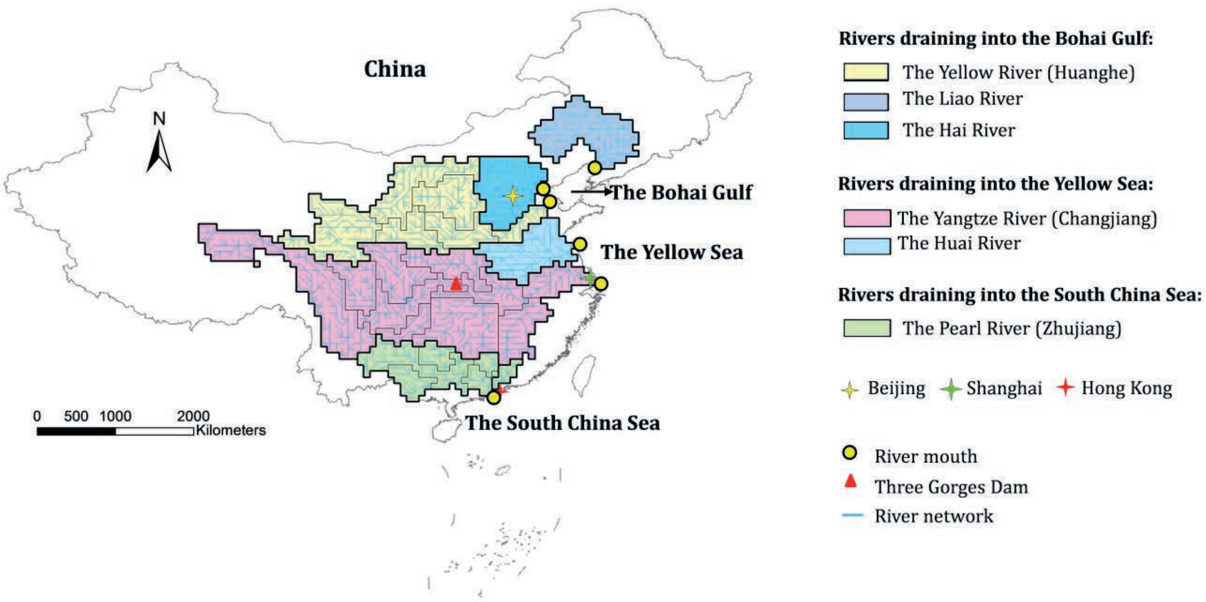

Figure 5.1. *Map of Chinese rivers that drain into the Bohai Gulf, Yellow Sea and South China Sea. Drainage areas of the rivers are from the Simulated Topological Network (STN-30, v6.01) (Mayorga et al. 2010; Vörösmarty et al. 2000b). The boundary of China is from GADM (2012).*

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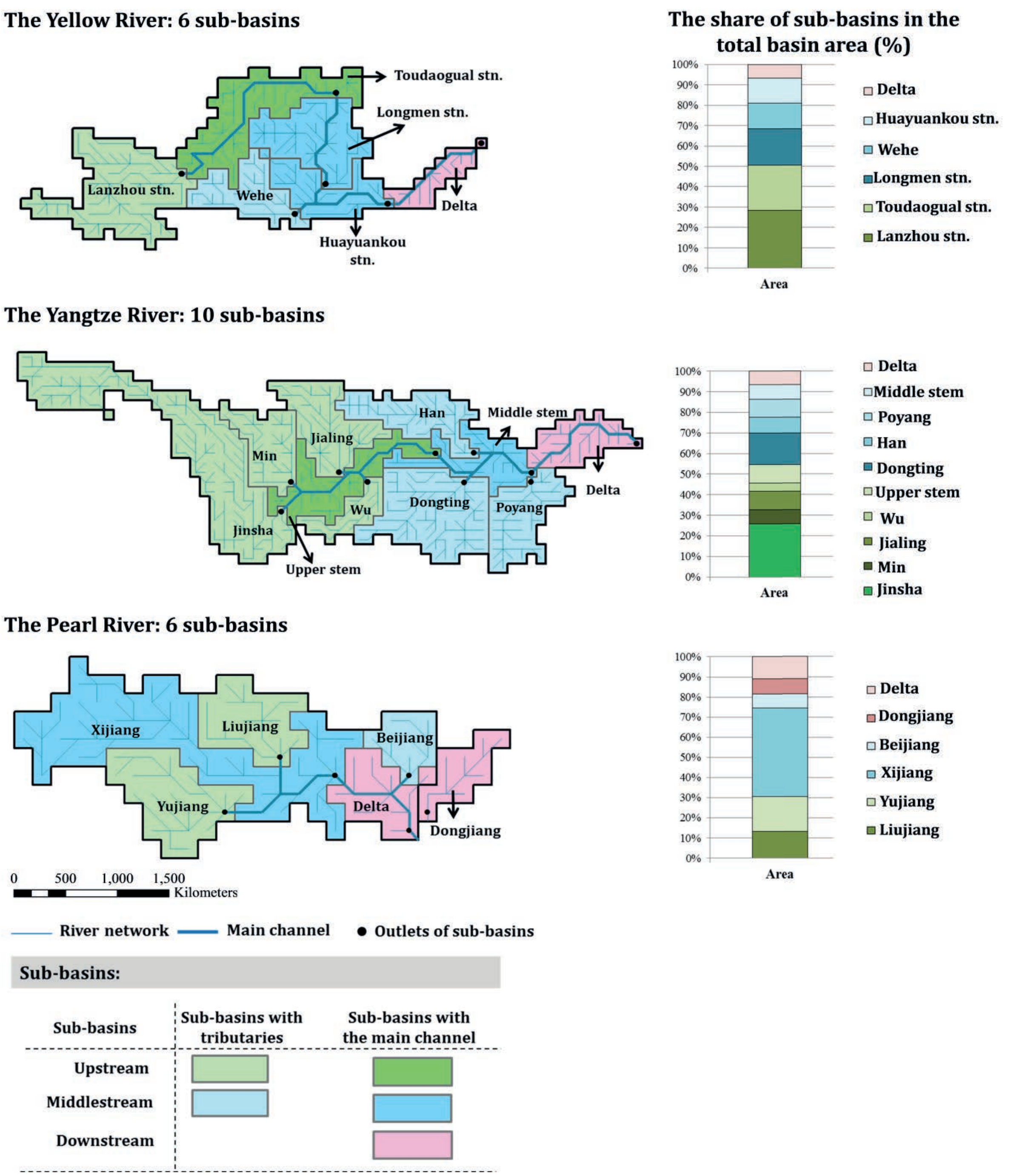


Figure 5.2. *Sub-basins of the Yellow, Yangtze and Pearl rivers, and the share of the sub-basins in the total basin area. See Figure 5.1 for locations of the rivers in China. Names of the sub-basins refer to existing names of outlets or to monitoring (hydrological) stations (indicated as stn. on the maps; see Section 5.1). Downstream sub-basins are named after their deltas. Outlets of the downstream sub-basins are the river mouths: the point where nutrients are discharged into the sea. Drainage areas of the sub-basins are from the Simulated Topological Network (STN-30, v6.01) (Mayorga et al. 2010; Vörösmarty et al. 2000b).*

5.2.2 Modeling river export of nutrients by source at the sub-basin scale

We integrated existing modeling approaches and sources of information into a new Model to Assess River Inputs of Nutrients to seAs (*MARINA*) for China. The *MARINA* Nutrient Model quantifies annual river export of nutrients by source at the sub-basin scale. We used the Global *NEWS*-2 model (Mayorga et al. 2010) as a starting point, but redesigned it largely for the six Chinese rivers. We started from quantifying nutrient inputs to the Chinese rivers at the sub-basin scale while accounting for direct manure discharges. The results are recently published in Strokal et al. (2016b).

Here we improved the previous study by accounting for other missing sources of nutrients in Chinese rivers namely human waste from unconnected sewage populations. We did this by using approaches and information from Ma et al. (2012) and Morée et al. (2013). We developed a sub-basin modeling approach (Strokal et al. 2015) with updated reservoir information (Lehner et al. 2011a). We published a first version of this sub-basin approach for the Pearl River for dissolved inorganic N and P (Strokal et al. 2015). In the current study we improved the approach, and extended it to other Chinese basins and other nutrient forms. Below we summarize model description and inputs, with a focus on the differences from the original Global *NEWS*-2 approach. Supplementary Materials provide details on model equations and inputs, and additional information on model outputs.

The *MARINA* Nutrient Model quantifies river export of N and P in dissolved inorganic (DIN, DIP) and organic (DON, DOP) forms for 1970, 2000 and 2050 by sub-basin (see Section 5.2.1 and Figure 5.2 for the sub-basins). The amount of nutrients exported by rivers to Chinese seas depends on human activities on land (e.g., agriculture, sewage), hydrology, and basin characteristics (e.g., land use). Important factors are retention and losses of nutrients in soils and rivers, and traveling distance of nutrients to the river mouth (coastal waters).

The *MARINA* Nutrient Model quantifies river export of nutrients in three steps. First, inputs of nutrients from diffuse or point sources to surface waters are quantified. Second, the river export of nutrients to the outlet of each sub-basin is quantified. Third, export of nutrients from sub-basin outlets to river mouths (the point where nutrients are discharged into the sea) is quantified (Figure 5.3). The Hai, Huai and Liao rivers do not have sub-basins and drain directly into coastal waters.

The overall equation to quantify export of nutrient form (F: DIN, DON, DIP, and DOP) to the river mouth ($M_{F.y.j}$, kg) by source y from sub-basin j is:

$$M_{F.y.j} = RS_{F.y.j} \cdot FE_{riv.F.outlet.j} \cdot FE_{riv.F.mouth.j} \qquad (5.1)$$

Here $RS_{F.y.j}$ is inputs of nutrient form F to surface waters from source y in sub-basin j (kg year^{-1}). In our model we distinguish between diffuse ($RSdif_{F.y.j}$) and point ($RSpnt_{F.y.j}$) sources (Box S5.1 and Table S5.1). $FE_{riv.F.outlet.j}$ is the fraction of $RS_{F.y.j}$ exported to the outlet of sub-basin j (0-1, Box S5.2 and Table S5.2). $FE_{riv.F.mouth.j}$ is the fraction of $RS_{F.y.j} \cdot FE_{riv.F.outlet.j}$ exported to the river mouth (0-1, Box S5.3, Table S5.3 and Figures S5.1-S5.3). Text S5.1 provides extended information on *MARINA*.

Step 1: Quantifying $RS_{F.y.j}$ ($RSdif_{F.y.j}$ and $RSpnt_{F.y.j}$)

Diffuse sources ($RSdif_{F.y.j}$) of nutrients in rivers include use of synthetic fertilizers, animal manure and human excreta in agriculture (for DIN, DIP, DON and DOP), atmospheric N deposition on agricultural and non-agricultural areas (for DIN), biological N fixation by agricultural crops (e.g., legumes) and natural vegetation (for DIN), weathering of P-contained minerals (for DIP), and leaching of organic matter from agricultural and non-agricultural soils (for DON and DOP).

Nutrient inputs to Chinese rivers from these diffuse sources are quantified using Global *NEWS*-2 approaches (Bouwman et al. 2009; Mayorga et al. 2010), except for animal manure and human excretion. In Global *NEWS*-2, nutrient inputs to rivers from synthetic fertilizers, atmospheric N deposition and biological N fixation are quantified as a function of nutrient inputs to land from each source, corrected for nutrient removal via crop harvesting, animal grazing and soil retention. Nutrient inputs to rivers from P weathering and organic leaching are quantified as a function of annual runoff from land to stream. We summarized these approaches for our sub-basins in Text S5.1, Box S5.1 and Table S5.1. Model inputs for synthetic fertilizers, atmospheric N deposition, biological N fixation, and annual runoff for 1970, 2000 and 2050 were derived from gridded databases (0.5 latitude by 0.5 longitude) of Global *NEWS*-2 (Bouwman et al. 2009; Fekete et al. 2010; Mayorga et al. 2010). These gridded data were aggregated to sub-basins in ArcGIS, except for annual runoff (calculated using natural water discharges from outlets of sub-basins and their areas; details are in Figure S5.4, Tables S5.4-S5.9).

For animal manure we improved Global *NEWS*-2 approaches (Bouwman et al. 2009; Mayorga et al. 2010) as explained in our earlier study (Strokal et al. 2016b). Global *NEWS*-2 only accounts for diffuse inputs of manure in rivers. Here, we also account for point source inputs, which can be considerable (see equation 5.4 below). To this end, we used information from the NUFER model of Ma et al. (2012) (see details in Strokal et al. (2016b)). Thus, inputs of nutrient form (F: DIN, DON, DIP, and DOP) to surface waters from animal manure use in agriculture in sub-basin j ($RSdif_{F.ma.j}$, kg year^{-1}) are quantified (Box S5.1 and Table S5.1) as:

$$RSdif_{F.ma.j} = WSdif_{E.ma.j} \cdot G_{F.j} \cdot FE_{ws.F.j}$$
$$= [Eexc_{ma.j} \cdot (1 - (frE_{losses.ma.j} + frE_{sw.ma.j}))] \cdot G_{F.j} \cdot FE_{ws.F.j} \quad (5.2)$$

Here $WSdif_{E.ma.j}$ is inputs of nutrient element (E: N or P) to agricultural land from animal manure in sub-basin j (kg year^{-1}). $G_{F.j}$ is the fraction of nutrient (F) applied to agricultural land that is retained in soils after animal grazing and crop harvesting in sub-basin j (0-1). $G_{F.j}$ was calculated as the 1 – [the amount of element (N or P) that is exported from soils by harvesting and grazing (kg year^{-1}) / the total inputs of element (N or P) to agricultural soils from all diffuse sources (kg year^{-1})]. The required model inputs for 1970, 2000 and 2050 were aggregated from the gridded data (0.5 latitude by 0.5 longitude) of Global *NEWS*-2 (Bouwman et al. 2009; Mayorga et al. 2010) to sub-basins in ArcGIS (Figure S5.4 and Table S5.4). $FE_{ws.F.j}$ is the export fraction of nutrient form (F) entering surface waters in sub-basin j (0-1). This fraction is calculated as a function of annual runoff from land to streams. The fraction accounts implicitly for retention of nutrients in soils during their transport to rivers (e.g., transformation to unreactive forms, temporary accumulation, retentions in groundwater, riparian zones, (Mayorga et al. 2010)). $Eexc_{ma.j}$ is nutrient element (E: N or P) in animal excretion in sub-basin j (kg year^{-1}). Animal excretion equals the animal manure available for application to agricultural soils plus (only for N) ammonia losses during storage and housing. Animal manure available for application in 1970, 2000 and 2050 was from the gridded database (0.5 latitude by 0.5 longitude) of Global *NEWS*-2 (Bouwman et al. 2009; Mayorga et al. 2010), aggregated to sub-basins in ArcGIS (Figure S5.4 and Table S5.4). $frE_{losses.ma.j}$ is the fraction of nutrient losses to the air during animal manure storage and housing in sub-basin j (ammonia losses, 0-1). $frE_{sw.ma.j}$ is the fraction of nutrient element (E: N or P) in animal excretion that is directly discharged to surface waters in sub-basin j (0-1). $frE_{losses.ma.j}$ and $frE_{sw.ma.j}$ were derived from the provincial NUFER model of Ma et al. (2012) for 1970 and 2000. We aggregated these fractions from provinces to sub-basins by calculating area-weighted averages in ArcGIS (Figure S5.4 and Table S5.4). For 2050, we used values of 2000 for $frE_{losses.ma.j}$ and $frE_{sw.ma.j}$ assuming that the fraction of manure that directly discharges to rivers will not change largely in the coming years in China.

In China, human excreta are sometimes used as a fertilizer for vegetables and fruits. This holds in particular for rural areas (Ju et al. 2005; Ma et al. 2012; Miao et al. 2010; Xing & Yan 1999) lacking sewage connections (WHO/UNICEF 2014). This source is not accounted for in Global *NEWS*-2. Here we adjusted the Global *NEWS*-2. Nutrient inputs to surface waters from recycled human excreta in agriculture from unconnected populations ($RSdif_{F.hum.uncon,j}$, kg year^{-1}) were quantified (Box S5.1 and Table S5.1) as:

$$RSdif_{F.hum.uncon.j} = WSdif_{E.hum.uncon.j} \cdot G_{F.j} \cdot FE_{ws.F.j}$$
$$= [WSdif_{E.hum.uncon.rur.j} + WSdif_{E.hum.uncon.urb.j}] \cdot G_{F.j} \cdot FE_{ws.F.j} \quad (5.3)$$

Here $WSdif_{E.hum.uncon.j}$ is inputs of nutrient element (E: N or P) to agriculture from human excretion in sub-basin j (kg year^{-1}). $WSdif_{E.hum.uncon.rur.j}$ and $WSdif_{E.hum.uncon.urb.j}$ are nutrient element (E: N or P) in human excretion that is recycled in agriculture of sub-basin j from rural and urban population (not connected to sewage systems), respectively (kg year^{-1}). We defined urban population with and without sewage connections as in Morée et al. (2013). Rural population is defined as people without sewage connections and calculated as the total population minus urban population (with and without sewage connection). $WSdif_{E.hum.uncon.rur.j}$ is quantified by correcting rural human excretion for direct discharges to surface waters and for N losses to the air (ammonia losses). $WSdif_{E.hum.uncon.urb.j}$ is quantified by multiplying urban human excretion with the fraction that is recycled in agriculture after correcting for N losses to the air. We used the Global *NEWS*-2 approach and its gridded databases (0.5 latitude by 0.5 longitude) (Mayorga et al. 2010; Van Drecht et al. 2009) to quantify N and P human excretion from rural and urban population without a sewage connection (see Box S5.1, Figure S5.4 and Table S5.4 for details). The fraction of human excretion discharged to aquatic systems from rural people was available from Ma et al. (2010) for China. We applied this fraction to our sub-basins (Figure S5.4 and Table S5.6). The recycling fraction of urban human excretion in agriculture was derived from Morée et al. (2013) for China for 1970 and 2000. For 2050 we applied the fraction of 2000 (Figure S5.4 and Table S5.7).

<u>Point sources</u> ($RSpnt_{F.y.j}$) of nutrients in rivers include sewage effluents from human excretion (for DIN, DIP, DON, and DIP) and detergents (for DIP and DOP), and direct discharges of animal manure and human excretion (not connected to sewage systems) to surface waters (for DIN, DIP, DON and DOP). Global *NEWS*-2 only accounts for sewage inputs. Here we also quantify point source inputs of manure and direct discharge of human waste into rivers.

Nutrient inputs to rivers from sewage effluents are quantified following Global *NEWS*-2 (Mayorga et al. 2010; Van Drecht et al. 2009) as a function of urban excretion from population connected to sewage systems, corrected for nutrient removal during waste water treatment. Model inputs and parameters were from the gridded databases (0.5 latitude by 0.5 longitude) and basin information of Global *NEWS*-2 (see details in Box S5.1 and Table S5.1 for equations, Figure S5.4 and Table S5.4 for model inputs). An exception is the Dongjiang sub-basin of the Pearl River draining into Hong Kong (Figure 5.1). For this sub-basin we used the fraction of nutrient removal during treatment from Strokal et al. (2015).

Direct discharge of manure (as a point source) to rivers is a considerable contributor to nutrient pollution of aquatic systems in China. A reason for this is poor manure management and the on-going industrialization of animal farms that do not often recycle manure in cropland, and have poor waste treatment (Ju et al. 2005; Ma et al. 2012; Schneider 2011; Steinfeld et al. 2006) (see Sections 5.1 and 5.3.1). We included manure point source in our model by adjusting the Global *NEWS*-2 approach (Mayorga et al. 2010; Van Drecht et al. 2009) using information from Ma et al. (2012) for manure losses (see details in Strokal et al. (2016b), Box S5.1 and Table S5.1):

$$RSpnt_{F.ma.j} = RSpnt_{E.ma.j} \cdot FEpnt_{F.ma}$$
$$= [Eexc_{ma.j} \cdot frE_{sw.ma.j}] \cdot FEpnt_{F.ma} \quad (5.4)$$

Here $RSpnt_{E.ma.j}$ is point source inputs of nutrient element (E: N or P) to surface waters from manure in sub-basin j (kg year^{-1}). $FEpnt_{F.ma}$ is the fraction of element E (N or P) entering surface waters as form F (DIN, DON, DIP, and DOP) (0-1). This fraction was derived based on Huang et al. (2011), Jensen (2013), Li et al. (2014a), Schoumans (2015) (Table S5.9).

Direct discharges of human excretion to aquatic systems from unconnected sewage population can also contribute to water pollution in China (Ju et al. 2005; Ma et al. 2012; Morée et al. 2013; Xing & Zhu 2002). We accounted for this point source by adjusting the modeling approach of Global *NEWS*-2 and using information from Morée et al. (2013) for urban people and from Ma et al. (2012) for rural people Box S5.1 and Table S5.1) as:

$$RSpnt_{F.hum.uncon.j} = RSpnt_{E.hum.uncon.j} \cdot FEpnt_{F.hum.uncon}$$
$$= [RSpnt_{E.hum.uncon.rur.j} + RSpnt_{E.hum.uncon.urb.j}] \cdot FEpnt_{F.hum.uncon} \quad (5.5)$$

Here $RSpnt_{E.hum.uncon.j}$ is inputs of nutrient element (E: N or P) to surface waters from direct discharges of human excretion from unconnected population to sewage systems in sub-basin j (kg year^{-1}). $FEpnt_{F.hum.uncon}$ is the fraction of element E (N or P) input to rivers as form F (DIN, DON, DIP, and DOP) (0-1). This fraction was derived based on literature (Figure S5.4 and Table S5.9). $RSpnt_{E.hum.uncon.rur.j}$ and $RSpnt_{E.hum.uncon.urb.j}$ are inputs of nutrient element (E: N or P) to surface waters in sub-basin j from direct discharges of human excretion (not connected to sewage systems) from rural and urban population, respectively (kg year^{-1}). $RSpnt_{E.hum.uncon.rur.j}$ is quantified as N and P in human excretion (kg year^{-1}) multiplied by the fraction that is discharged to surface waters (0-1). This fraction accounts for N losses to the air (Ma et al. 2012). $RSpnt_{E.hum.uncon.urb.j}$ is quantified by correcting urban human excretion for N losses to the air and for recycling in agriculture (Morée et al. 2013).

Step 2: Quantifying $FE_{riv.F.outlet.j}$

The fraction of nutrient inputs to surface waters that reaches the outlet of sub-basin j ($FE_{riv.F.outlet.j}$, 0-1) is calculated following Global *NEWS*-2 (Box S5.2 and Table S5.2):

$$FE_{riv.F.outlet.j} = (1-D_{F.j}) \cdot (1-L_{F.j}) \cdot (1-FQrem_j) \quad (5.6)$$

Here $D_{F.j}$ is this fraction of nutrient form (DIN or DIP) retained in dammed reservoirs in sub-basin j (0-1), calculated using Global *NEWS*-2 approaches (Harrison et al. 2005b; Seitzinger et al. 2002). For 1970 and 2000, nutrient retention in each reservoir was calculated based on water residence time and depth (for DIN), using information from the Global Reservoir and Dam Database (GRanD) (Lehner et al. 2011a; Lehner et al. 2011b) (Figure S5.4 and Table S5.4). Then nutrient retentions of individual reservoirs were averaged for sub-basins using actual water discharges at the outlets of the sub-basins. Actual water discharges were from the gridded database of Global *NEWS*-2 (Fekete et al. 2010; Mayorga et al. 2010) (Figure S5.4 and Table S5.4). For 2050, we did not have information for individual reservoirs. Thus we multiplied sub-basin retentions for 2000 with a factor reflecting the increase between 2000 and 2050 following Strokal et al. (2015). This factor was calculated using basin retentions of 2000 and 2050 from Global *NEWS*-2 (Mayorga et al. 2010) (see details in Figure S5.4 and Table S5.4).

$L_{F.j}$ is the fraction of nutrient form (DIN or DIP) retained in or/and lost from water systems in sub-basin j (e.g., denitrification for DIN, sedimentation processes for DIP, 0-1). For DIN, this fraction was calculated as a function of sub-basin area, following Global *NEWS*-2 (Seitzinger et al. 2002). For DIP, we applied values based on Strokal et al. (2015) and literature (Figure S5.4 and Table S5.4).

$FQrem_j$ is the fraction of nutrients (generic for DIN, DIP, DON, and DOP) removed from sub-basin j for water consumption (0-1). This fraction was calculated as the ratio between the natural (before) and actual (after water is removed for consumption) water discharges at the outlet of sub-basin j. Equations for $D_{F.j}$, $L_{F.j}$ and $FQrem_j$ are presented in Box S5.2 and Table S5.2, and details on model inputs and parameters are in Figure S5.4 and Table S5.4.

Step 3: Quantifying $FE_{riv.F.mouth.j}$

We distinguished between sub-basins that include only tributaries (T) and sub-basins that include the main channel (C) of the river. We do this for up- (ju), middle- (jm) and downstream (jd) sub-basins of Yellow, Yangtze and Pearl rivers (Figure 5.3). Tributaries discharge nutrients to the main channel. The main channel transports these nutrients to the river mouth (Figure 5.3).

Fractions of nutrients exported from the outlets of upstream tributaries ($FE_{riv.F.mouth.juT}$, 0-1) and of upstream main channel ($FE_{riv.F.mouth.juC}$, 0-1) to the river mouth (Box S5.3, Table S5.3, Figures S5.1-S5.3) are calculated as follows:

$$FE_{riv.F.mouth.juT} = {}^{juT}FE_{riv.F.outlet.juC} \cdot {}^{juT}FE_{riv.F.outlet.jmC} \cdot {}^{juT}FE_{riv.F.outlet.jdC} \quad (5.7)$$

$$FE_{riv.F.mouth.juC} = {}^{juC}FE_{riv.F.outlet.jmC} \cdot {}^{juC}FE_{riv.F.outlet.jdC} \quad (5.8)$$

Here ${}^{juT}FE_{riv.F.outlet.juC}$, ${}^{juT}FE_{riv.F.outlet.jmC}$ and ${}^{juT}FE_{riv.F.outlet.jdC}$ are fractions of nutrient form (F) exported from the outlet of upstream tributary (upper case: juT) to the outlets of the main channel in upstream (lower case: juC), middlestream (lower case: jmC) and downstream (lower case: jdC) sub-basins (0-1). ${}^{juC}FE_{riv.F.outlet.jmC}$ and ${}^{juC}FE_{riv.F.outlet.jdC}$ are fractions of nutrient form (F) exported from the outlet of upstream sub-basin with the main channel (upper case: juC) to the outlets of the main channel in middlestream (lower case: jmC) and downstream (lower case: jdC) sub-basins (0-1).

Fractions of nutrients exported from the outlets of middlestream tributaries ($FE_{riv.F.mouth.jmT}$, 0-1) and of middlestream main channel ($FE_{riv.F.mouth.jmC}$, 0-1) to the river mouth (Box S5.3, Table S5.3, Figures S5.1-S5.3) are calculated as follows:

$$FE_{riv.F.mouth.jmT} = {}^{jmT}FE_{riv.F.outlet.jmC} \cdot {}^{jmT}FE_{riv.F.outlet.jdC} \quad (5.9)$$

$$FE_{riv.F.mouth.jmC} = {}^{jmC}FE_{riv.F.outlet.jdC} \quad (5.10)$$

${}^{jmT}FE_{riv.F.outlet.jmC}$ and ${}^{jmT}FE_{riv.F.outlet.jdC}$ are fractions of nutrient form (F) exported from the outlet of the middle tributary (upper case: jmT) to the outlets of the main channel in middlestream (lower case: jmC) and downstream (lower case: jdC) sub-basins (0-1). ${}^{jmC}FE_{riv.F.outlet.jdC}$ is the fraction of nutrient form (F) exported from the outlet of the middlestream sub-basin with the main channel (upper case: jmC) to the outlets of the main channel in downstream (lower case: jdC) sub-basins (0-1).

Downstream sub-basins have the main channel and nutrients from their outlets discharge directly to the river mouth. Thus $FE_{riv.F.mouth.jd}$ for downstream sub-basins is assumed to be one. This also holds for Hai, Huai and Liao.

All fractions indicated in equations 5.7-5.10 were quantified as in step 2 above, and account for nutrient retention in reservoirs ($D_{F.j}$) and losses ($L_{F.j}$) during river transport and as a result of consumptive water use ($FQrem_j$). To this end, we corrected sub-basin specific values for $D_{F.j}$, $L_{F.j}$ and $FQrem_j$ for the drainage area of the main channel exporting nutrients towards the river mouth (see Figures S5.1-S5.3). The main channel is defined based on the Strahler Order from the Simulated Topological Network (STN-30, v6.01) (Vörösmarty et al. 2000a; Vörösmarty et al. 2000b) implemented in Global *NEWS*-2 (Mayorga et al. 2010). For Chinese rivers, the main channel is often formed by streams with the Strahler Order between three (smaller streams) and five (larger streams). The drainage area of the main channel for each sub-basin was quantified from the STN-30 information (Vörösmarty et al. 2000a; Vörösmarty et al. 2000b). Box S5.3, Table S5.3 and Figures S5.1-S5.3 give details on equations and on values for the drainage areas of the main channel by sub-basin.

In summary, we used most of the model inputs for human activities in 1970, 2000 and 2050 from gridded databases (0.5 latitude by 0.5 longitude) of Global *NEWS*-2 that were prepared in earlier studies (Bouwman et al. 2009; Fekete et al. 2010; Mayorga et al. 2010; Van Drecht et al. 2009). For 2050 these gridded model inputs were prepared based on storylines of the four Millennium Ecosystem Assessment (MEA) scenarios (Alcamo et al. 2005; Seitzinger et al. 2010). We implemented one of the MEA scenarios (Global Orchestration: GO) as an illustrative example in the *MARINA* Nutrient Model.

The GO scenario illustrates the consequences of globalization and a reactive environmental management approach for nutrient pollution. Details on the storylines of GO are published in earlier studies (Alcamo et al. 2005; Seitzinger et al. 2010) for agriculture (Bouwman et al. 2009), sewage (Van Drecht et al. 2009) and hydrology (Fekete et al. 2010). Strokal et al. (2016b) describe GO for Chinese sub-basins that are studied here (see also Section 5.3.1). Strokal et al. (2016b) modified the original GO scenario (Alcamo et al. 2005; Seitzinger et al. 2010) by incorporating industrialization of animal production in China. In this study we used the GO of Strokal et al. (2016b) with some additional assumptions for human waste for 2050.

The GO scenario assumes that people will apply large amounts of synthetic fertilizers to produce crops to feed the increasing urban population (see Strokal et al. 2016b and Bouwman et al. 2009). Animal production will increase because of the increasing demand for food especially in cities. This will drive construction of large industrial animal farms. However, GO also assumes that in 2050 the management of animal manure will be of the same type as in 2000. This implies that large amounts of manure will be still discharged to rivers (as in 2000, see Section 5.3.1 and Strokal et al. 2016b). The GO scenario assumes a rapid urbanization for China (Van Drecht et al. 2009). The number of people connected to sewage systems will consequently increase. However, the efficiency of nutrient removal during treatment will be relatively low compared to the economic and technological development in China (secondary-oriented treatment with 40-50% of the removal efficiency, see Van Drecht et al. 2009). We assume that management of human waste from rural populations will remain as in 2000. This implies that rural people will be without a sewage connection, and thus some amounts of human waste will be used as fertilizers and the remainder will be discharged to rivers as waste. Urban waste that is not collected is assumed to be managed as in 2000 (see Section 5.3.1). GO projects northern sub-basins to be drier while middle- and southern sub-basins to be wetter in 2050 compared to 2000 (see Section 5.3.1 and Fekete et al. 2010). Increases in food production will drive increases for the demand for irrigation, especially in dry regions (e.g., northern areas in Figure 5.1). As a result, more dams are assumed to be constructed, leading to more nutrient retentions. This may decrease nutrient export to the sea.

The Chinese government has recently introduced a number of environmental policies to reduce adverse-effects of human activities, especially of animal production on water pollution. Examples are the Discharge Standard of Pollution for Livestock Production (2001), the Criteria for Evaluating Environmental Quality of Livestock Farms (2004) (Zheng 2013), and the first manure regulation for collective feedlots and industrial livestock farms, introduced in the beginning of 2014 (http://www.gov.cn/). The later regulation should be implemented at the county level. This regulation is strict in terms of selecting areas for new industrial farms with better treatment facilities and improving manure treatment in existing farms (if they are located in permitted areas, if not, then these farms should be removed). The regulation facilitates recycling of manure in crop production (e.g., building crop-livestock production systems). All these may reduce nutrient inputs to rivers from direct discharges of animal manure and thus improve water quality of the Chinese seas in the coming years. In our GO scenario we assumed that the fraction of manure that directly discharges to rivers will not change largely in the coming years in China (as mentioned in the previous paragraph). We did this to illustrate what amounts of nutrients can potentially be avoided by implementing effectively recent and future environmental policies. Results are shown in Section 5.3.1 for the characteristics of human activities at the sub-basin scale, and in Sections 5.3.2 and 5.3.3 for nutrient exports by rivers to the Chinese seas and their associated coastal eutrophication in the past and future.

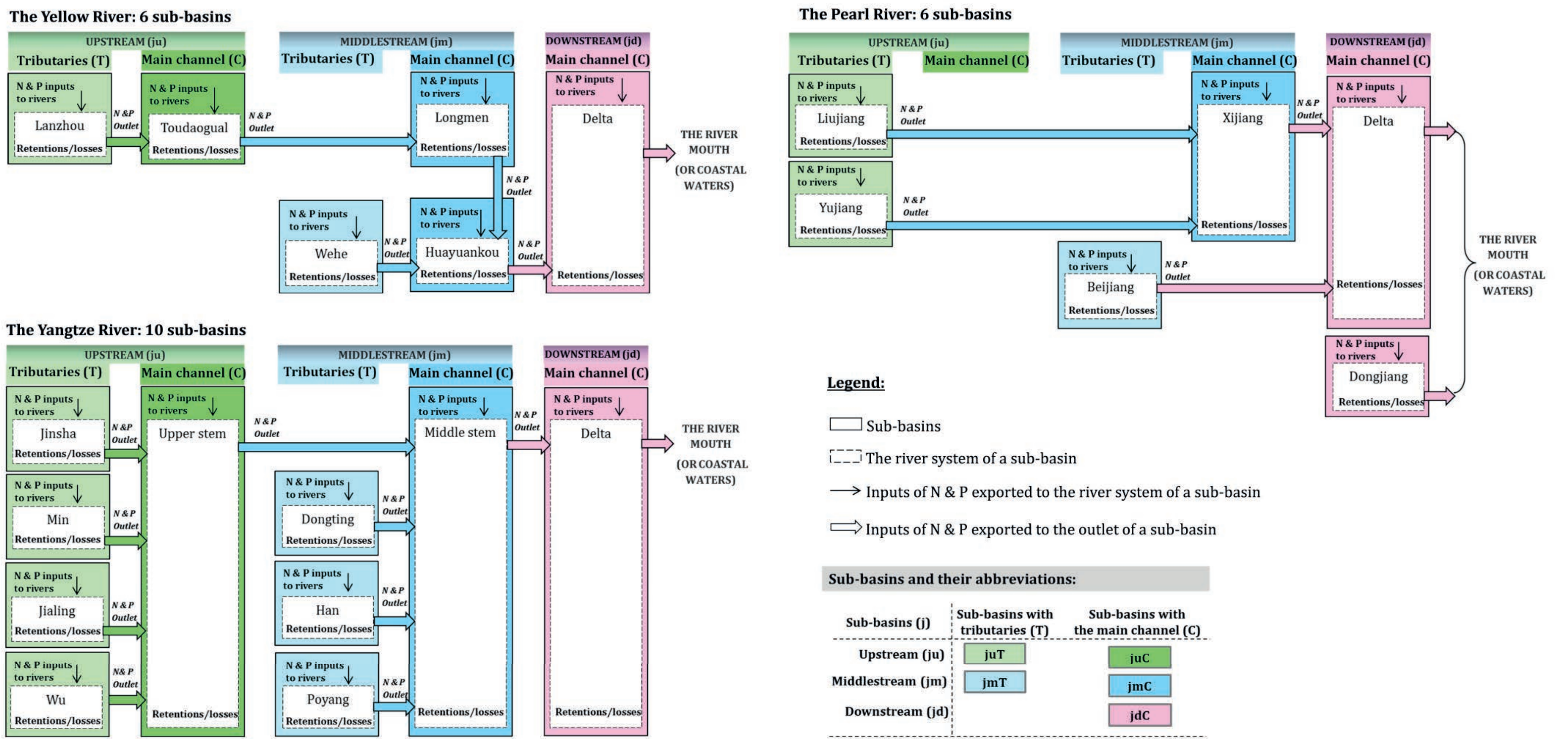


Figure 5.3. *Schematic overview of the sub-basin scale modeling framework for the Yellow, Yangtze and Pearl rivers in the MARINA Nutrient Model (Model to Assess River Inputs of Nutrients to seAs). See Figures 5.1 and 5.2 for locations of the rivers in China and their sub-basins (abbreviations of the sub-basins are also given in Figures S5.1-S5.3).*

5.2.3 Indicator for Coastal Eutrophication Potential

We use the Indicator for Coastal Eutrophication Potential (ICEP) to assess effects of nutrient export by rivers on coastal eutrophication in China. Billen and Garnier (2007) developed this indicator. ICEP shows the potential of producing non-siliceous harmful algae in coastal waters under excess of riverine N and P fluxes taking into account the requirements for growth (the Redfield ratio of N:P:Si:C = 16:1:20:106). Either N-ICEP or P-ICEP (kg C-eq. km^{-2} day^{-1}) is calculated depending on limiting nutrient (Garnier et al. 2010; Strokal et al. 2014b):

$$\text{N-ICEP} = [TN_{flx}/(14 \cdot 16) - DSi_{flx}/(28 \cdot 20)] \cdot 106 \cdot 12, \text{ if N is limiting (N:P < 16)} \quad (5.11)$$

$$\text{P-ICEP} = [TP_{flx}/31 - DSi_{flx}/(28 \cdot 20)] \cdot 106 \cdot 12, \text{ if P is limiting (N:P > 16)} \quad (5.12)$$

TN_{flx}, TP_{flx} and DSi_{flx} are the fluxes of total N, P and dissolved silica (Si) exported by rivers to coastal waters, respectively (kg km^{-2} $year^{-1}$). TN is the sum of dissolved inorganic, dissolved organic and particulate N. TP is the sum of dissolved inorganic, dissolved organic and particulate P. Dissolved forms of N and P are from the *MARINA* Nutrient Model (see Section 5.2.2). Particulate N and P and dissolved Si are from Global *NEWS*-2 (Beusen et al. 2009; Mayorga et al. 2010; Strokal et al. 2014b). Particulate N and P are calculated using an empirical relationship with total suspended solids (TSS) through a regression analysis (details are in Beusen et al. 2005 and Mayorga et al. 2010). A similar approach was implemented in the Global *NEWS*-2 model to quantify river export of dissolved Si (details are in Beusen et al. (2009)). We calculated daily fluxes as annual fluxes of TN, TP and DSi divided by 365. Positive ICEP values indicate a high risk for harmful algal blooms because rivers discharge N and P to coastal waters in excess over DSi. Negative ICEP values indicate low risks.

5.2.4 Model evaluation and uncertainties

We validated the *MARINA* Nutrient Model by comparing modeled values with measurements. Measurements were available for DIN and DIP export by the Yellow, Yangtze and Pearl (Table S5.10). Measured DIN and DIP yields (kg km^{-2} $year^{-1}$) were calculated from concentrations and water discharges normalized by basin areas; DIN is the sum of nitrite (NO_2^-), nitrate (NO_3^-), ammonium (NH_4^+), and DIP reflects phosphate (PO_4^{3-}) (Table S5.10). Tong et al. (2015) provide measured total N (TN) and total P (TP) in river mouths of the Yellow, Yangtze, Pearl, Huai, Hai and Liao (only TP) for the period of 2006-2012. However, Tong et al. (2015) do not provide measured DIN, DIP, DON and DOP for 2000. Nevertheless, we compared modeled river exports of TN (the sum of DIN, DON and particulate N) and TP (the sum of DIP, DOP and particulate P) with measured

TN and TP from Tong et al. (2016). We used particulate N and P from Global *NEWS*-2 (Mayorga et al. 2010; Strokal et al. 2014b).

Figure 5.4a shows ranges in the measured yields of DIN, DIP, TN and TP compared to modeled values for individual rivers. We assessed the model performance using R_P^2 (the Pearson's coefficient of determination, 0-1), R_{NSE}^2 (the Nash-Sutcliffe efficiency, 0-1) and ME (Model Error, %) according to Moriasi et al. (2007). R_P^2 shows the proportion of the variance in measured values that can be explained by the model. R_{NSE}^2 indicates how well the points of the modeled and measured values fit the 1:1 line, and ME indicates the difference between modeled and measured values. These indicators show a reasonable performance of our model: R_P^2 is 0.84, R_{NSE}^2 is 0.78 and ME is 8%. DIN export by the Yellow and Yangtze is within the range of measurements. The model results for DIN export by the Yellow River are slightly lower than measured values. For DIP export by the Yangtze, Pearl and Yellow rivers they are slightly higher. Modeled TN and TP export by the Yangtze and Pearl is in agreement with measured values. For northern rivers (the Yellow, Huai, Liao and Hai) measured TN and TP are lower than modeled values (Figure 5.4a, Tong et al. 2015). The difference between modeled and measured DIN, DIP, TN and TP yields can be associated with uncertainties in model inputs and parameters as well as with uncertainties in the measured values. Another reason can be the ratios of dissolved to particulate nutrients in TN and TP as these ratios may differ between modeled and measured nutrients. We compared nutrients modeled for 2000 with nutrients measured for different years (e.g., TN and TP for 2006-2012 from Tong et al. 2015). This may also explain the differences between modeled and measured values. However, the number of available measurements is still limited in literature, making it difficult to validate the model results in particular for dissolved organic N and P.

We compared total exports of TN and TP by six rivers with measured TN and TP from Tong et al. (2016) (Table S5.11). Our estimates show that six rivers exported 2143 kton of TN and 319 kton of TP to the Chinese seas in 2000 (see Section 5.3.2). These estimates are close to the range of minimum and maximum values from Tong et al. (2016) (2136-3096 kton of TN and 159-275 kton of TP). Furthermore, our study indicates the large contribution of the Yangtze and Pearl rivers to nutrient pollution of the Chinese seas (see Section 5.3.2), which is also in line with Tong et al. (2015).

We also compared results of the *MARINA* Nutrient Model with reported trends in nutrient fluxes and their spatial variability. The model quantifies increasing trends in nutrient export by Chinese rivers since 1970 (see Section 5.3.2). These model results are generally in line with existing literature indicating increases in nutrient concentrations in Chinese rivers. For example, Ti et al. (2012) quantified increasing trends for DIN export by the Yangtze, Yellow and Pearl rivers. Tao et al. (2010) indicate increases in N inputs to downstream stations of the Yellow River since the 1970s. Wang (2006) and Li et al. (2007) report on increases in N and P inputs to the Yangtze since the 1960s. Increasing trends in nutrients are also reported by Sun et al. (2013a) for upstream stations (before the Three Gorges Dam: TGD) and by Sun et al. (2013b) for middlestream stations (after the TGD) of the Yangtze. Xu et al. (2014) indicate higher concentrations of nutrients in the northern rivers (e.g., Yellow, Huai) than in the southern rivers (e.g., Pearl) of China. This is because the northern rivers discharge less water. These results are all in line with our model results. Tong et al. (2016) indicate higher nutrient inputs in the river system of downstream areas than in upstream areas of the Yellow River. This is, generally, in agreement with our findings (see Section 5.3.2). Section 5.3.4 provides discussion on comparing our model results with other modeling studies.

The *MARINA* Nutrient Model was developed by implementing Global *NEWS*-2 approaches to the sub-basin scale, and by improving the modeling of animal manure and human waste, and reservoirs. On one hand, this improves the modeling of nutrient export by large Chinese rivers. On the other hand, applying Global *NEWS*-2 approaches to sub-basins, while using model parameters and inputs from various sources may increase uncertainties. For example, the approaches of Global *NEWS*-2 do not account for DON and DOP retentions in dammed reservoirs (only losses from rivers via water withdrawal) (Mayorga et al. 2010). This may lead to an overestimation of river export of these nutrients. Most model inputs for sub-basins are from gridded databases of Global *NEWS*-2 (0.5 latitude by 0.5 longitude), which were prepared from national assessments such as FAO (Bouwman et al. 2009). For some parameters we used sub-basin specific values, for instance fractions of manure losses to rivers were calculated from provincial data. For others we used basin- (e.g., nutrient removal during sewage treatment) or country-specific (e.g., fractions of human excretion discharges) values (Figure S5.4).

We tested the sensitivity of model outputs to variations in model inputs and parameters. We found that model results for 2000 are sensitive to changes in the following model inputs and parameters: animal manure production, direct losses of this manure to surface waters of sub-basins (fractions), use of synthetic fertilizers and water discharges (Figures S5.5 and S5.6). For example, increasing direct losses (fractions) of animal manure to aquatic systems by 50% for all sub-basins resulted in a 25-40% increase in river export of DIP, DON and DOP. The DIN export by rivers of the Bohai Gulf is more sensitive to changes in animal manure and less sensitive to changes in synthetic fertilizers compared to the DIN export by rivers of the Yellow Sea and South China Sea (Figure S5.5). This is because of differences in the relative contribution of sources to DIN export and in hydrology (see Section 5.3.1). Human waste is an important factor of P river export (Figures S5.5 and S5.6). The results of this sensitivity analysis give us a better understanding of the relevant model inputs and parameters for further check.

We also compared selected model inputs to an independent Chinese county dataset (RESDC 2014). This gives an indication of the quality of our sub-basin model inputs for animal manure, synthetic fertilizers, population and sub-basin areas (Figure 5.4b). For these inputs, we aggregated information from counties to sub-basins in ArcGIS, and then compared with our sub-basin inputs. The results of this comparison increase our confidence in using sub-basin model inputs derived from these gridded databases: Pearson's coefficients of determination (R_P^2) range between 0.87 and 0.99 (see Figure 5.4b).

Fractions of manure losses to rivers are important parameters in our model that determine river pollution by animal manure (Section 5.3). We calculated these fractions from provincial data derived from the NUFER model (Ma et al. 2012). NUFER was developed for China based on statistical data (ECCAP 2006; MOA 2006), field surveys of about 50 thousand farms between 1999-2008 (NATESC 1999) and literature (Ma et al. 2010; Ma et al. 2012). This builds our trust in using these fractions in our model to quantify manure losses to Chinese rivers. Furthermore, some model parameters that were derived on the basis of limited literature were checked with local experts in China (e.g., Chinese Academy of Sciences, Peking University). These are, for example, the fractions of nutrient removal during treatment in the Dongjiang sub-basin and the fraction of N or P in animal manure and human waste that is discharged to rivers in a form (e.g., DIN or DON).

Uncertainties in Global *NEWS*-2 approaches were addressed in earlier studies (Bouwman et al. 2009; Fekete et al. 2010; Mayorga et al. 2010; Van Drecht et al. 2009). Global *NEWS*-2 has been widely used for the regional analyses of nutrient export by rivers draining into the coastal waters of China (Li et al. 2011b; Qu & Kroeze 2010; Qu & Kroeze 2012; Strokal et al. 2014b; Yan et al. 2010), Indonesia (Suwarno et al. 2014a; Suwarno et al. 2014b), the Bay of Bengal (Sattar et al. 2014; Zinia & Kroeze 2015), Europe (Thieu et al. 2010), Africa (Yasin et al. 2010) and North America (McCrackin et al. 2013; McCrackin et al. 2014). Global *NEWS*-2 was calibrated and validated for large rivers worldwide, including large Chinese rivers (the Yellow, Yangtze and Pearl) (Mayorga et al. 2010). The validation results indicate an acceptable performance of Global *NEWS*-2: R_{NSE}^2 is 0.51-0.90 depending on nutrient form (Mayorga et al. 2010). Furthermore, validation results of Strokal et al. (2014b) indicate an acceptable performance of Global *NEWS*-2 for DIN and DIP export by the Yangtze, Pearl, Yellow and Liao rivers. For example, according to Strokal et al. (2014b) R_P^2 is 0.96, R_{NSE}^2 is 0.42 and

ME is 18%. For the *MARINA* Nutrient Model R_{NSE}^2 is much higher and ME is lower (Figure 5.4a), indicating better performance.

Based on the above, we consider the *MARINA* Nutrient Model appropriate for its purpose: to quantify river inputs of N and P to Chinese seas by source at the sub-basin scale.

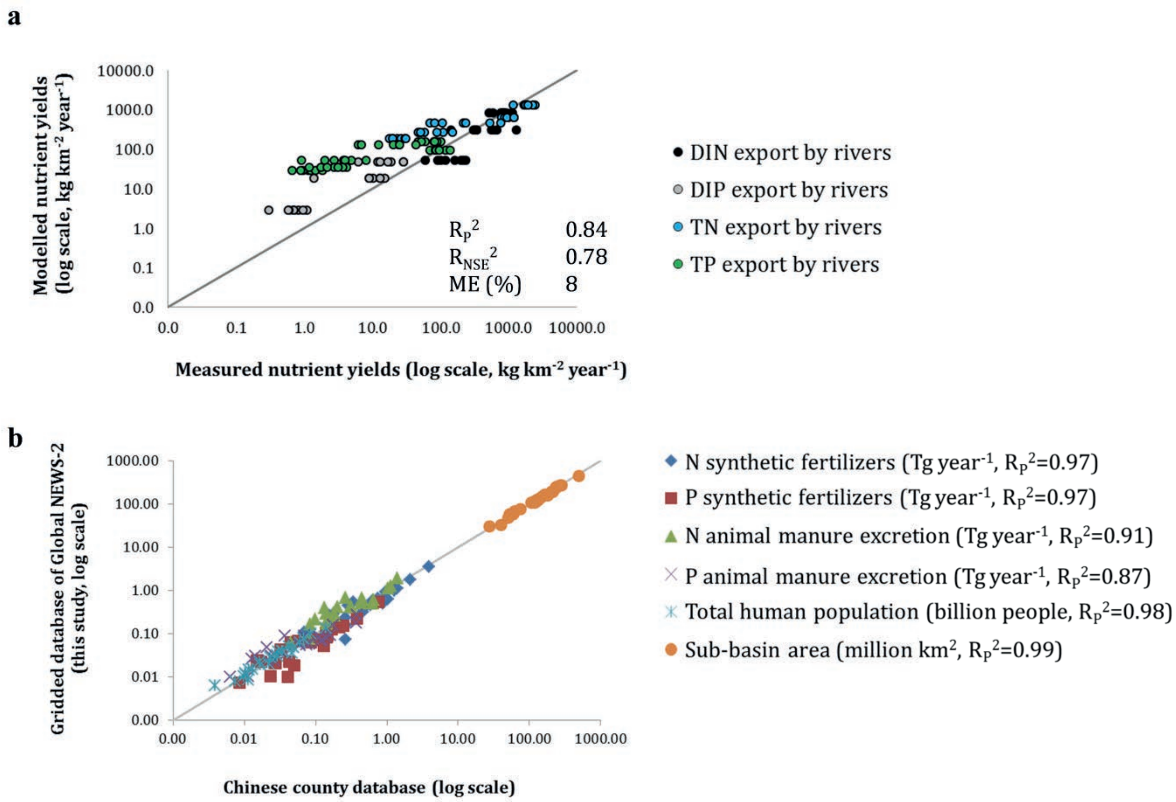


Figure 5.4. *(a) Measured versus modeled dissolved inorganic nitrogen (DIN), dissolved inorganic phosphorus (DIP), total nitrogen (TN) and total phosphorus (TP) export by the Yangtze, Yellow and Pearl, Hai, Huai and Liao rivers to the coastal waters in China (yields, kg km^{-2} year^{-1}), and (b) sub-basin model inputs from gridded database (0.5 latitude by 0.5 longitude) of Global NEWS-2 (Mayorga et al. 2010) versus model inputs from the Chinese county database (RESDC 2014) for 2000. R_P^2, R_{NSE}^2 and ME are the Pearson's coefficient of determination (0-1), the Nash-Sutcliffe efficiency (0-1) and the Model Error (%), respectively. Modeled yields of DIN and DIP were derived from the MARINA Nutrient Model for 2000. Measured yields of DIN and DIP for the Pearl, Yangtze and Yellow rivers were calculated from concentrations of DIN and DIP using annual water discharges and drainage areas of the basins (see Table S5.10 for values and their sources). Modeled yields of TN were calculated from the sum of DIN (from the MARINA Nutrient Model), dissolved organic N (from the MARINA Nutrient Model) and particulate N (from Mayorga et al. 2010) for 2000. The same holds for modeled TP export by rivers. Tong et al. (2015) provided measured TN and TP in river mouths in ton year^{-1} for the period of 2006-2012. We converted tons of TN and TP to kg km^{-2} year^{-1} using drainage areas of the basins from Tong et al. (2015).*

5.3 An analysis of nutrient export by rivers to Chinese seas from 1970 to 2050

5.3.1 Characteristics of human activities at the sub-basin scale

Our analyses show that urbanization and agriculture are strongly interconnected and influence N and P export by Chinese rivers at the sub-basin scale (Figures 5.5 and 5.6, and Figures S5.7-S5.11). The total population in the studied sub-basins increased from 0.6 billion in 1970 to 0.9 billion in 2000 (Figure S5.8) in line with rapid economic development (Strokal et al. 2014b). As a result, N and P human excretion doubled in this period (Figure 5.5). In 1970 most of the human excretion was from the rural population (Figure 5.5), of which more than half was applied on land to grow crops, fruits or vegetables (Figure S5.11) as also indicated in national studies (Ju et al. 2005; Ma et al. 2012; Miao et al. 2010). By 2000 the urban population accounted for 40% of the total population (the range for sub-basins is 0-68%), where half of the urban population was connected to sewage systems (the range for sub-basins is 0-100%; Figure S5.11), in line with national estimates of WHO/UNICEF (2014). Sewage systems were not effective in removing N and P from human excretion (Figure S5.9). Uncollected urban excretion was mainly discharged to surface waters in 2000 (Figure S5.10). In general, urbanization was taking place mostly in downstream areas (Figure 5.5, and Figures S5.8-S5.11).

Chinese agriculture has been industrializing because of food security reasons and changes in diets (Bai et al. 2014; Ju et al. 2005; Ma et al. 2013b; Schneider 2011). In 1970 approximately 5.8 Tg of N and 0.8 Tg of P in animal excretion were produced in the study area (Figure 5.6). Most animal manure was used to grow crops (Figure 5.6) (Bai et al. 2014; Ju et al. 2005). Synthetic fertilizer was hardly used (Figure S5.7). By 2000 N and P in animal excretion had doubled (a factor of 1-5 increase among sub-basins). Over one-third of N and half of P are calculated to have been discharged directly to surface waters from animal manure, and the remainder was used to fertilize soils in agriculture (after ammonia losses; Figure 5.6). Industrial animal farms often did not recycle manure on land and had poor treatment facilities (see also Section 5.1). This explains the large manure losses to surface waters in 2000. Arable farmers increasingly used synthetic fertilizers because of the low prices and low labor demand (Bai et al. 2013; Bai et al. 2014; Ju et al. 2005; Li et al. 2013; MOA 2011b). Thus, the total use of N synthetic fertilizers in the sub-basins increased from 2.6 Tg in 1970 to 16.3 Tg in 2000. For P synthetic fertilizers this increase was from 0.3 Tg to 2.4 Tg during this period (Figure S5.7). In general, middle- and downstream sub-basins have intensive agriculture (see Figure 5.2 for locations of the sub-basins, Figure 5.6 and Figure S5.7 for agricultural activities). For example, Huai, Hai and the delta of the Yellow River are part of the North

China Plain where over half of national wheat is produced (Kendy et al. 2003). Our estimates for the total manure and fertilizer use are generally lower than national totals (e.g., Ma et al. 2010) because our study area covers only part of China.

In the future there may be ongoing urbanization and industrialization of agriculture in the study area. The GO scenario assumes a rapid economic development for the study area (e.g., at least a 10-fold increase in GDP at purchasing power parity between 2000 and 2050). The total population in the sub-basins is projected to increase slightly between 2000 and 2050 (Figure S5.8). However, a fast urbanization is assumed because of migration from rural to urban areas. As a result, the urban population with a sewage connection is projected to increase by a factor of 1-7 in the sub-basins (Figure S5.8). As a result, nutrient inputs to Chinese rivers from the urban population are calculated to increase (Figure 5.5, Figure S5.9) unless efficient sewage treatment is implemented (Alcamo et al. 2005; Van Drecht et al. 2009). Agricultural activities in the study area are projected to intensify in GO (Alcamo et al. 2005; Bouwman et al. 2009). Reasons for this are a slight decrease in agricultural areas (due to, for instance, urbanization; Strokal et al. 2015), and a large increase in animal manure production (associated with increasing demand for meat) and in synthetic fertilizer use (Figure 5.6, Figure S5.7). More industrial farms may emerge, preferably around cities with a high demand for meat products. Since in our GO scenario the fraction of manure that is directly discharged to rivers will not change largely in the coming years, large amounts of manure are projected to discharge to surface waters in 2050 (Figure 5.6).

Hydrology, in-river processes, water consumption and dams influence N and P export by Chinese rivers (Figures S5.12-S5.17). Rivers draining into the Bohai Gulf (the Yellow, Hai and Liao) are drier than rivers draining into the Yellow Sea (Yangtze, Huai) or the South China Sea (Pearl). For example, water discharges of the Bohai Gulf rivers are relatively low (<15 km^3 $year^{-1}$) (Figure S5.14) because of low runoff and precipitation (Tang et al. 2013; Zhai et al. 2005). This results in lower nutrient export from land to the rivers. Part of the nutrients are retained in or lost from rivers before reaching coastal waters. Water consumption removes nutrients from rivers, and it is generally higher in areas with intensive agriculture (e.g., Hai, Huai, delta sub-basins of the Yellow and Yangtze rivers; Figures 5.6, S5.7 and S5.15). Considerable N can be lost via in-river denitrification (Figure S5.16), while P may be accumulating. Nutrient retentions in dammed reservoirs increased between 1970 and 2000 because many more dams were constructed (Figures S5.12 and S513 and Table S5.12). An example is the world's largest Three Gorges Dam (TGD), built in the middle of the Yangtze (the Upper stem sub-basin, Figures 5.1 and 5.2), which has an impact on downstream areas (Hu et al. 2011; Yang et al. 2011; Yang & Lu 2014a). The traveling distance of nutrients to the river mouth is another factor (Figure

S5.17). In general, not all nutrients from upstream activities reach the river mouth because of losses and retentions during traveling. This traveling is shorter for nutrients from downstream activities, thus more nutrients reach the river mouth (Figure S5.17). GO projects that dry basins (Yellow, Hai and Liao in our study) may become drier between 2000 and 2050, while wet basins (Yangtze, Pearl, Huai) may become wetter (Figure S5.14). Nutrient retentions in and losses from rivers may increase because of more dams and higher water consumption (Figures S5.13-S5.15).

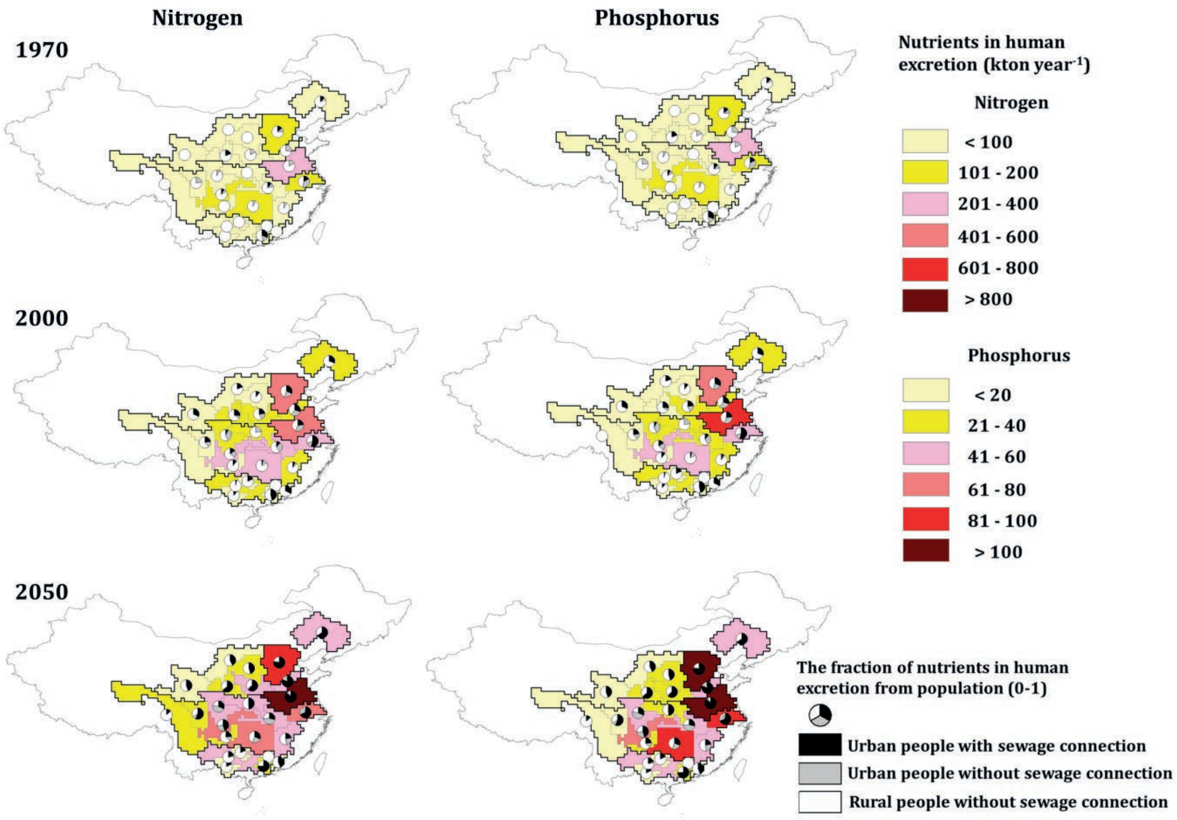


Figure 5.5. *Nitrogen and phosphorus in human excretion (kton year^{-1}) by sub-basin in 1970, 2000 and 2050. Pie charts show fractions of human excretion resulting from the urban population with and without sewage connections, and from the rural population (all without sewage connection). 2050 is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Seitzinger et al. 2010). See Section 5.2.2 for model inputs and parameters.*

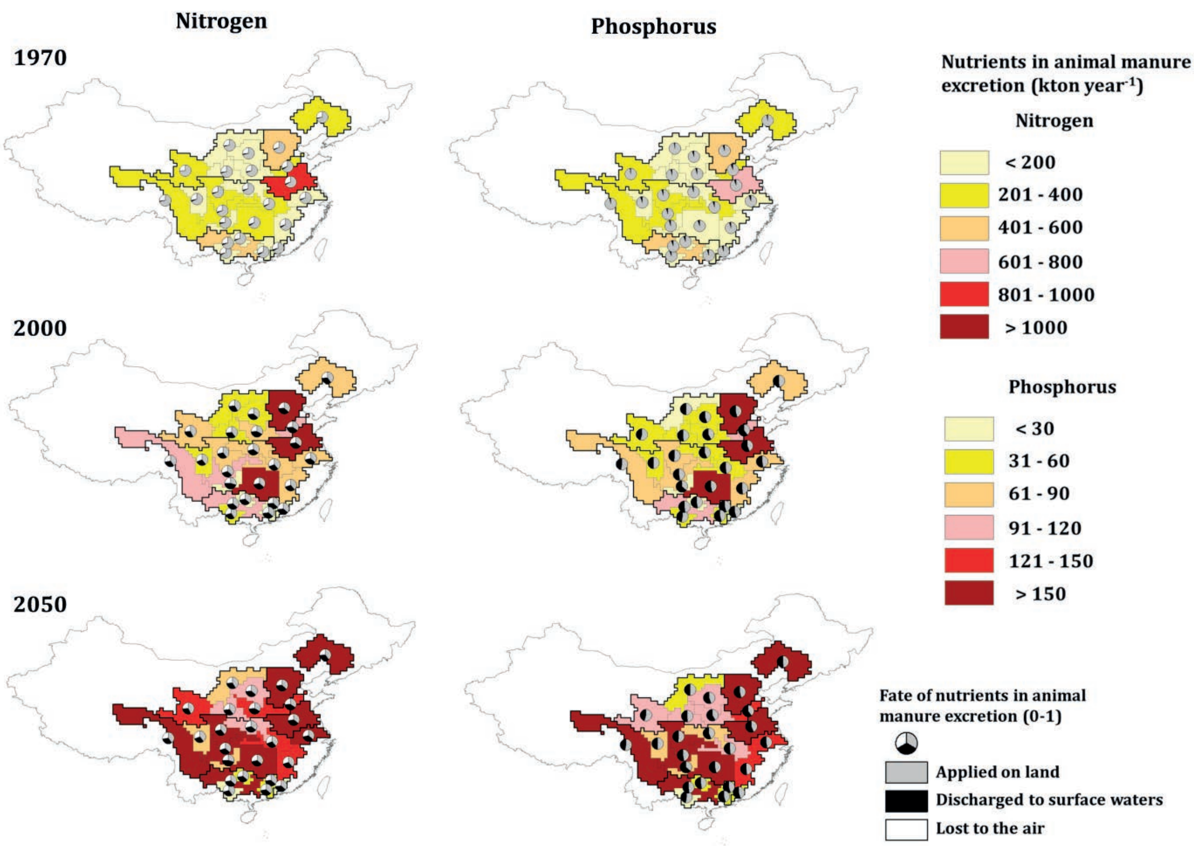


Figure 5.6. *Nitrogen and phosphorus in animal manure excretion (kton year^{-1}) by sub-basin in 1970, 2000 and 2050. Pie charts show fractions of nutrients in animal manure that are applied to land, discharged to surface waters and lost to the air as ammonia. 2050 is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Seitzinger et al. 2010). See Section 5.2.2 for model inputs and parameters.*

5.3.2 River export of nutrients by source at the sub-basin scale

We calculate that nutrient export by the six rivers increases rapidly from 1970 to 2050 (Figures 5.7, 5.9 and 5.10). In 1970 the six rivers exported 520 kton of DIN, 262 kton of DON, 26 kton of DIP and 37 kton of DOP to Chinese seas (Figure 5.7). Between 1970 and 2000 the total river export of DIN and DIP doubled, and DON and DOP increased by a factor of 4-5. By 2050 the river export of nutrients is projected to further increase (Figure 5.7). The three large rivers (Yangtze, Yellow and Pearl) are dominant exporters of nutrients to the Yellow Sea, Bohai Gulf and South China Sea, respectively (Figure 5.8, Figure S5.18). Below we describe nutrient export by rivers to each of these three Chinese seas. We finish this section by comparing our results with other modeling studies.

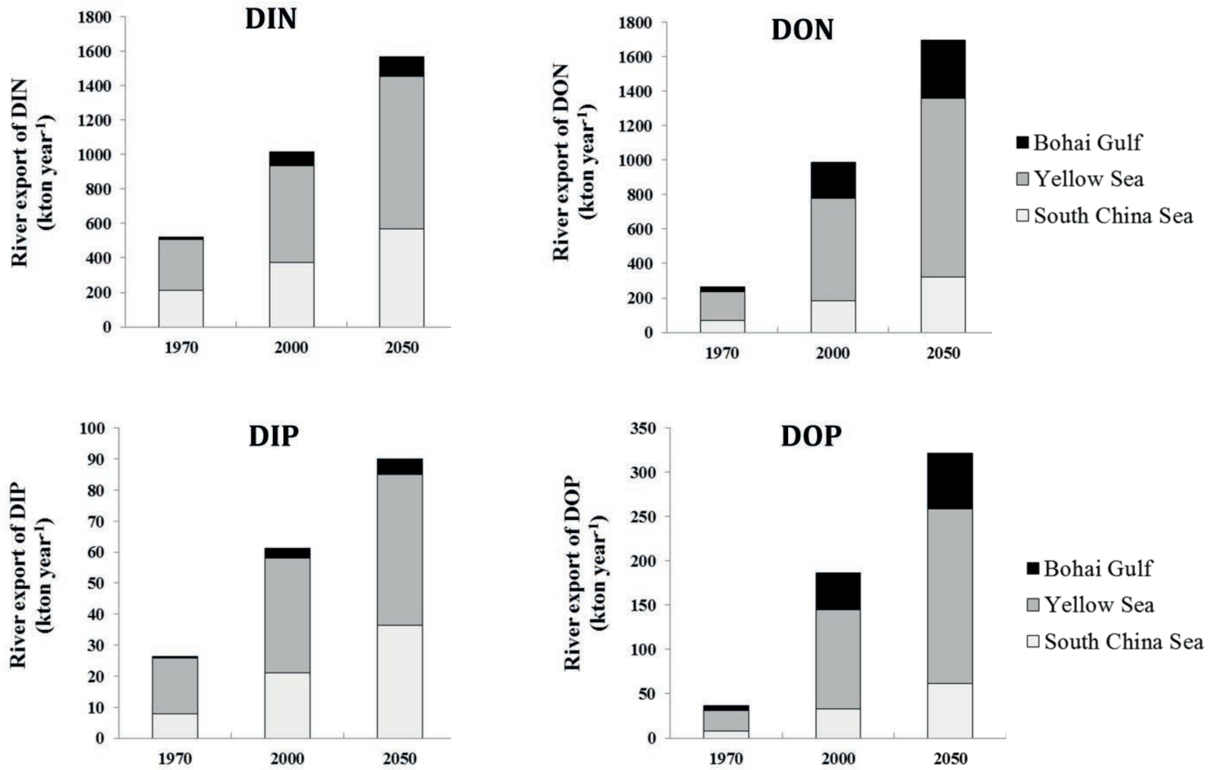


Figure 5.7. *Modeled dissolved inorganic (DIN, DIP) and organic (DON, DOP) nitrogen (N) and phosphorus (P) export by rivers to the Bohai Gulf, Yellow Sea and South China Sea in 1970, 2000 and 2050 (kton year^{-1}). These nutrients are exported by the Yellow, Hai and Liao rivers to the Bohai Gulf, by the Yangtze and Huai rivers to the Yellow Sea, and by the Pearl River to the South China Sea (see Figure 5.1 for river locations and seas). The share of these rivers in the total nutrient loads is shown in Figure 5.8. 2050 is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Seitzinger et al. 2010). See Section 5.2 for model description and inputs.*

The Bohai Gulf receives increasing amounts of nutrients from the Yellow, Hai and Liao rivers. The Yellow River, which is the second largest rivers in China, transported 6 kton of DIN, 0.4 kton of DIP, 14 kton of DON and 3.3 kton of DOP in 1970 (Figure 5.9). By 2000 the export of these nutrients increased by a factor of 7-8. Around 70% of DIN and DIP and one-third of DON and DOP fluxes originated from human activities in the downstream sub-basin of the Yellow River. The remainder amount of the nutrients is mainly from middlestream sub-basins (Figure 5.9). The Hai and Liao transported 6 kton of DIN, 0.2 kton of DIP, 12 kton of DON and 2.4 kton of DOP to the Bohai Gulf in 1970 (Figure 5.10). Nutrient export by Hai and Liao increased by 4-8 times between 1970 and 2000 as a result of human activities (see Section 5.3.1). In 1970 direct discharges of human waste to surface waters accounted for over half of the nutrients entering the Bohai Gulf, except for DIN export by the Liao, where other sources also contributed (Figures 5.11 and 5.12). However, in 2000 60-78% of the nutrient inputs to the Bohai

Gulf were from direct discharges of animal manure to surface waters. These manure discharges occurred because of poor manure management (see Section 5.3.1). Sewage was an important source of DIP in the Bohai Gulf rivers. By 2050 nutrient export by the Yellow, Hai and Liao rivers is projected to be 30-70% higher than in 2000 depending on river and nutrient form, except for DIN export by Hai (Figures 5.9 and 5.10). The Hai River is projected to export less DIN because of increased DIN retentions in the river system as a result of damming (see Section 5.3.1). In our scenario, direct discharges of manure remains a dominant source of nutrients in the Bohai Gulf. And sewage contributes more to dissolved inorganic N and P in rivers than in the past, in particular in downstream areas (Figures 5.11 and 5.12).

The Yellow Sea receives nutrients from the Yangtze and Huai rivers in this study. DIN export by the Yangtze (the largest Chinese river) increased from 295 kton to 556 kton between 1970 and 2000 (Figure 5.9). Between these years the Yangtze export of DIP increased from 18 kton to 33 kton, of DON from 155 kton to 500 kton, and of DOP from 20 kton to 93 kton. In 2000 activities in the downstream sub-basin contributed to 12 - 27% of DIN and DIP, and to 6-7% of DON and DOP export by the Yangtze to the coastal waters. The share of middlestream activities was 45-70% and 18-45% of upstream activities depending on the nutrient form (Figure 5.9). The Huai River transported 0.7 kton of DIN, 0.5 kton of DIP, 12.6 kton of DON and 3 kton of DOP to the Yellow Sea in 1970 (Figure 5.10). These exports increased by a factor of 6-8 between 1970 and 2000 because of increasing human activities (Section 5.3.1). In 1970 atmospheric N deposition and biological N fixation were dominant sources of DIN, and organic matter leaching of DON in the rivers draining into the Yellow Sea, except for the Huai River (Figure 5.11). Direct discharges of human waste to surface waters were important sources of DON and DOP in that time (Figure 5.12). This was different for 2000: direct discharges of manure were responsible for 62-69% of DON, DOP and DIP, and for 22% of DIN inputs to the Yellow Sea (Figures 5.11 and 5.12). Other sources such as the use of synthetic fertilizers were important sources of DIN in rivers in 2000 (Figure 5.11). Nutrient export by the Yangtze and Huai rivers are projected to further increase (Figures 5.9 and 5.10) as a result of agriculture (synthetic fertilizer use for river export of DIN, and manure discharges for river export of the other nutrient forms) and urbanization (for river export of DIP) (Figures 5.11 and 5.12). Sewage in the downstream sub-basin is in particular an important source of DIP exported by the Yangtze.

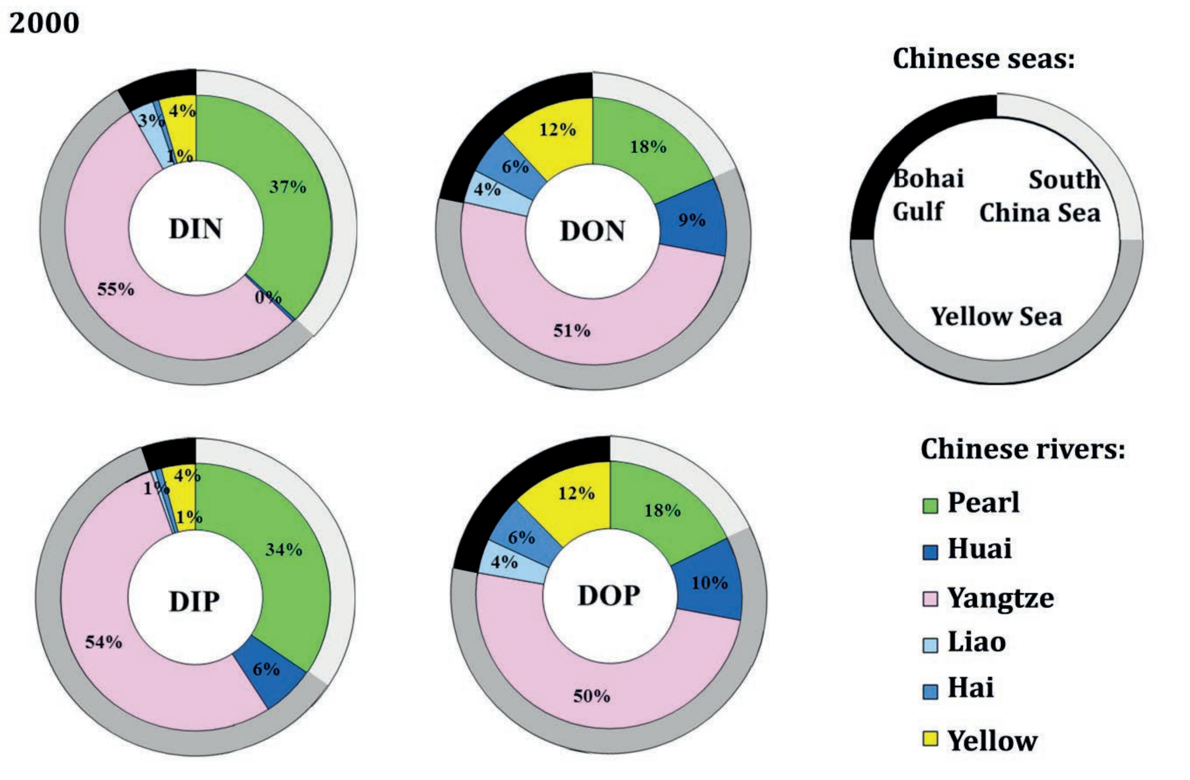
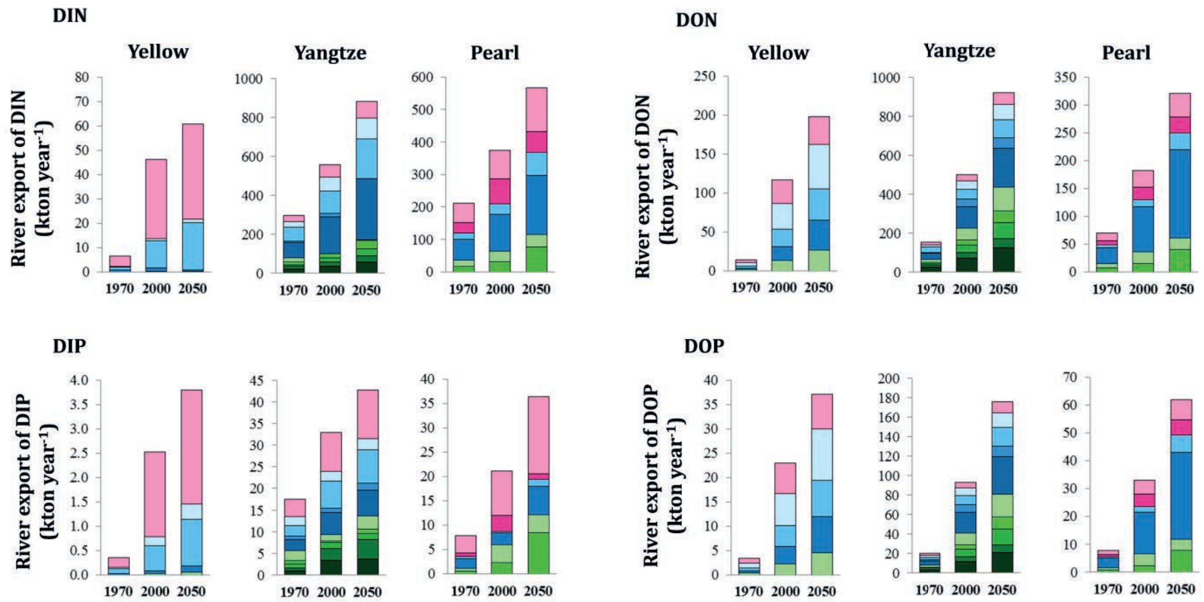


Figure 5.8. *The share of rivers in the modeled dissolved inorganic (DIN, DIP) and organic (DON, DOP) nitrogen (N) and phosphorus (P) export to the Bohai Gulf, Yellow Sea and South China Sea in 2000 (%). For 1970 and 2050 this share is shown in Figure S5.18.*

We analyzed nutrient inputs to the South China Sea by the Pearl River (third largest river in China). Similar to the Bohai Gulf and Yellow Sea, the coastal waters of the South China Sea are polluted by increasing amounts of nutrients (Figure 5.9). River export of DIN almost doubled between 1970 (212 kton) and 2000 (375 kton) and DIP export by the Pearl River almost tripled between 1970 (8 kton) and 2000 (21 kton). For DON this increase was from 69 kton in 1970 to 182 kton in 2000, and for DOP from 8 kton in 1970 to 33 kton in 2000 (Figure 5.9). In 2000 over half of DON and DOP in the coastal waters of the South China Sea were from human activities in the middlestream sub-basins of the Pearl River, 20% from activities in upstream sub-basins and around 30% from activities in downstream sub-basins (Figure 5.9). For DIN and DIP export the share of middlestream activities was 13-39%, 17-28% for upstream activities and 44-59% for downstream activities in 2000. Atmospheric N deposition and biological N fixation were important sources of DIN, while organic leaching was for DON in the Pearl River in 1970 (Figure 5.11). Less than half of DIP and DOP inputs to the sea were from direct losses of human waste to surface waters of the Pearl basin. Only in the Delta sub-basin (see Figure 5.2 for location) did sewage contribute considerably to DIP export (Figure 5.12). By 2000, the share of sources had changed: synthetic fertilizers with direct manure discharges were large contributors of DIN fluxes to the South China Sea. Manure discharges were responsible for 61-74% of DON, DOP, and DIP, and for 20% of DIN in the South China Sea. Sewage was an important source of DIP in the water systems of the Delta sub-basin where urbanization is profound (Figures 5.11 and 5.12). In our scenario for 2050 the Pearl River transports more nutrients to the South China Sea than in 2000 (Figure 5.9) because of manure discharges resulting from livestock production (Figures 5.11 and 5.12). Fertilizer use may remain an important source of DIN export by the river, and sewage of DIP (see Section 5.3.1 for the characteristics of human activities at the sub-basin scale).



Sub-basins of the large rivers draining into the Bohai Gulf, Yellow Sea and South China Sea:

	Yellow River (the Bohai Gulf)	Yangtze River (the Yellow Sea)	Pearl River (the South China Sea)
Downstream:	Delta	Delta	Delta Dongjiang
Middlestream:	Huayuankou Wehe Longmen	Middle stem Poyang Han Dongting	Beijiang Xijiang
Upstream:	Toudaogual Lanzhou	Upper stem Wu Jialing Min Jinsha	Yujiang Liujiang

Figure 5.9. *Modeled dissolved inorganic (DIN, DIP) and organic (DON, DOP) nitrogen (N) and phosphorus (P) export by the Yellow, Yangtze and Pearl rivers from sub-basins in 1970, 2000 and 2050 (kton year^{-1}). 2050 is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Seitzinger et al. 2010). Figure 5.2 provides locations of the sub-basins. See Section 5.2 for model description and inputs.*

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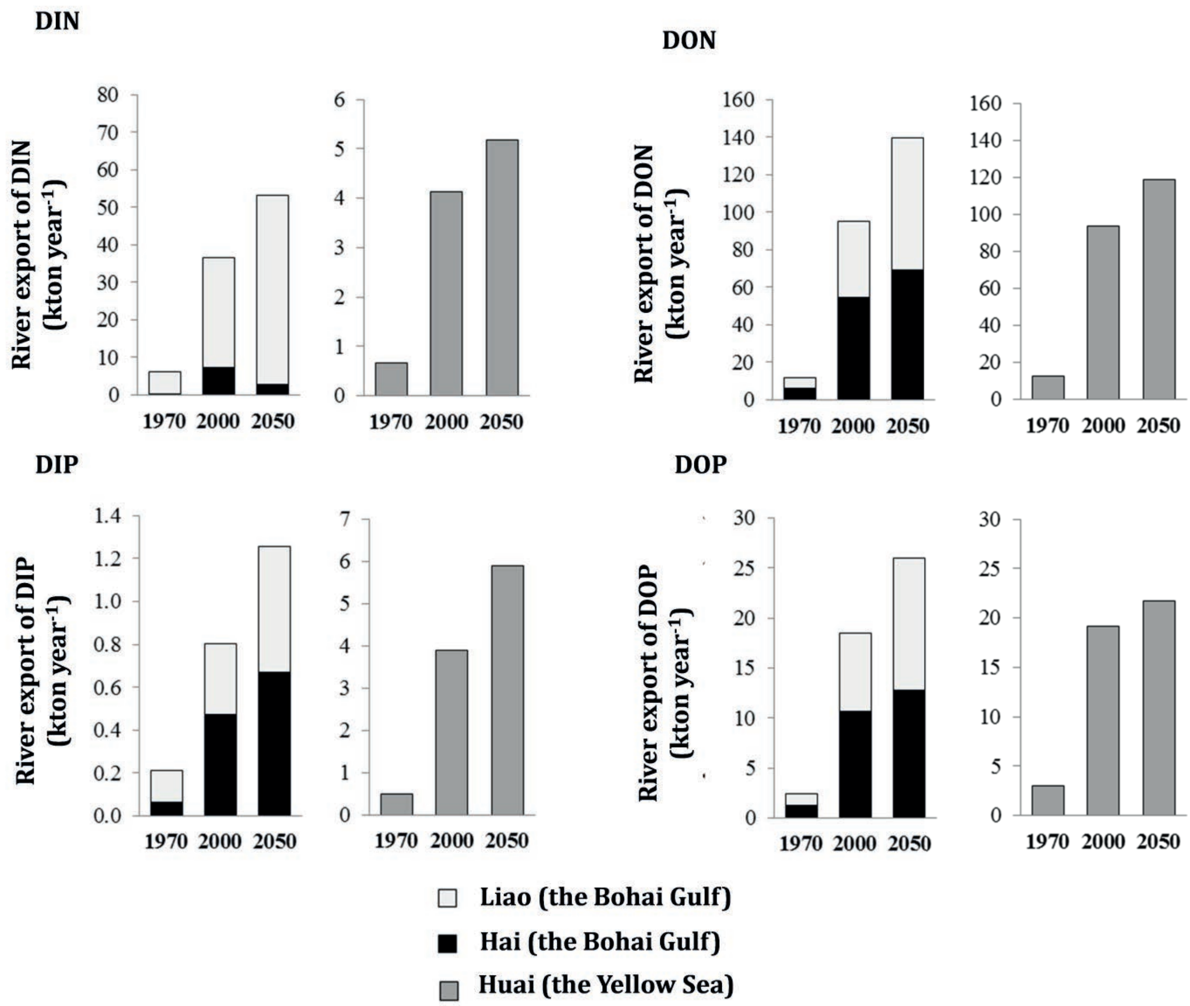

Figure 5.10. *Modeled dissolved inorganic (DIN, DIP) and organic (DON, DOP) nitrogen (N) and phosphorus (P) export by the Liao, Hai and Huai rivers in 1970, 2000 and 2050 (kton year^{-1}). 2050 is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Seitzinger et al. 2010). Figure 5.1 provides locations of the river basins. See Section 5.2 for model description and inputs.*

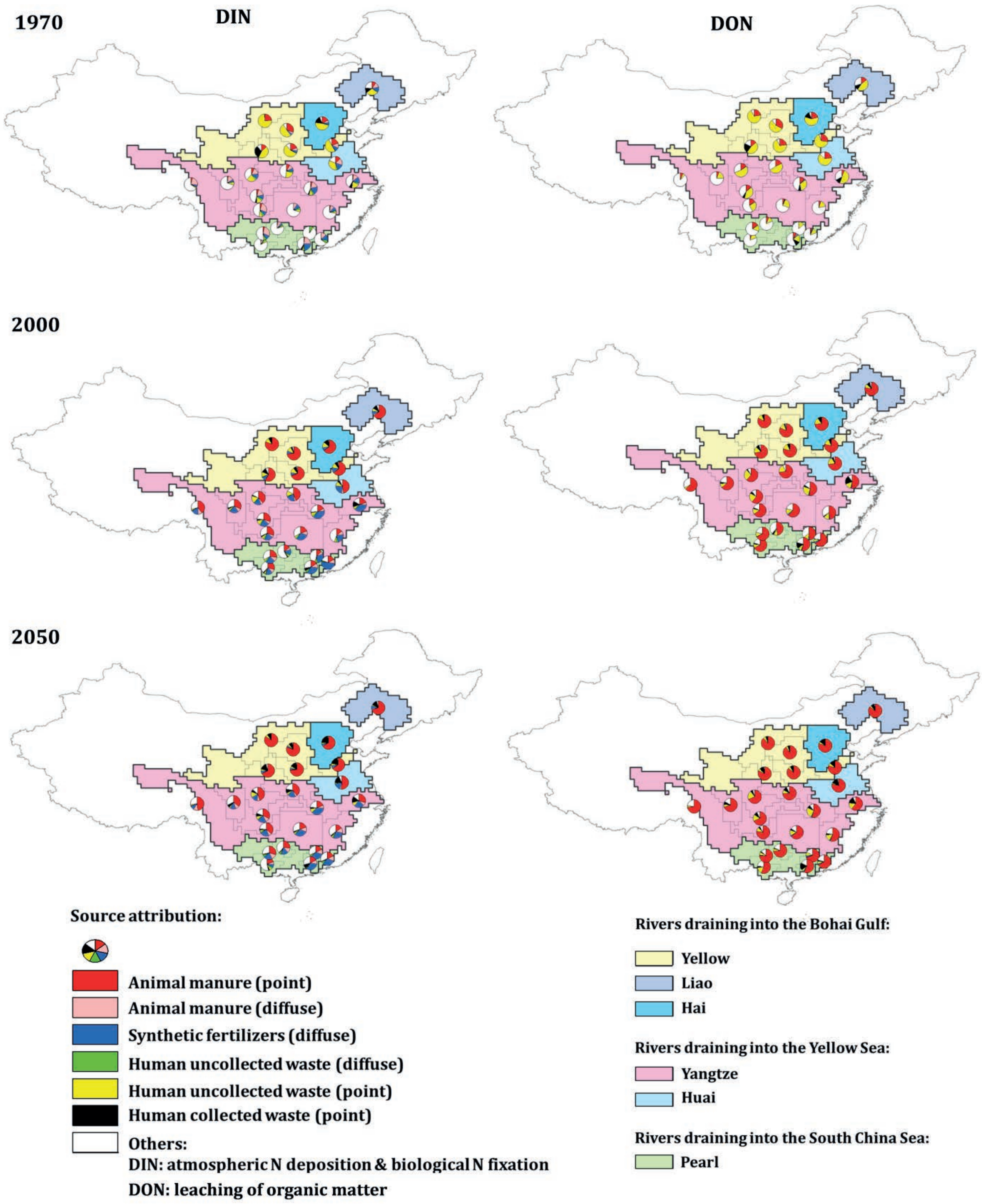

Figure 5.11. *The share of sources in dissolved inorganic (DIN) and organic (DON) nitrogen (N) export by the Chinese rivers draining into the Bohai Gulf, Yellow Sea and South China Sea in 1970, 2000 and 2050 (0-1). 2050 is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Seitzinger et al. 2010). See Section 5.2 for model description and inputs.*

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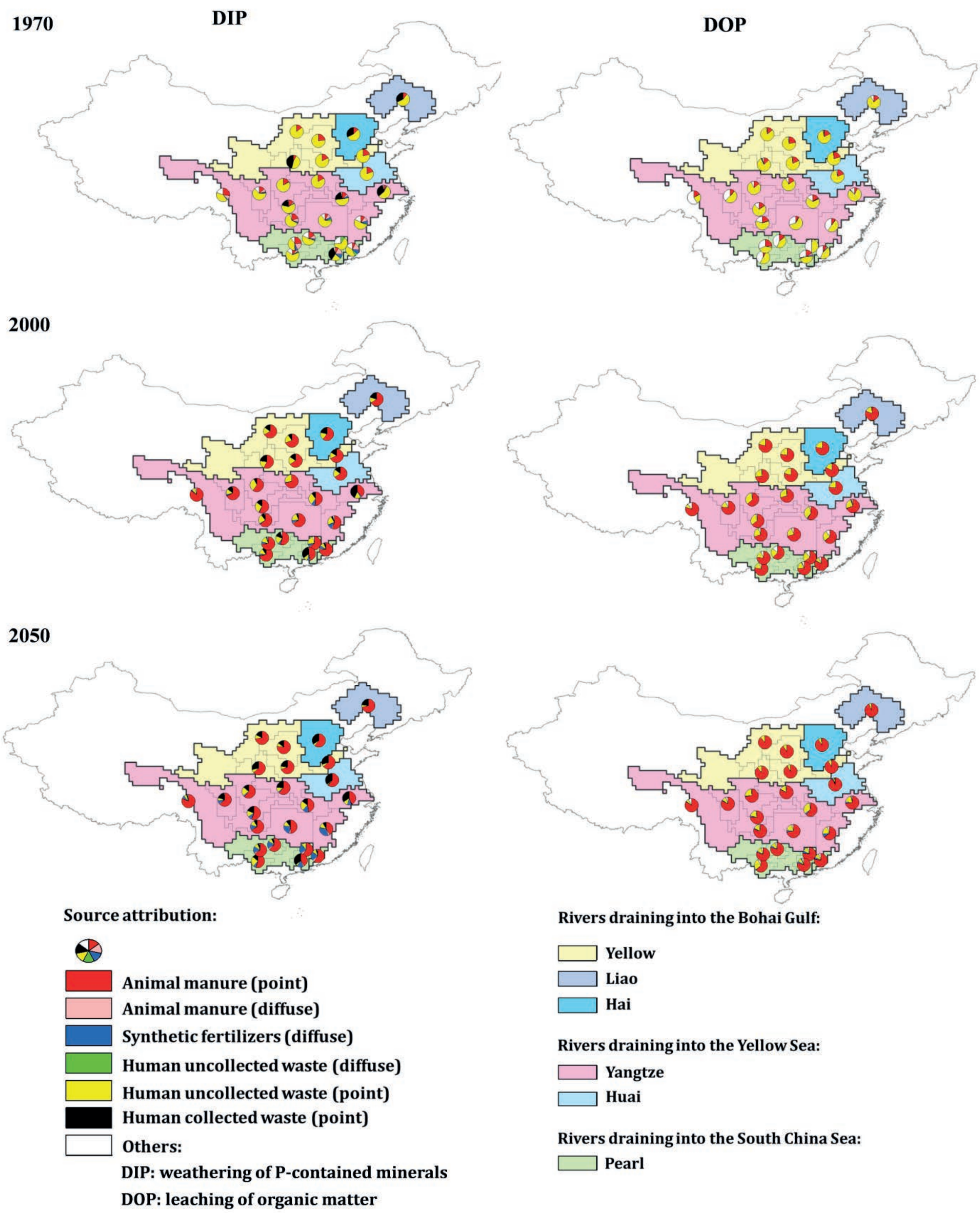

Figure 5.12. *The share of sources in dissolved inorganic (DIP) and organic (DOP) phosphorus (P) export by the Chinese rivers draining into the Bohai Gulf, Yellow Sea and South China Sea in 1970, 2000 and 2050 (0-1). 2050 is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Seitzinger et al. 2010). See Section 5.2 for model description and inputs.*

5.3.3 Increasing risks for coastal eutrophication as a result of increasing river export of nutrients

The results show that the risk of rivers to cause coastal eutrophication and thus harmful algae blooms increases from 1970 to 2050 (Figure 5.13). For 1970 ICEP values of the rivers ranged between -1.8 and 0.8 kg C-eq. km^2 $year^{-1}$. For 2000 these values were higher with a range of 2.4-11 kg C-eq. km^2 $year^{-1}$ among the rivers (Figure 5.13). We calculated N-ICEP for most of the rivers because N was limited (the Redfield ratio of N:P>16, see Figure S5.19). The Pearl and Liao (2000) rivers were P limited (N:P<16, Figure S5.19) and thus P-ICEP was calculated. Positive ICEPs indicate a high risk for harmful algae to develop in coastal waters as a result of N and P levels in excess over DSi. This holds in particulate for dissolved forms of N and P for which we calculated increasing trends between 1970 and 2000 (Section 5.3.2). To calculate ICEP we used river export of dissolved nutrients from this study, and of particulate N and P and DSi from Strokal et al. (2014b) (Section 5.2.3). The share of particulate N and P to the total N and P river export decreased between 1970 and 2000 (Figure S5.20). Strokal et al. (2014b) indicate that river export of particulate N and P to the Bohai Gulf decreased, and slightly increased to the Yellow and South China seas between 1970 and 2000. River export of DSi decreased during this period as well. Particulate N, P and DSi exports decreased because of river damming. However, the effect of river damming may be underestimated by Strokal et al. (2014b) compared to our study because we account for more dams (Figure S5.12). Nevertheless, the results of our study are generally in agreement with various existing studies that indicate current eutrophication problems in aquatic systems of China (Diaz & Rosenberg 2008; Huang et al. 2003; Li et al. 2014b; Wang et al. 2008; Xiao et al. 2007).

In the future the risk for coastal eutrophication may further increase. In the GO scenario rivers have higher values for N-ICEP or P-ICEP than in 2000 (Figure 5.13 and Figure S5.19). This is because rivers are projected to transport more dissolved nutrients (especially from animal production and urbanization; Section 5.3.2) than DSi (Strokal et al. 2014b) (Figure S5.19). As a result, the coastal waters of the Bohai Gulf, Yellow Sea and South China Sea may be at the risk for more eutrophication-related problems in the future.

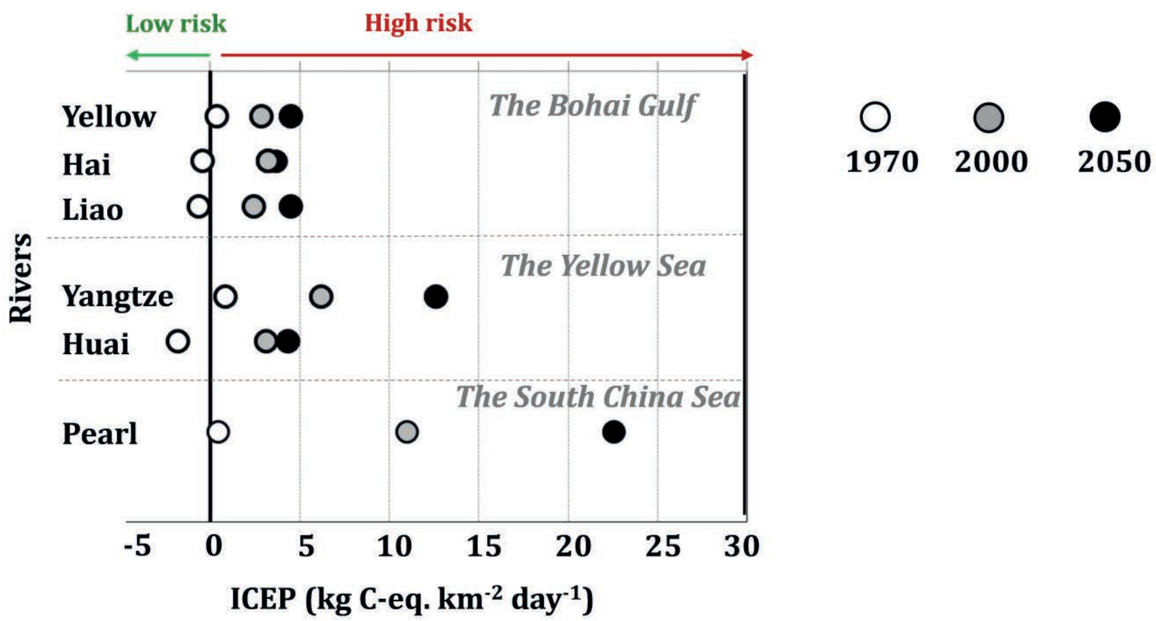

Figure 5.13. *Indicator for Coastal Eutrophication Potential (ICEP, kg C-eq. km^{-2} day^{-1}) for the Chinese rivers in 1970, 2000 and 2050. Positive ICEP indicates high risks for coastal eutrophication, and thus for blooms of harmful algae because of excess nutrients in coastal waters over DSi. Negative ICEP indicates low risks for coastal eutrophication. Either N- or P-ICEP is calculated depending on which nutrient (N or P) is limiting (see Figure S5.19). See Section 5.2.3 for ICEP description. 2050 is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Seitzinger et al. 2010).*

5.3.4 Comparison with other studies

We compared our results to other modeling studies (Li et al. 2011b; Qu & Kroeze 2010; Qu & Kroeze 2012; Strokal et al. 2015; Strokal et al. 2014b; Ti et al. 2012; Yan et al. 2010). These modeling studies indicate an increase in nutrient export by Chinese rivers since the 1970s, which is line with our findings.

Our estimates for nutrient export by individual rivers differ from estimates of Global *NEWS*-2 studies for China (Li et al. 2011b; Strokal et al. 2015; Strokal et al. 2014b; Yan et al. 2010). Strokal et al. (2014b), Qu and Kroeze (2010) and Qu and Kroeze (2012) analyzed nutrient export by sixteen rivers to Chinese seas using the original Global *NEWS*-2 model (Mayorga et al. 2010). We quantify higher nutrient export by Chinese rivers than those modeling studies. Exceptions are DIN export by the Yangtze, Huai and Hai rivers, and DIP export by the Yangtze (in 2000), Huai (in 2000), Hai and Liao for which our estimates are lower. For example, our estimates for 2000 are 3% higher for DIN and 24% for DIP exports by the Pearl than estimates of Strokal et al. (2014b), Qu and Kroeze (2010) and Qu and Kroeze (2012) (Table S5.11). For the Yangtze, our estimates for DIN are 55% lower, and for DIP 18% (Table S5.11). We have higher or lower values compared to Global *NEWS*-2 modeling studies because our model accounts for direct discharges of animal manure and human waste (increasing nutrient export), and for more dams (increasing nutrient retention).

Strokal et al. (2015) modeled DIN and DIP export by the Pearl River for 1970-2050 at the sub-basin scale with updated reservoir information, but without direct inputs of human waste and animal manure to rivers. This explains why our estimates are higher (375 kton of DIN and 21 kton of DIP in 2000) than by Strokal et al. (2015) (189 kton of DIN and 4.5 kton of DIP in 2000; see Table S5.11). Our study shows that middlestream activities are important contributors to DIN and DIP export by the Pearl River in addition to downstream activities. This is different from Strokal et al. (2015) where downstream activities play a dominant role. The difference is because in the current study we improved the sub-basin scale approach of Strokal et al. (2015) for nutrient export within the main channel towards the river mouth (Section 5.2 for methods).

Yan et al. (2010) and Li et al. (2011b) modeled DIN and DIP export by the Yangtze River using Global *NEWS*-2, but with the provincial information. We model higher DIP (33 kton) and lower DIN (556 kton) exports by the Yangtze River than these studies (22-25 kton of DIN and 1611 kton of DIN) (Table S5.11). The reason for these differences is that our model accounts for direct losses of animal manure and human waste to rivers (leading to higher values) and for nutrient retentions in rivers of the sub-basins (leading to lower values). These existing modeling studies quantify DIN and DIP export at the basin scale. We, however, account for differences in nutrient retentions among sub-basins.

We compared our results for coastal eutrophication with other world seas and oceans (Diaz & Rosenberg 2008; Garnier et al. 2010; Selman et al. 2008). Coastal waters of Europe (e.g., the Baltic Sea, North Atlantic EU) and of North America (e.g., the North Atlantic US) are eutrophied because of human activities. However, the risk for coastal eutrophication may become lower in 2050 (in the GO scenario) because of effectively implementing environmental policies (Garnier et al. 2010). This is different for China where coastal eutrophication may occur in the future if manure management is not improved (Section 5.3.3). Similar to China, coastal waters of Indian, South Atlantic and Arctic oceans may have higher risks for eutrophication in 2050 than in 2000. Possible reasons are projected intensive human activities in the coming years (Garnier et al. 2010). This also holds for the Black Sea where urbanization in addition to agriculture is an important cause of nutrient pollution (Strokal et al. 2014c).

5.4 Conclusions

We analyzed the main sources of dissolved inorganic (DIN, DIP) and organic (DON, DOP) N and P export by six large rivers at the sub-basin scale for 1970, 2000 and 2050. To this end, we developed the *MARINA* Nutrient Model to quantify river export of nutrients by source at the sub-basin scale. This was done by redesigning the existing Global *NEWS*-2 model to the sub-basin scale with updated information for reservoirs, and by accounting for direct losses of manure and human waste.

River export of dissolved N and P increased by a factor of 7-8 for the Bohai Gulf, a factor of 6-8 for the Yellow Sea and a factor of 2-4 for the South China Sea between 1970 and 2000. These increases are caused largely by manure losses to rivers. Most nutrients are exported by the Yangtze (>50%), Pearl (<40%) and Yellow (<12%). In 1970 uncollected sewage from rural people was the main source of most nutrients in rivers. Manure losses to rivers were small because of manure recycling on land. By 2000 manure losses became responsible for 60-78% of the nutrients in the Bohai Gulf, and for 20-74% of the nutrients in the Yellow Sea and South China Sea. These losses resulted from limited manure recycling on land, and poor manure management in animal production, leading to direct discharge of manure to rivers. Synthetic fertilizers contributed largely to DIN export by the Yangtze and Pearl. Sewage effluents from urbanized areas were important sources of DIP in downstream areas. The Yellow River exported up to 70% of DIN and DIP from the downstream sub-basin and of DON and DOP from middlestream sub-basins. The Yangtze and Pearl rivers exported over half of DIN, DIP, DON and DOP fluxes from both downstream and middlestream sub-basins with a higher contribution from the middlestream activities. In our scenario for 2050 most rivers transport more nutrients from animal production and urban areas, increasing the risk for coastal eutrophication. This risk, however, may be reduced by effective implementation of recent and future environmental policies.

Transport of nutrients from human activities on land to sea depends on hydrology, in-river retentions and losses, and traveling distance towards the sea. Rivers draining into the Bohai Gulf transport fewer nutrients than other rivers because their drainage areas have less precipitation and runoff. This also explains why these rivers receive fewer nutrients from diffuse sources such as synthetic fertilizers than the Yangtze and Pearl. Nutrient retentions in dammed reservoirs increased between 1970 and 2000 because more dams were constructed. An example is the large Three Gorges Dam located in the middle of the Yangtze main channel and reduces nutrient export towards the sea. Water consumption for irrigation removes nutrients from rivers. This consumption is higher in particular in sub-basins covering the North China Plain with intensive agriculture.

The *MARINA* Nutrient Model is the first sub-basin scale model for China to quantify river export of DIN, DIP, DON and DOP by source for the past and future. We validated the model using available measurements from the literature. The *MARINA* Nutrient Model performs reasonably well for its purpose (R_{NSE}^2 is 0.78, ME is 8%). However, the number of measurements is limited, making validation difficult, especially for dissolved organic N and P. Uncertainties in model results are associated with model inputs, parameters and approaches. We, thus, compared our model inputs with an independent Chinese dataset and performed a sensitivity analysis. This all builds trust in the model. Suggestions for future analyses include an analysis of uncertainties in the model. Re-calibrating model parameters for Chinese sub-basins is desirable and can improve modeling of river export of dissolved inorganic and organic nutrients. However, this will require more measurements of DIN, DIP, DON and DOP in river mouths. Despite these model limitations, we believe that our *MARINA* Nutrient Model provides useful quantitative information on the sub-basin scale analyses of river export of nutrients and their sources for the past and future.

Our study can help to develop effective nutrient management strategies in China. Effective manure management is important to reduce coastal water pollution. Awareness of coastal eutrophication has been increasing in China in recent years, as illustrated by recent policies aimed at reducing nutrient losses to the environment. We provide quantitative and spatially explicit information on the extent to which human activities contribute to nutrient pollution of the Chinese seas. This can help to allocate management options. For example, if we want to reduce DIN and DIP export by the Yellow River, it is more effective to invest in management of manure in the downstream sub-basins than in upstream. Improved manure management in both middlestream and downstream sub-basins can help to reduce DON and DOP export by large rivers. Urbanization will likely continue in China, as well as industrialization of animal production around cities. Our *MARINA* Nutrient Model can be helpful in exploring future development pathways that can be considered sustainable.

Acknowledgments

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Chapter 6.

Reducing future coastal water pollution in China in optimistic scenarios

Abstract

The coastal waters of China are rich in nitrogen (N) and phosphorus (P) and thus often eutrophied. This is because rivers export increasing amounts of nutrients to coastal seas. Animal production and urbanization are important sources of nutrients in Chinese rivers. In this study we explored the future from an optimistic perspective. We present two optimistic scenarios for 2050 (OPT-1 and OPT-2) for China. Maximized recycling of manure on land in OPT-1 and OPT-2, and strict sewage control in OPT-2 (e.g., all sewage is collected and treated efficiently) are essential nutrient strategies in these scenarios. We also analyzed the effect of the current policy (CP) plans aiming at "Zero Growth in Synthetic Fertilizers after 2020" (the CP scenario). We used the *MARINA* (a Model to Assess River Inputs of Nutrients to seAs) model to quantify dissolved N and P export by Chinese rivers to the Bohai Gulf, Yellow Sea and South China Sea and the associated coastal eutrophication potential (ICEP). The Global Orchestration (GO) scenario of the Millennium Ecosystem Assessment was used as a basis. GO projects increases in river export of dissolved N and P (up to 89%) between 2000 and 2050 and thus a high potential for coastal eutrophication (ICEP>0). In contrast, the potential for coastal eutrophication is low in optimistic scenarios (ICEP<0). This is because in 2050 loads of most dissolved N and P in Chinese seas are around their 1970 levels. Maximizing manure recycling can reduce nutrient pollution of Chinese seas considerably. Sewage control is effective in reducing P export by rivers from urbanized areas. The CP scenario, on the other hand, shows that current policy plans may not be sufficient to avoid coastal eutrophication in the future. Our study may help policy makers in formulating strategies to ensure clean coastal waters in China in the future.

Under revision as:

Strokal M, Kroeze C, Wang M, Lin M. Reducing future coastal water pollution in China in optimistic scenarios. This manuscript is under revision after positive review comments. It will be resubmitted soon to the Science of the Total Environment

6.1 Introduction

Rivers supply increasing amounts of nitrogen (N) and phosphorus (P) nutrients to the coastal waters of China (Huang et al. 2003; Li et al. 2014b; Strokal et al. 2015; Sumei et al. 2008). This causes eutrophication and blooms of harmful algae (Liu & Diamond 2005; Qu & Kroeze 2012; Wang 2006; Xiao et al. 2007; Zhang et al. 2013). These environmental problems pose a threat to ecosystems, and decrease the suitability of water for, for instance, fishing and recreation (Vörösmarty et al. 2010). Food production and urbanization contribute largely to these nutrient pollution problems where animal production plays a leading role (Bai et al. 2015; Liu & Diamond 2005; Ma et al. 2012; MEP et al. 2010; Strokal et al. 2016b). Chinese agriculture has been under transitions: changes in farming practices from traditional- to industrial-dominated systems (Strokal et al. 2016b). Animal production started industrializing since the 1990s to produce more food especially for cities. Large industrial animal farms became disconnected from crop production. Thus animal manure is currently not applied often on land, but largely discharged to nearby water systems, causing nutrient pollution (Bai et al. 2015; Ma et al. 2012; MEP et al. 2010; Strokal et al. 2016b). These point source inputs of manure to rivers exceed the diffuse losses of nutrients from fertilized soils in large parts of China (Strokal et al. 2016b). Human waste is another important source of increasing nutrients in rivers. Sewage systems discharge in many rivers and are an important source of dissolved inorganic P in rivers (Strokal et al. 2015; Strokal et al. 2014b; Van Drecht et al. 2009). Rural people in China, however, often lack sewage connections. For example, in 2000 less than 5% of rural people were connected to sewage systems (WHO/UNICEF 2014). As a result, human waste is often also directly discharged to rivers, but untreated. Thus, human excreta pollute rivers all over China (Ju et al. 2005; Morée et al. 2013; Strokal et al. 2014b).

In the future, the potential for coastal water pollution may increase. This is because both industrialization of animal production and urbanization will likely further develop. River pollution and thus coastal eutrophication will depend on how efficiently nutrients are managed in both sectors. Various studies analyzed different scenarios for nutrient management in China. However, these studies are either limited to agriculture and fresh waters (no connection with coastal waters) or do not account for the effects of industrialized animal production on coastal waters in China. For example, several studies (Bai et al. 2015; Ma et al. 2013a; Ma et al. 2013b) analyzed the effects of efficient nutrient management in animal (e.g., precisions feeding, more manure use on land instead of synthetic fertilizers) and crop production (e.g., balanced fertilization) on nutrient losses to fresh waters by 2030. However, their national and provincial analyses do not account for urbanization. Other studies (Qu & Kroeze 2010; Qu & Kroeze 2012) analyzed the effects of nutrient management strategies in agriculture and urbanization on reducing river export of nutrients and thus coastal water pollution by 2050. However, they do not account for the effects of industrialized animal production in China. As a result these studies may underestimate nutrient pollution and thus may overestimate the effectiveness of their strategies.

In this study we explore the future from an optimistic perspective to reduce river export of nutrients and thus coastal eutrophication in China by 2050. Optimistic perspectives are limited in existing scenario studies (e.g., Ma et al. 2013b; Qu & Kroeze 2012). We developed optimistic scenarios that assume more efficient nutrient management in agriculture and sewage than typical business as usual scenarios. In addition, we analyzed effects of the recently introduced policy on "Zero Growth in Synthetic Fertilizers after 2020" aimed at reducing water pollution from agriculture by increasing nutrient use efficiencies (MOA 2015). We compared effects of this policy with effects of optimistic scenarios, and discuss whether this policy is sufficient to reduce coastal water pollution in 2050. We used the Global Orchestration (GO) scenario of the Millennium Ecosystem Assessment as a basis for our scenarios because it assumes an economy-driven world with a rapid urbanization (see Section 6.2.2). We used the *MARINA* Nutrient Model (a Model to Assess River Inputs of Nutrients to seAs) to quantify river export of nutrients and the associated coastal eutrophication (Strokal et al. 2015; Strokal et al. 2016a; Strokal et al. 2016b).

6.2 Methodology

6.2.1 Model description

The *MARINA* Nutrient Model quantifies the river export of dissolved inorganic (DIN and DIP) and dissolved organic (DON and DOP) N and P by source from sub-basins for 1970, 2000 and 2050. The model was developed by Strokal et al. (2016a) based largely on approaches and information of the Global *NEWS*-2 model (*N*utrient *E*xport from *W*ater*S*heds) (Mayorga et al. 2010). The *MARINA* Nutrient is described in earlier studies including model validation (Strokal et al. 2015; Strokal et al. 2016a; Strokal et al. 2016b) (see Text S6.1 in the Supplementary Materials on details). The model is implemented for six large rivers draining into the coastal waters of China (Figure 6.1). The Yellow (Huanghe), Hai and Liao drain into the Bohai Gulf, and the Yangtze (Changjiang) and Huai drain into the Yellow Sea. The South China Sea receives nutrients from the Pearl River. The drainage areas of the Yangtze, Yellow and Pearl rivers are divided into up-, middle- and downstream sub-basins, based on literature (Cui et al. 2007; Huang et al. 2009a; Niu & Chen 2010; Wang et al. 2010; Yang & Lu 2014b; Zhou et al. 2013). The amount of nutrients reaching coastal waters from each sub-basin depends on human activities on land (e.g., agriculture, sewage), sub-basin characteristics (e.g., land use, retentions in soils), and retentions and losses of nutrients within the river network (see Text S6.1 in the Supplementary Materials).

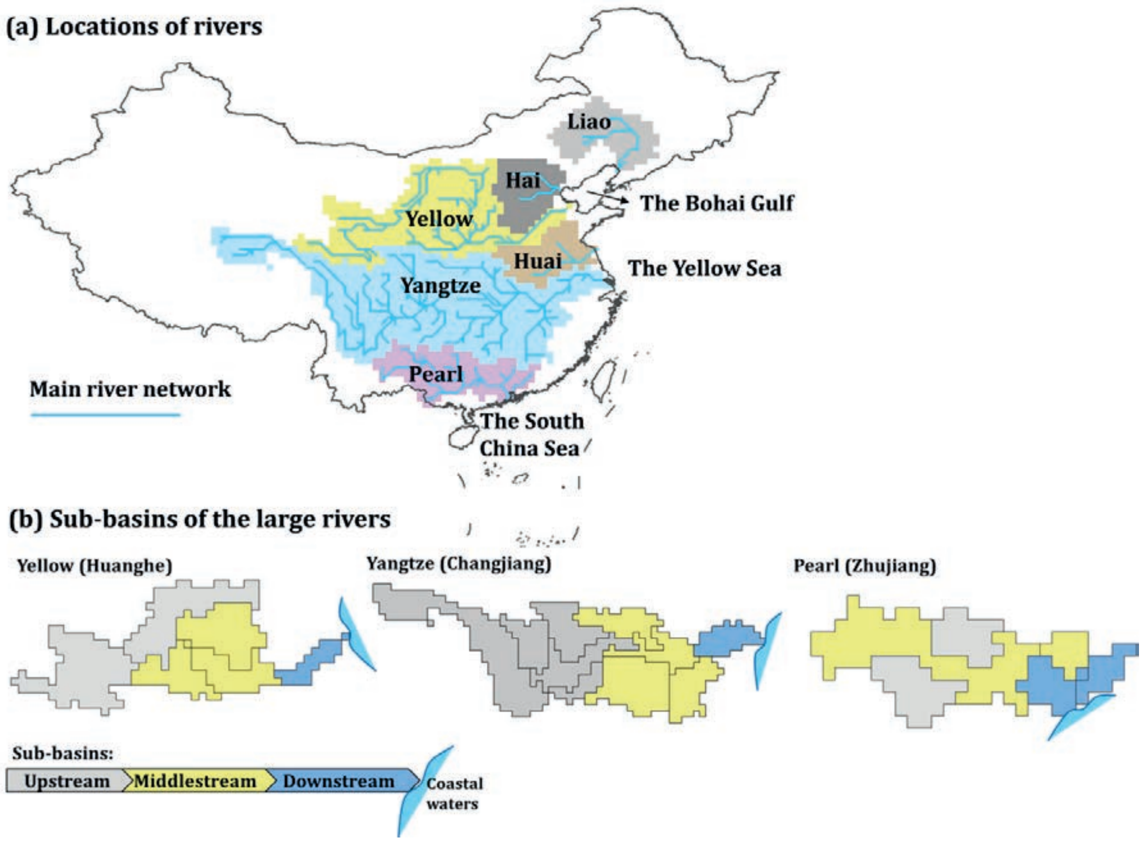

Figure 6.1. *(a) Locations of rivers and their drainage areas in China, and (b) sub-basins of the large rivers. Drainage areas of the sub-basins are from the Simulated Topological Network (STN-30, v6.01) (Mayorga et al. 2010; Vörösmarty et al. 2000b).*

Human activities add nutrients to Chinese rivers. We distinguish between diffuse and point sources of nutrients in rivers. Diffuse sources are the use of synthetic fertilizers, animal manure and human excretion on land, atmospheric N deposition (for DIN) and biological N fixation (for DIN). Nutrient inputs to rivers from these sources are quantified by correcting nutrient inputs to land for crop harvesting and animal grazing, and for nutrient retentions in soils. Weathering of P-contained minerals is a diffuse source of DIP in rivers while leaching of organic matter is for DON and DOP. Nutrient inputs to rivers from these sources are quantified as a function of annual runoff (details are in Strokal et al. 2016b and Strokal et al. 2016a). Point sources of nutrients include direct discharges of animal manure to rivers and of uncollected human waste from rural and urban populations as well as sewage effluents and detergents (for only DIP, DOP). Direct discharges of animal manure are calculated by multiplying animal excretion with the fraction of direct manure discharges. Direct discharges of uncollected human waste are calculated in a similar way. Sewage effluents and detergents result from the urban population that is connected to sewage systems. Some N and P in sewage influents are removed during waste water treatment (depending on the efficiency of treatment) and the remainder are discharged to rivers (details are in Strokal et al. 2016b, Strokal et al. 2016a and the Supplementary Materials).

Nutrients are retained and/or lost within the river network of sub-basins as a result of water consumptions (for nutrient forms), denitrification (for DIN), sedimentation processes (for DIP) and by construction of dams (for DIN and DIP). In the *MARINA* Nutrient Model, the nutrient retentions and losses are quantified according to the Global *NEWS*-2 model (Mayorga et al. 2010), but with updated information for reservoirs and included sedimentation for DIP in rivers (see Strokal et al. 2016a). Nutrients in rivers of upstream sub-basins travel in general longer towards coastal waters (the river mouth) than nutrients in rivers of middle- and downstream sub-basins. During this traveling some nutrients are lost or/and retained in the river before reaching coastal waters (see Text S6.1 in the Supplementary Materials).

We used the *MARINA* Nutrient Model to quantify the Indicator for Coastal Eutrophication Potential (ICEP, expressed in kg C-eq. km^{-2} day^{-1}) (Garnier et al. 2010). ICEP indicates a high potential for coastal eutrophication (ICEP>0) when rivers supply total N (TN) and total P (TP) in excess over dissolved silica (Si). TN and TP are the sum of dissolved inorganic, dissolved organic and particulate forms of nutrients. As a result, harmful algae develop fast, considering nutrient requirements for phytoplankton growth (the Redfield ratio of C:N:P:Si = 106:16:1:20). Either N- or P-ICEP is calculated depending on limiting nutrient (see details in Garnier et al. (2010)).

In the *MARINA* Nutrient Model most model inputs to quantify river export of nutrients for 1970, 2000 and 2050 were derived from the gridded datasets (0.5 by 0.5 degree cell) of Global *NEWS*-2 (Bouwman et al. 2009; Fekete et al. 2010; Mayorga et al. 2010; Seitzinger et al. 2010; Van Drecht et al. 2009). These gridded datasets (e.g., synthetic fertilizer use, manure, atmospheric N deposition etc.) were aggregated to sub-basins in ArcGIS. Inputs for reservoirs were from the Global Reservoir and Dam Database (GRanD) (Lehner et al. 2011a; Lehner et al. 2011b). Model parameters to account for manure point sources were from Ma et al. (2012). Model parameters to account for management of uncollected human waste were from Ma et al. (2012) for rural population and from Morée et al. (2013) for urban population. Details on sources of model inputs and parameters are given in Strokal et al. (2016a). Supplementary Materials provide inputs for main model variables (Figures S6.3-S6.12).

6.2.2 Scenario description

We developed three alternative scenarios for 2050 using GO as a starting point: CP (current policy), OPT-1 (optimistic 1) and OPT-2 (optimistic 2). In the *MARINA* Nutrient Model (Strokal et al. 2016a) the original GO scenario of Global *NEWS*-2 (Alcamo et al. 2005; Seitzinger et al. 2010) was slightly modified to account for industrialization trends in Chinese animal production and for the management of uncollected human waste. Strokal et al. (2016b) provide details on GO and the Supplementary Materials provide details on the alternative scenarios (Figures S6.1-S6.11 and Tables S6.1 and S6.2). Here we summarize the scenarios in Figure 6.2 and Table 6.1. GO is considered as the worst case for the environment whereas OPT-1 and OPT-2 are the best cases.

<u>The GO scenario</u> assumes globalization trends for socio-economic development, a reactive approach in environmental management (Alcamo et al. 2005; Seitzinger et al. 2010), further industrialization of animal production and urbanization (Table 6.1). Animal numbers will increase, producing more manure (doubling between 2000 and 2050, Figures S6.3-S6.5). More industrial farms will emerge to grow animals, but with manure management that will not differ largely from today: considerable amounts of manure will be ignored as a fertilizer, increasing losses of nutrients to the environment (details in Strokal et al. 2016b and Strokal et al. 2016a). Use of synthetic fertilizers is projected to increase by a factor of 1.5-3.2 between 2000 and 2050 (Bouwman et al. 2009, Figures S6.6-S6.8). On the other hand, people are assumed to be moving to urban areas for better jobs, increasing the demand for food and producing more human waste (Figure S6.9, Table 6.1). However, not all human waste will be collected by sewage systems and nutrient removal during treatment will remain moderate (secondary-oriented treatment with around 40-50% of N and P removal, Van Drecht et al. (2009), Figures S6.9-S6.11). As for manure, management of uncollected human waste from urban and rural areas will not differ largely from today: some human excretion is assumed to remain on land (mainly in rural areas), some is lost to the air (for N as a result of poor sanitation) and some is discharged to nearby water systems as in 2000, according to Ma et al. (2012) (Strokal et al. 2016a, Figures S6.10 and S6.11). Furthermore, in the future more dams are assumed to be constructed and consumption of water for, for example, irrigation may increase especially in drier basins (e.g., Yellow, Hai, Liao) (see details in Fekete et al. (2010) on assumptions for hydrology and climate). All this will affect the nutrient retention in rivers (see Figure S6.12).

<u>The CP scenario</u> differs from GO in that it assumes implementation of the recent policy on “Zero Growth in Synthetic Fertilizers after 2020”. This policy aims at zero growth in the use of synthetic fertilizers from the year 2020 (to avoid over-fertilization), and 60% recycling of available manure on land (to avoid manure losses to rivers) (MOA 2015,

Table 6.1 and Figure 6.2). We thus assumed that between 2020 and 2050 the use of synthetic fertilizers will stay at the level of 2020 and 60% of available manure (after correcting for N losses) will be applied on land (see details on assumptions in Tables S6.1 and S6.2). The policy does not explicitly describe how to manage the remaining manure. We assume that it is discharged to aquatic systems (and to air for N, Figure 6.2, Figures S6.3-S6.5).

<u>The OPT-1 scenario</u> describes an optimistic world assuming full implementation of efficient nutrient management in agriculture (Table 6.1, Figure 6.2). It assumes maximum manure use on land and the full use of best available technologies to reduce nutrient losses to the environment (see references below). In this world people will use nutrients as efficiently as possible while maintaining crop yields and avoiding losses of nutrients to water systems from both crop and animal production.

N and P excretion by animals is 20% (N) and 30% (P) lower than in GO as a result of improved animal feeding (e.g., via precision feeding) based on Ma et al. (2013a) (Tables S6.1 and S6.2). Ammonia emissions during storage and housing are assumed to be reduced by 50% relative to GO as a result of mitigation options (e.g., covering storage facilities, collecting urine rapidly, and decreasing areas with uncovered manure storage based on different studies (Bittman et al. 2014; Loyon et al. 2016; Oenema et al. 2012). The available manure is assumed to be managed in such a way that all P and almost all N in manure will be applied on land as fertilizers (only some N will be lost during treatment and/or transportation, and application of manure will be via injection to avoid nutrient losses to the environment). This is because in this scenario animal manure is considered important as nutrient fertilizer for crops and to increase the quality of soils (e.g., via increasing organic matter) as indicated in recent experimental studies for China (Qin et al. 2015; Wang et al. 2016) (Tables S6.1 and S6.2).

We assume that animal production will be strongly connected to crop production in this scenario. The amount of synthetic fertilizers needed for crops is determined based on available manure and other sources of nutrients (atmospheric N deposition, biological N fixation, human excretion). We quantified percentages of synthetic fertilizer reductions relative to CP based on information from Ma et al. (2013a) and Wang et al. (under review) who used a balanced fertilization method, in which the amount of nutrients applied to land follows from crop requirements (see Table S6.1 for details on calculations). Following these assumptions, the synthetic fertilizer use in OPT-1 is 52% lower for N and 58% for P relative to CP (average for 25 sub-basins, see Figure S6.1 for sub-basin values).

The OPT-2 scenario differs from OPT-1 in that it also assumes efficient nutrient management for sewage (Figure 6.2, Table 6.1, Tables S6.1 and S6.2). Agriculture develops as in OPT-1. For sewage the scenario assumes full access to improved sanitation. We assume that decentralized sanitation systems will be implemented fully in rural areas (Massoud et al. 2009; Oakley et al. 2010; Singh et al. 2015). Treated effluents from these decentralized systems are assumed to be reused for irrigation in agriculture. Urban population will have a full access to centralized sewage systems. We assume the implementation of new technologies (e.g., Khiewwijit 2016) in both decentralized (in rural areas) and centralized (in urban areas) systems that remove 80% of N and 90% of P from sewage influents. We assume that the Chinese government will ban P-based detergents. All P-detergents will be replaced by non-P detergents. This could be realized by adopting European practices where some countries have moved to Zeolite, which is a P-free detergent (Glennie et al. 2002).

Table 6.1. *Main characteristics of scenarios for agriculture and sewage control in China for 2050. See Tables S6.1 and S6.2 for details on assumptions.*

Scenario	Main characteristics
GO	*Global Orchestration: globalization trends with reactive environmental management*
	Agriculture: o Industrialization of animal production o Limited manure application on land o Ongoing increase in fertilizer use until 2050 Sewage control: o Urbanization o Not all urban people connected to sewage systems o Rural people without access to sewage systems o Moderate efficiency of nutrient removal during treatment (secondary-oriented)
CP	*Current Policy plans: focus on the "Zero Growth in Synthetic Fertilizer after 2020" policy*
	Agriculture: o Enhanced manure application on land o Zero increase in synthetic fertilizer use after 2020
OPT-1	*Optimistic scenario 1: focus on efficient nutrient management in agriculture*
	Agriculture: o Maximized manure application on land o Improved animal feeding and facilities to store and collect manure o Decreasing synthetic fertilizer use after 2020 to avoid over-fertilization of soils
OPT-2	*Optimistic scenario 2: focus on efficient nutrient management in agriculture and in sewage*
	Agriculture as in OPT-1 Strict sewage control: o Decentralized sewage systems for collecting all rural waste o Centralized sewage systems for collecting all urban waste o High efficiency of nutrient removal during treatment (tertiary-oriented) o Phase out of P-based detergents

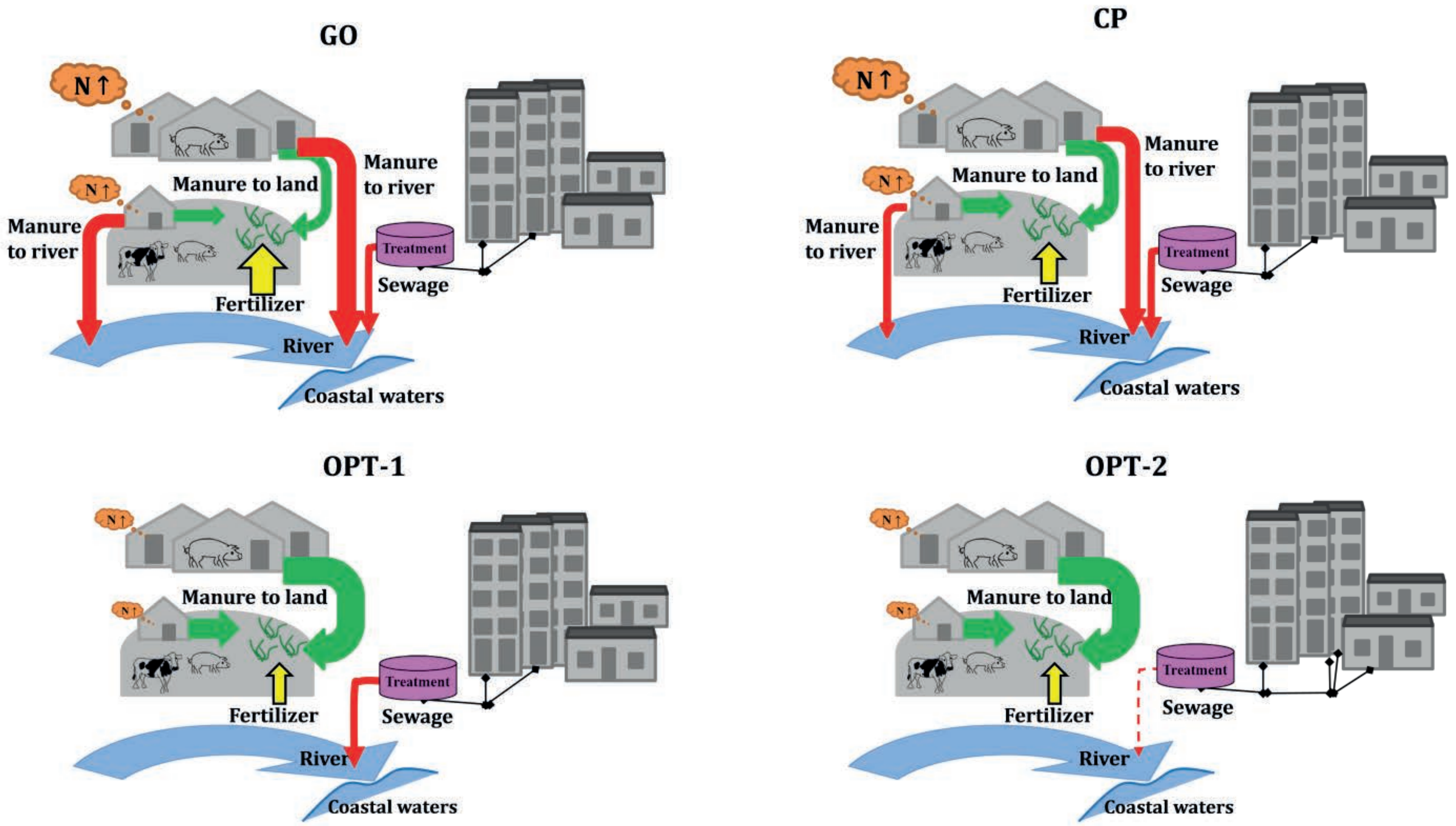

Figure 6.2. *Overview of scenarios for agriculture and sewage control in 2050. CP, OPT-1 and OPT-2 are alternative scenarios, based on Global Orchestration (GO) of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Mayorga et al. 2010; Seitzinger et al. 2010). CP is a scenario that incorporates roughly current policy plans for agriculture. OPT-1 and OPT-2 are optimistic scenarios that assume full implementation of improved nutrient management in agriculture (OPT-1 and OPT-2) and highly efficient sewage management (OPT-2). See also Table 6.1 and Tables S6.1 and S6.2 for details on scenario assumptions.*

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6.3 Reducing future coastal water pollution in China

The loads of N and P in Chinese rivers are at alarming levels. This is mainly because of agricultural transitions increasing the direct discharge of animal manure in rivers (Strokal et al. 2016b). Thus the potential for coastal eutrophication is high. We show that by 2000 dissolved N and P export by rivers to the Bohai Gulf had increased by a factor of 6-7, to the Yellow Sea by a factor of 2-5 and to the South China Sea by a factor of 2-4 relative to 1970 (ranges are for nutrient forms, Figure 6.3, Text S6.2). As a result, in 2000 the Bohai Gulf received 83-212 kton of N and 3-41 kton of P, the Yellow Sea received 561-594 kton of N and 37-112 kton of P, and the South China Sea received 182-375 kton of N and 21-33 kton of P (the ranges are for dissolved inorganic and dissolved organic forms) (Figure 6.3, Text S6.2). ICEP values are positive for 2000, indicating the potential for coastal eutrophication (Figure 6.6). Strokal et al. (2016a) show details in past trends in nutrient export by rivers to Chinese seas and main causes of the increasing trends. We summarized these trends in the Supplementary Materials (Text S6.2).

In 2050, the projected coastal water pollution is low, but only in optimistic scenarios (OPT-1 and OPT-2, Figures 6.3-6.5). The GO scenario, which is the worst case for the environment, projects a high potential for coastal eutrophication (ICEP>0, Figure 6.6). This is because rivers are projected to export roughly 10-90% more nutrients in 2050 than in 2000 (range for rivers, Figure 6.3). An exception is DIN export by the Hai River to the Bohai Gulf for which we calculate a decrease between 2000 and 2050 because of river damming. The large Yellow, Yangtze and Pearl rivers are projected to transport relatively large amounts of dissolved N and P to the Bohai Gulf, Yellow Sea and South China Sea, respectively (Figures 6.3-6.5, Figure S6.14). Over half of the nutrients from these rivers may result from middle- and downstream activities (Figure S6.15). Manure point sources will remain a major polluter of rivers (except for DIN) because animal production will continue to industrialize without proper manure management (Figures 6.4, 6.5, and S6.14). Synthetic fertilizers will further contribute to DIN inputs in the Yangtze and Pearl. The share of urban human waste in river pollution is projected to increase (Figure S6.14) especially for DIP in rivers of downstream sub-basins (Figure 6.5, Figure S6.15). This is driven by urbanization (Figure S6.9).

Implementing current policy plans in agriculture (the CP scenario) may not be effective enough to reduce future nutrient pollution of most Chinese rivers (Figures 6.3-6.5). Thus the high potential for coastal eutrophication may remain in 2050 (ICEP>0, Figure 6.6). In the CP scenario nutrient export by most rivers is projected to increase between 2000 and 2050, but not as fast as in GO. In particular P export is lower (Figures 6.3-6.5, S6.14). The differences between CP and GO are associated with manure recycling on land and use of synthetic fertilizers in 2050. CP incorporates the recent policy (MOA 2015) in which manure recycling (60%) is somewhat higher than in GO (around 50%, Table S6.1). Thus in CP the contribution of manure point sources to river pollution is somewhat lower than in GO. This explains why in CP nutrient export by most rivers is only slightly lower than in GO. In contrast, DOP export by Hai and Huai is decreasing between 2000 and 2050 in CP. Thus recycling 60% of the manure may be enough to reduce DOP export by these rivers. Synthetic fertilizers remain important sources of DIN in the Yangtze and Pearl rivers, as in GO. This is because the use of N synthetic fertilizers in CP is comparable to that in GO (Figure S6.8). This is different for P synthetic fertilizers. CP projects much lower use of P fertilizers than GO. This explains why effects of this policy on DIP export by the Yangtze and Pearl Rivers are more visible than on other nutrient forms (Figures 6.3-6.5, S6.14). In other rivers the majority of DOP and DIP is from point sources (Figure 6.5) and thus effects of reduced P fertilizers on land is negligible.

In scenarios OPT-1 and OPT-2, river export of nutrients is projected to reduce to levels with low potentials for coastal eutrophication (ICEP<0, Figures 6.3-6.6). For 2050 these optimistic scenarios project river export of most nutrients to be at around levels of 1970 (Figures 6.3, S6.14). Only DIN export by the Yangtze and Pearl is projected to be at around levels of 2000 (Figures 6.3-6.5). Low river export of nutrients in OPT-1 is largely associated with improved manure management (Tables 6.1, S6.1 and S6.2). In particular, maximum recycling of the manure on land avoids direct discharge of manure to Chinese rivers.

As a result, the nutrient pollution of Chinese seas reduced considerably (Figures 6.3-6.5, S6.14). Furthermore, the use of N and P synthetic fertilizers is projected much lower in OPT-1 than in GO and CP (Figures S6.6-S6.8, Tables 6.1, S6.1 and S6.2). This further reduces DIN export by the Yangtze and Pearl rivers in 2050 for which synthetic fertilizers are important sources (Figure S6.14). In OPT-1 human waste becomes the dominant source of nutrients in many rivers (Figures 6.4 and 6.5). Leaching of organic matter is an important source of DON in OPT-1 and for biological N fixation and atmospheric N deposition of DIN in the Yangtze and Pearl.

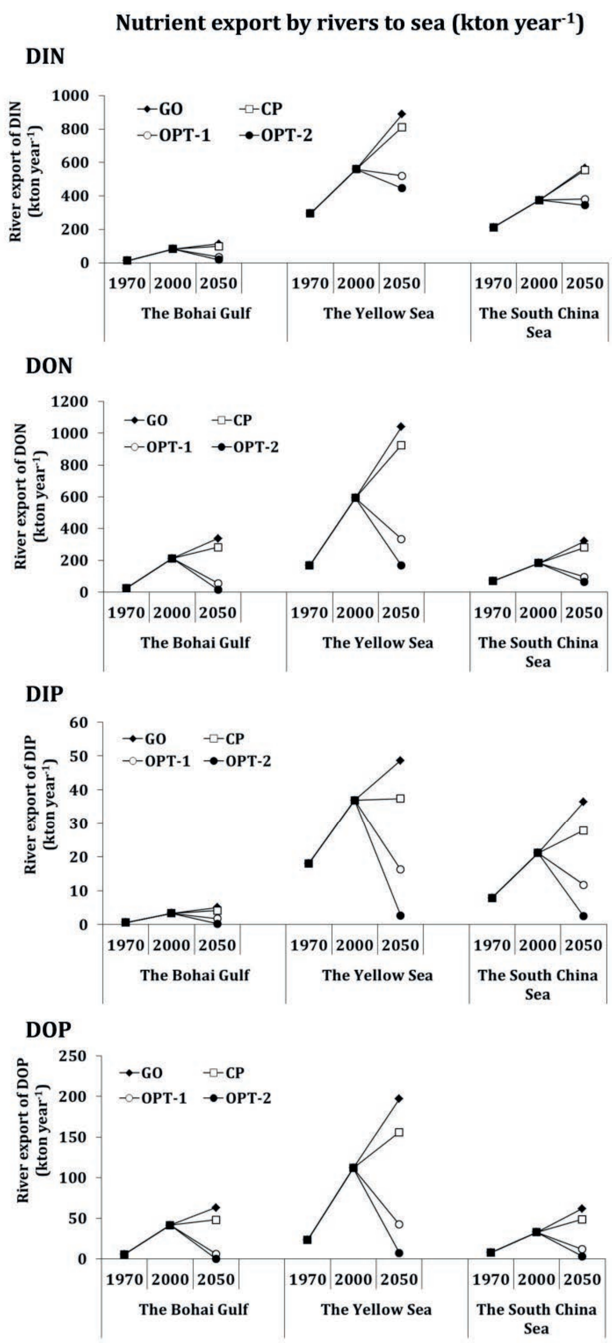
In OPT-2, low river export of nutrients in 2050 is a result of assumptions on the efficient use of nutrients both in agriculture (as in OPT-1) and sewage (Tables 6.1, S6.1 and S6.2). In general, this OPT-2 scenario projects lower river export of most nutrients than OPT-1 (Figures 6.3-6.5). This is because OPT-2 assumes full access to improved sanitation with efficient nutrient treatment in centralized (for urban people) and decentralized (for rural people) systems as well as banned P-based detergents. The effect of this improved sanitation is larger on P export by rivers from urbanized sub-basins. In 2050, urbanization may especially be higher in downstream sub-basins (e.g., Hai, Huai, downstream sub-basins of the Yellow, Yangtze and Pearl rivers), and in some middlestream sub-basins of the Yangtze and Yellow rivers (see Figure 6.1 for location of sub-basins, Figures S6.9-S6.11 for urbanization). In OPT-2 the share of other sources in nutrient export (e.g., leaching of organic matter, P weathering, biological N fixation and atmospheric N deposition) may increase in 2050 especially for rivers of the Yellow Sea and South China Sea (Figures 6.4, 6.5, and S6.14).

6.4 Concluding remarks

In our optimistic scenarios river export of most nutrients is projected to be back to levels around 1970 (except for DIN export by the Yangtze and Pearl, which is projected to be back to levels around 2000). Thus the potential for coastal eutrophication is low (ICEP<0). Our study shows the importance of manure recycling to avoid direct discharges of manure to rivers and thus to achieve the low potentials for coastal eutrophication. If these discharges are not avoided, it will be difficult to reduce river and coastal water pollution in China. We also show the importance of efficient sewage management in urban areas.

Our optimistic scenarios demonstrate that it is technically possible to reduce pollution in aquatic systems in China to very low levels. However, one may question whether full implementation of high recycling rates of animal manure and high efficiencies of nutrient removal in sewage during treatment is feasible. It may not always be easy to realize in practice. Nevertheless, we believe it is not impossible by 2050. Our optimistic scenarios only consider technologies that are already currently available (Bittman et al. 2014; Burton & Turner 2003; Jarret et al. 2012; Khiewwijit 2016; Loyon et al. 2016; Magrí et al. 2013; Oenema et al. 2012). For example, technologies for manure management (e.g., composting, anaerobic digestion) and application on land (e.g., deep and shallow injection, even distribution over periods and crops) have been successfully applied in several parts of the world (especially in European countries) (Bonmatí et al. 2003; Burton & Turner 2003; Loyon et al. 2016; Magrí et al. 2013; Oenema et al. 2009). Likewise, recent technological developments (e.g., Khiewwijit 2016) increase the efficiency of nutrient removal in sewage during treatment by up to 90%.

Environmental awareness in China has been increasing. Nevertheless, facilitating the implementation of best available technologies is needed. This could be done by, for instance, policy support for manure processing, transportation and application. Advisory services would also help farmers to change their practices. The economy in China develops fast, opening an opportunity to implement best available technologies. Thus, our optimistic scenarios may be challenging, but not impossible to realize. We, therefore, consider them not only technically effective, but also practically feasible. Our results may support decision making on strategies to ensure clean rivers and coastal waters in China in the future.



Changes in nutrient export by individual rivers between 2000 and 2050 (%)

DIN

River	GO	CP	OPT-1	OPT-2	Sea
Yellow	31	12	-59	-79	The Bohai Gulf
Hai	-62	-67	-88	-95	
Liao	72	54	-48	-67	
Yangtze	59	45	-7	-20	The Yellow Sea
Huai	25	16	-42	-59	
Pearl	51	48	1	-8	The South China Sea

DON

River	GO	CP	OPT-1	OPT-2	Sea
Yellow	69	39	-74	-93	The Bohai Gulf
Hai	27	6	-74	-92	
Liao	75	55	-74	-90	
Yangtze	84	66	-38	-68	The Yellow Sea
Huai	27	5	-72	-89	
Pearl	76	54	-47	-65	The South China Sea

DIP

River	GO	CP	OPT-1	OPT-2	Sea
Yellow	50	21	-48	-93	The Bohai Gulf
Hai	42	20	-44	-93	
Liao	77	50	-50	-95	
Yangtze	30	1	-58	-93	The Yellow Sea
Huai	51	24	-41	-92	
Pearl	72	32	-44	-89	The South China Sea

DOP

River	GO	CP	OPT-1	OPT-2	Sea
Yellow	61	21	-83	-99	The Bohai Gulf
Hai	20	-8	-89	-99	
Liao	71	37	-86	-99	
Yangtze	89	51	-56	-92	The Yellow Sea
Huai	13	-18	-93	-99	
Pearl	88	48	-64	-90	The South China Sea

Changes in nutrient export between 2000 and 2050 (%):	Increase	Decrease
	> 50%	< -50%
	15 - 50%	-15 - -50%
	0 - 15%	0 - -15%

Figure 6.3. *River export of dissolved inorganic (DIN, DIP) and organic (DON, DOP) nitrogen (N) and phosphorus (P) to the Bohai Gulf, Yellow Sea and South China Sea in 1970, 2000 and 2050 (shown in graphs, kton year*$^{-1}$*), and changes in nutrient export by individual rivers between 2000 and 2050 (shown in tables, %). CP, OPT-1 and OPT-2 are alternative scenarios based on Global Orchestration (GO) of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Mayorga et al. 2010; Seitzinger et al. 2010). CP is the scenario that incorporates current policy plans for agriculture. OPT-1 and OPT-2 are optimistic scenarios that assume full implementation of improved nutrient management in agriculture (OPT-1 and OPT-2) and better sewage control (OPT-2). See Tables 6.1, S6.1 and S6.2 for scenario description and assumptions. Figures 6.4, 6.5, S6.13 and S6.14 provide source attribution for nutrient exports.*

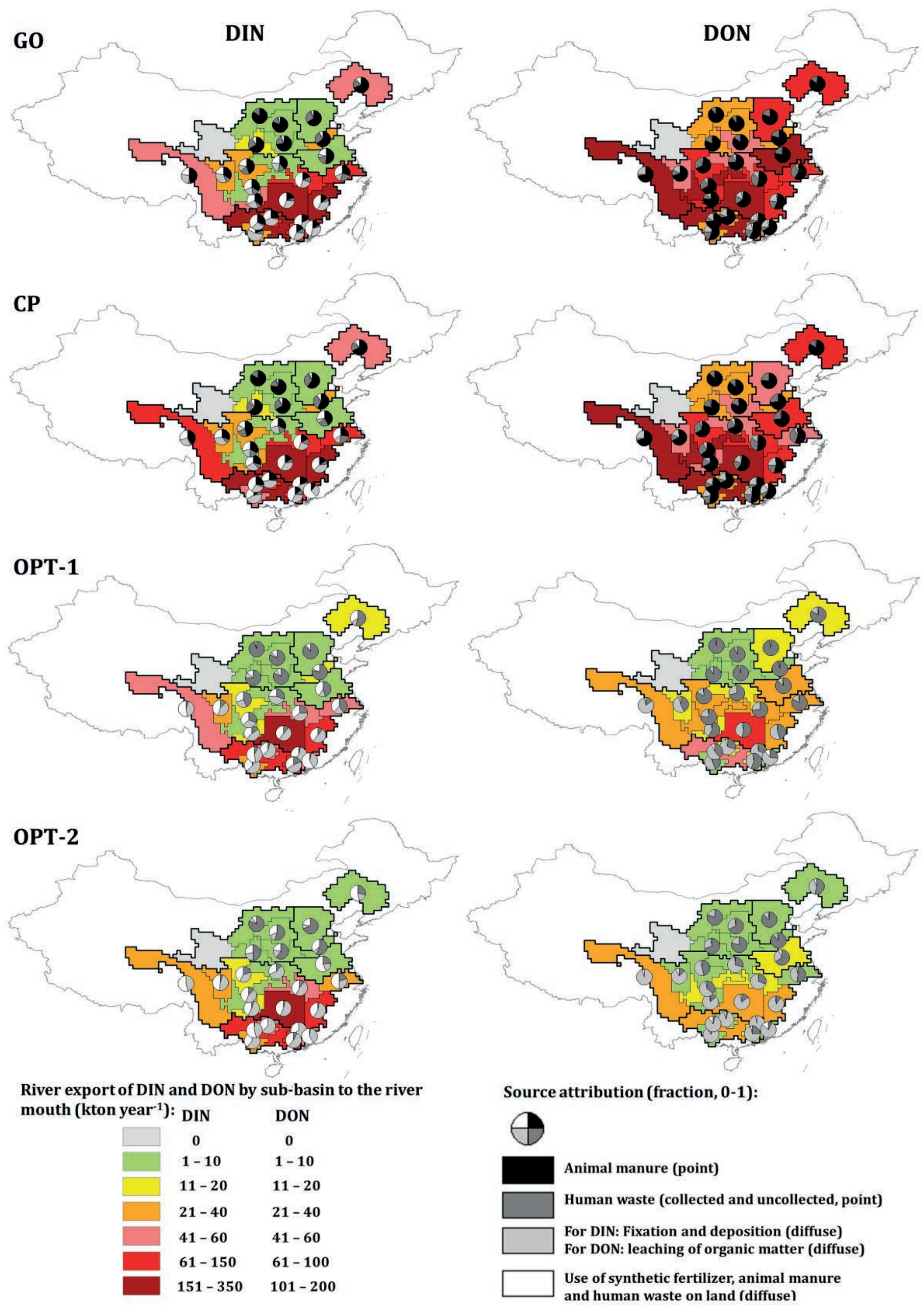


Figure 6.4. *River export of dissolved inorganic (DIN) and organic (DON) nitrogen (N) by sub-basin to the river mouth and their source attribution in 2050 according to the GO, CP, OPT-1 and OPT-2 scenarios. CP, OPT-1 and OPT-2 are alternative scenarios based on Global Orchestration (GO) of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Mayorga et al. 2010; Seitzinger et al. 2010). CP is the scenario that incorporates current policy plans for agriculture. OPT-1 and OPT-2 are optimistic scenarios that assume full implementation of improved nutrient management in agriculture (OPT-1 and OPT-2) and better sewage control (OPT-2). See Tables 6.1, S6.1 and S6.2 for scenario description.*

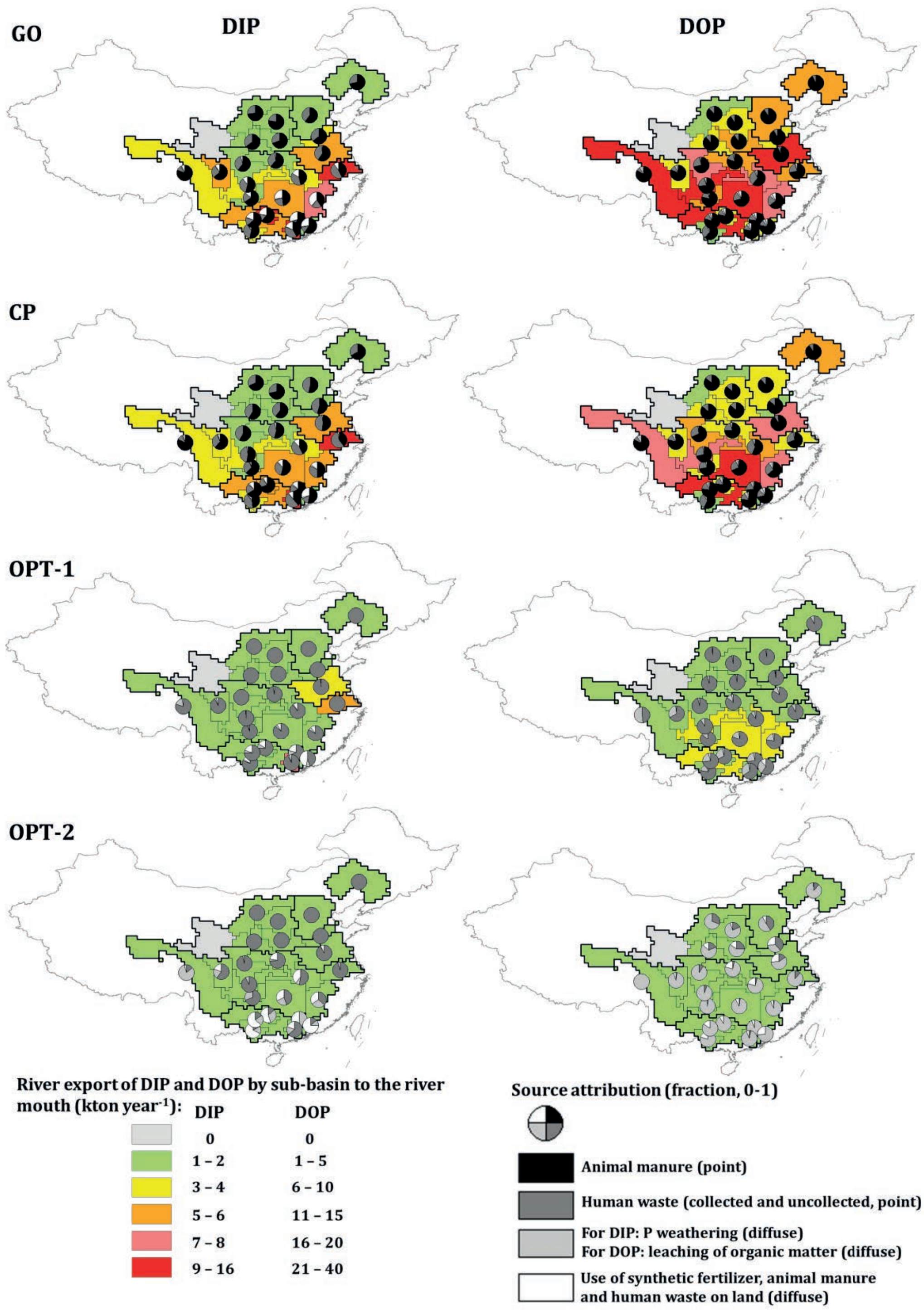


Figure 6.5. *As Figure 6.4, but for river export of dissolved inorganic (DIP) and organic (DOP) phosphorus (P) by sub-basin to the river mouth and their source attribution in 2050 according to the GO, CP and OPT scenarios.*

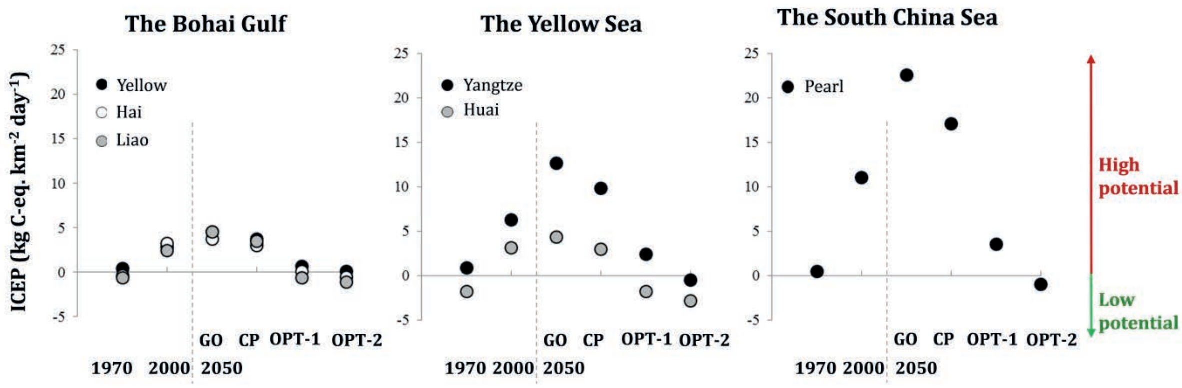


Figure 6.6. *Indicator for Coastal Eutrophication Potential (ICEP, kg C-eq. km^{-2} day^{-1}) for the Chinese rivers in 1970, 2000 and 2050 according to the GO, CP and OPT scenarios. High potentials for coastal eutrophication are indicated by positive ICEP, indicating that river transport of nutrients is in excess over that of dissolved silica. Low potentials are indicated by negative ICEP. Either N- or P-ICEP is calculated depending on which nutrient (N or P) is limiting (see Figure S6.16). CP, OPT-1 and OPT-2 are alternative scenarios based on Global Orchestration (GO) of the Millennium Ecosystem Assessment (Alcamo et al. 2005; Mayorga et al. 2010; Seitzinger et al. 2010). CP is the scenario that incorporates current policy plans for agriculture. OPT-1 and OPT-2 are optimistic scenarios that assume full implementation of improved nutrient management in agriculture (OPT-1 and OPT-2) and better sewage control (OPT-2). See Tables 6.1, S6.1 and S6.2 for scenario description and assumptions.*

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Chapter 7.
Overall discussion and conclusions

7.1 Introduction

The main research objective of this PhD thesis is to better understand trends in river export of nutrients to the coastal waters of China by source from sub-basins, and the associated coastal eutrophication. To achieve this objective I developed the *MARINA* model (Model to Assess River Inputs of Nutrients to seAs).

I justified the research objective in Chapter 1. In Chapter 2, I analyzed coastal eutrophication in China that is associated with river export of nitrogen (N), phosphorus (P) and silica (Si) for 1970-2050. For this I used the original, basin scale Global *NEWS*-2 model (Nutrient Export from WaterSheds). In Chapter 3, I developed a sub-basin scale modeling approach for the Pearl River (the third largest river in China). I implemented it to Global *NEWS*-2 with updated reservoir information. This was done to better understand the spatial variability in nutrient sources within large, data-poor basins such as the Pearl. In Chapter 4, I focused on a missing source of nutrients in Chinese rivers: direct manure discharges to rivers, referred to as manure point sources. In Chapter 5, I presented the new *MARINA* model for China. In the *MARINA* model I implemented the improved sub-basin approach (from Chapter 3) to other large Chinese rivers and to other nutrient forms. Moreover, the *MARINA* model includes manure point sources (from Chapter 4) as well as uncollected human waste as a new source of nutrients in rivers. In Chapter 6, I explored options to reduce river export of nutrients and thus coastal eutrophication in China in 2050.

In this final Chapter I discuss the methodological aspects of nutrient models (Section 7.2). Next, I discuss the results of the *MARINA* model by comparing them to other studies for China and to other world regions (Section 7.3). Finally, I provide the main lessons for nutrient modeling, summarize findings of the PhD thesis (Section 7.4) and discuss future outlook (Section 7.5).

7.2 Modeling nutrient flows to aquatic systems in China

In this section, I focus on modeling approaches for China and the niche of the new *MARINA* model. I start with a brief overview of a number of the existing nutrient models for China (Section 7.2.1). Then, I describe the *MARINA* model (Section 7.2.2), its strengths (Section 7.2.3) and weaknesses (Section 7.2.4) in comparison to other nutrient models. Finally, I reflect on how to build scientific trust in nutrient models (Section 7.2.5).

7.2.1 Nutrient models for China

Many models exist to quantify nutrient flows to aquatic systems. In an earlier study (Strokal & de Vries 2012) I described 27 water quality models performing at different scales: ranging from the field, to the basin and to the global scale. In this section, I do not intend to give a complete model overview. Rather, I intend to compare illustrative examples of models for China. To this end, I selected models that cover the whole of China, or that are widely applied to individual basins of China (Tables 7.1 and 7.2). These are Global *NEWS*-2, IMAGE-GNM (Integrated Model to Assess the Global Environment–Global Nutrient Model), NUFER (Nutrient flows in Food chains, Environment and Resources use), SWAT (Soil and Water Assessment Tool) and AGNPS (AGricultural Non-Point Source pollution model or Agricultural NonPoint Source). Other models such as SPARROW (SPAtially Referenced Regressions On Watersheds, Schwarz et al. 2006), RVERSTRAHLER (Sferratore et al. 2005), and GREEN (Geographic Regression Equation for European Nutrient losses, Grizzetti et al. 2012) are also interesting. However these models are, so far, mainly applied to basins of the United States and Europe and thus are not included in this comparison (Tables 7.1-7.3, see also Chapter 1 for model descriptions).

o Models for China as a whole

Global *NEWS*-2 (Mayorga et al. 2010) quantifies river export of nutrients by source (Tables 7.1 and 7.2). This is done for over 6000 rivers as a function of human activities on land and basin characteristics. Human activities are, for example, agriculture and sewage. Basin characteristics include land use, hydrology etcetera. Nutrient retentions in soils and rivers are taken into account under a steady state approach. Important factors influencing nutrient retentions in and removal from the river network are denitrification, water withdrawal and reservoirs. The temporal level of detail is annual. The spatial level of detail is the grid scale of 0.5 by 0.5 degree cell for most of the model inputs and a basin scale for model outputs. Model outputs are quantified river export of N, P, Si and carbon (C) in dissolved inorganic, dissolved organic and particulate forms. The other outputs are the Indicator for Coastal Eutrophication Potential (ICEP) and the source attribution. In the source attribution, diffuse and point sources of nutrients in rivers are accounted for. Diffuse sources include the use of synthetic fertilizers and manure on cropland, atmospheric N deposition, biological N fixation, P weathering and organic matter leaching. Point sources include sewage effluents from human waste and detergents. All model outputs are for the past (1970 and 2000) and future (2030 and 2050). Future trends are based on the scenarios of the Millennium Ecosystem Assessment (MEA) (Alcamo et al. 2005; Seitzinger et al. 2010). The model has been used for global analyses of nutrient export by rivers (Maavara et al. 2015; Mayorga et al. 2010; Seitzinger et al. 2010; Van Cappellen & Maavara 2016). Many studies applied the 2010 version of the Global *NEWS*-2 model to analyze water pollution in Indonesia (Suwarno et al. 2013; Suwarno et al. 2014a; Suwarno et al. 2014b), the Bay of Bengal (Sattar et al. 2014; Zinia & Kroeze 2015), Europe (Thieu et al. 2010), Africa (Yasin et al. 2010), North America (McCrackin et al. 2013; McCrackin et al. 2014), the Black Sea (Strokal & Kroeze 2013; Strokal et al. 2014c) and in China (Li et al. 2011b; Qu & Kroeze 2010; Qu & Kroeze 2012; Yan et al. 2010). I discuss these studies in Section 7.3.2.

IMAGE-GNM is another global model for river export of nutrients, but performs at the grid scale of 0.5 by 0.5 degree cell (Tables 7.1 and 7.2). It takes dynamic and spiraling approaches for nutrient cycling, meaning that soils and sediments have a “memory”. This affects the release of N and P to water systems via surface and sub-surface runoff. The spiraling approach for in-stream nutrient retentions accounts for denitrification, sedimentation and nutrient uptake by aquatic plants. Reservoirs and lakes serve as sinks and thus influence the transport of nutrients to sea. The temporal level of detail is annual, as in Global *NEWS*-2. The model outputs are total N and P fluxes exported from land to sea by grid. The other output is the source attribution. Here the diffuse and point sources of nutrients are considered as in Global *NEWS*-2. However, IMAGE-GNM includes additional sources such as allochthonous biomass inputs to rivers, aquaculture, direct N deposition to water systems and industrial wastewaters. So far, IMAGE-GNM has been applied for global analyses to understand cycles of N and P over the 20th century. Application of the model to individual-specific regions is rare (see Table 1 for references and Table 7.2 for a description).

NUFER differs from the global river export models in that it quantifies nutrient use efficiencies in the food chain of China and the associated losses to the environment (Tables 7.1 and 7.2). This information is annual and available for total N and P by country and by province for 1980, 2005 (Ma et al. 2012), 2013 and 2030 (Ma et al. 2013a; Wang et al. under review). Wang et al. (in preparation) have recently developed a county version of the model. NUFER is largely based on local statistical databases for model inputs (ECCAP 2006; MOA 2006) and Chinese field surveys (NATESC 1999). In

NUFER, inputs of nutrients to surface waters are from crop and animal production systems as well as from household and food processing activities. This model accounts for direct manure discharges to nearby water systems. These manure point sources are not accounted for in the global river export models. Human waste from rural people is another nutrient source in NUFER, but it is underrepresented in river models such as Global *NEWS*-2 (see Tables 7.1 and 7.2 for references and model descriptions).

- Models that are mainly applied at the level of individual basins in China

SWAT and AGNPS are typical examples of the models (e.g., Ongley et al. 2010; Shen et al. 2012) that aim at analyzing impacts of agricultural activities on water quality (Tables 7.1 and 7.2). These are, in general, distributed, physically-based models with well represented water flows and dynamics in nutrient processes. Both models can take a different temporal and spatial level of detail to quantify nutrient fluxes. SWAT can perform at hourly, daily, monthly and annual basis. AGNPS focuses on simulating nutrient flows during events such as storms. SWAT performs at hydrological response unities (HRUs) that are then aggregated to sub-basins or basins. AGNPS divides a basin into grids of, for example, 200m by 200m. Thus, input data for these models are needed at a much higher spatial and temporal resolution than for basin (Global *NEWS*-2) and province (NUFER) models. Outputs of SWAT for nutrient flows to aquatic systems are mainly for total N and P. Outputs of AGNPS are for dissolved and total N and P flows. However, these models do not explicitly provide the source attribution of nutrient loadings to surface waters as river export models do (Global *NEWS*-2, IMAGE-GNM). SWAT and AGNPS can be used for future predictions depending on the research objective (see Tables 7.1 and 7.2 for references and model descriptions).

***Table 7.1**. Abbreviations and main references of the selected nutrient models for China, compared to the MARINA model.*

Model	Abbreviation	Main references
Illustrative examples of nutrient models		
Global *NEWS*-2	Nutrient Export from WaterSheds	Mayorga et al. (2010), Seitzinger et al. (2010)
IMAGE-GNM	Integrated Model to Assess the Global Environment–Global Nutrient Model	Beusen et al. (2015a) Beusen et al. (2015b)
NUFER*	Nutrient flows in Food chains, Environment and Resources use	Ma et al. (2012) Ma et al. (2010)
SWAT	Soil and Water Assessment Tool	Arnold et al. (2012) Gassman et al. (2014)
AGNPS as an example**	AGricultural Non-Point Source pollution model or AGricultural NonPoint Source	Shen et al. (2012) Jianchang et al. (2008)
This study		
MARINA	Model to Assess River Inputs of Nutrients to seAs	Strokal et al. (2016a) Strokal et al. (2016b) Strokal et al. (2015)

* Except for NUFER, a few other models exist to quantify nutrient balances for agricultural systems in all of China (e.g., Chen et al. 2016; Chen et al. 2008a). NUFER was chosen here for the comparison because it quantifies nutrient flows in the food chain that consists of not only agricultural systems (crop and animal), but also food processing and household systems. And, NUFER has been widely applied for the past, present and future analyses of nutrient flows in the food chain at the national and provincial scales (see details in Section 7.2.1). ** A typical representative of the group of models that are widely used in China for individual basins to assess effects of diffuse pollution sources on water quality (e.g., Shen et al. 2012).

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Table 7.2. *Summarized overview of selected nutrient models for China, compared to the MARINA model. See Table 7.1 for model abbreviations and references.*

Model characteristics	Global *NEWS*-2	IMAGE-GNM	NUFER	SWAT	AGNPS	*MARINA*
Purpose	Analyze impacts of human activities on coastal waters	Understand changes in nutrient cycles over the 20th century	Analyze nutrient cycling in the food chain	Analyze effects of management practices on water resources	Analyze impacts of diffuse pollution sources on water quality	Analyze impacts of human activities from sub-basins on coastal waters
Model type	Lumped, quasi-empirical model	Distributed, physically-based model	Lumped model with combined approaches	Distributed, physically-based model	Distributed, physically-based model	Lumped, quasi-empirical model
Main model inputs	o Human activities o Economy & demography o Basin characteristics	o Human activities o Economy & demography o Basin characteristics	o Food chain activities o Demography	o Digital elevation map o Soil type and land use o Weather data	o Land use, slope o Soil characteristics o Hydrology	o Human activities o Economy & demography o Sub-basin characteristics
Main model outputs	o River export of nutrients o Source attribution o Indicator for coastal eutrophication (ICEP)	o River export of nutrients o Source attribution	o Nutrient use efficiencies o Nutrient losses to the environment	o Water quality that can be in terms of nutrient loadings to rivers and their outlets	o Nutrient losses from agricultural areas	o River export of nutrients o Source attribution o Indicator for coastal eutrophication (ICEP) o Sub-basin contribution
Nutrient forms*	DIN, DIP, DON, DOP, PN, PP, DSi, DOC, POC	TN, TP	TN, TP	TN, TP**	TN, TP, dissolved N and P	DIN, DIP, DON, DOP
Spatial level of detail: o Model inputs o Model outputs	 o $0.5^{o} \times 0.5^{o}$ grid o Basin	 o $0.5^{o} \times 0.5^{o}$ grid o $0.5^{o} \times 0.5^{o}$ grid	 o Country, province o Country, province	 o Flexible*** o Basin (watershed)	 o Grid (e.g., 200m × 200m) o Basin (watershed)	 o $0.5^{o} \times 0.5^{o}$ grid o Sub-basin
Temporal level of detail	Annual	Annual	Annual	Hour, day, month, annual	Event-based****	Annual
Spatial extent	Global, regional	Global	China	Individual basins	Individual basins	China
Temporal extent	1970-2050	1900-2000	1980-2030	Flexible*****	Uncertain*****	1970-2050
Modeling approach for nutrient cycling	Steady state	Dynamic and account for spiraling	Steady state	Dynamic	Dynamic	Steady state
Diffuse sources of nutrients in rivers that are included in the model	o Fertilizers (synthetic & manure) o N deposition & fixation o P weathering o Organic matter leaching	o Losses via surface & sub-surface runoff from agricultural & natural areas o Allochthonous biomass inputs[(a)] o P weathering	o Losses from agricultural areas with applications of fertilizers (synthetic, manure, human excreta), straw residues & seeds	o Losses from areas having different management practices (e.g., with crop production)	o Losses from areas with different management practices	o Fertilizers (synthetic & manure) o N deposition & fixation o P weathering o Organic matter leaching o Uncollected human waste
Point sources of nutrients in rivers that are included in the model	o Sewage effluents	o Sewage effluents[(b)] o Aquaculture[(c)] o Direct N deposition to water systems	o Direct inputs of: animal manure & uncollected human waste	o Sewage effluents (optional)	o Sewage effluents (optional)	o Sewage effluents o Direct inputs of manure o Direct inputs of uncollected human waste

* DIN = dissolved organic nitrogen. DIP = dissolved inorganic phosphorus. DON = dissolved organic nitrogen. DOP = dissolved organic phosphorus. DSi = dissolved silica. DOC = dissolved organic carbon. POC = particulate organic carbon. TN = total nitrogen. TP = total phosphorus. ** Nitrate and phosphate are modeled. But the outputs at river mouths are mainly given in forms of TN and TP. *** Depends on the study purpose and available maps for model inputs. **** The focus is on simulating nutrient fluxes during events such as storms, see Jianchang et al. (2008). *****Future predictions are possible, see Shen et al. (2012). **(a)** These are inputs from wetlands and floodplains. For example, during flooding some litter can enter surface waters and be a source of nutrients. **(b)** Includes industrial wastewater. **(c)** Aquaculture includes finfish and shellfish.

Table 7.3. *As Table 7.2, but for main strengths and weaknesses of selected models for modeling nutrient export from land to sea in China.*

Model characteristics	Global *NEWS*-2	IMAGE-GNM	NUFER	SWAT	AGNPS	*MARINA*
Strengths	o Past and future trends o Different nutrient forms: dissolved inorganic, dissolved organic and particulate o Many diffuse and point sources o Indicator for coastal eutrophication o Widely applied o Transparent	o Grid scale for local analyses o Dynamic & spiraling approaches for in-soil and in-stream retentions of nutrients o Many diffuse and point sources including aquaculture and direct N deposition to water systems	o Developed for China o Based on Chinese statistical data and surveys o Diffuse and point sources from agriculture including direct manure inputs to rivers and human waste	o Dynamics in nutrient cycling o Flexibility for inter-annual analyses o Includes other pollutants like bacteria and pesticides o Flexibility to add nutrient sources such as sewage effluents	o The impact of erosion o Event-based, which is better for addressing seasonal problems o Includes chemical oxygen demand & pesticides o Flexibility to add nutrient sources such as sewage effluents	o Comprehensiveness in terms of: - Nutrient export by river - Multiple nutrient forms - Many diffuse and point sources - Sub-basin contribution - Indicator for coastal eutrophication - Past and future trends o Sub-basin scale for large rivers to identify polluting areas with human activities o Improved modeling of pollution by including missing sources: direct inputs of manure to rivers and uncollected human waste o Coupling with NUFER o Updated reservoir information o Transparency
Weaknesses	o Basin scale for local analyses o Annual values o Missing sources: - direct inputs of manure to rivers - uncollected human waste - aquaculture - direct N deposition on seas o No P retentions in rivers due to sedimentation o Steady state approach for nutrient cycling o Outdated information for dams	o High data demand at the grid scale o Proper validation is challenging o Annual values o Missing sources: - direct manure inputs to rivers - direct N deposition on sea o No water consumption o No indicator for coastal eutrophication o No nutrient forms o No future trends yet	o Focus on agriculture o No river export of nutrients o Annual values o Steady state approach for nutrient cycling o No coastal eutrophication o No nutrient forms	o High data demand o High computation time o Validation / calibration needs a lot of water quality data o No explicit source attribution for model outputs o No coastal eutrophication o No nutrient forms o Focus mainly on agricultural areas	o Similar to SWAT	o Spatial and temporal level of detail for local or inter-annual water pollution problems o Missing local sources* o Missing reservoir retentions for DON and DOP** o Steady state approach for nutrient cycling o Calibration

*Aquaculture, industry & direct N deposition on sea. **DON = dissolved inorganic nitrogen. DOP = dissolved organic phosphorus.

7.2.2 *MARINA* model description

The *MARINA* model quantifies past and future trends in river export of nutrients by source from sub-basins and the potential for coastal eutrophication (Figure 7.1, Tables 7.1-7.3). This model builds largely on approaches and gridded input datasets of Global *NEWS*-2, but it accounts for missing sources of nutrients in Chinese rivers at the sub-basin scale (Strokal et al. 2016a). Global *NEWS*-2 was chosen as a basis for three reasons (see Section 7.2.1). First, Global *NEWS*-2 quantifies past and future trends in river export of nutrients, source attribution and the associated coastal eutrophication. Second, Global *NEWS*-2 accounts for nutrients in different forms: dissolved inorganic, dissolved organic and particulate. Third, Global *NEWS*-2 has been broadly accepted as illustrated by its wide applications in many studies for water pollution problems, including China (see Section 7.3.2).

In the *MARINA* model, river export of nutrients is quantified for dissolved inorganic and organic N and P for 1970, 2000 and 2050 (Tables 7.1-7.3). The model is applied to six main rivers of China: Yellow, Hai, Liao, Yangtze, Huai and Pearl. These rivers drain into the Bohai Gulf, Yellow Sea and South China Sea. The Yangtze, Yellow and Pearl rivers have the largest drainage areas of 1.8, 0.89 and 0.44 million km^2 respectively. The drainage areas of these rivers (covering around one-third of China with intensive economic activities) are subdivided into 10, 6 and 6 sub-basins, respectively (Figure 5.2). The other three smaller rivers have drainage areas of between 0.24 and 0.27 million km^2 and are considered as individual sub-basins in the model. The sub-basins are classified as up-, middle- and downstream based on literature (Cui et al. 2007; Huang et al. 2009a; Niu & Chen 2010; Wang et al. 2010; Yang & Lu 2014a; Zhou et al. 2013). I used the Simulated Topological Network (STN-30) of Global *NEWS*-2 (Vörösmarty et al. 2000a; Vörösmarty et al. 2000b) to delineate the sub-basins. This STN-30 is at the 0.5 by 0.5 degree cell resolution and based on the Strahler order (Strahler 1957). In this study, I delineated sub-basins with tributaries and with the main channel in up- middle- and downstream areas. Tributaries typically have a Strahler order of less than three. The main channel often has a Strahler order of more than three.

River export of nutrients is quantified as a function of human activities and sub-basin characteristics (Figure 7.1, Table 7.2). The principle is that human activities add nutrients to tributaries. These tributaries export nutrients to the main channel of the river. Through the main channel nutrients are transported to the river mouth (coastal waters in this study). In general, nutrients from upstream activities travel a longer distance to the river mouth than nutrients from downstream activities. During this travel, nutrients can be lost or retained in soils and water systems. For example, bioavailable N can be denitrified by anaerobic bacteria and lost to the air as gaseous N.

Water consumption can remove nutrients from the river whereas reservoirs can retain nutrients in the river. In *MARINA*, these nutrient losses and retentions are modeled as in Global *NEWS*-2, but with updated information for reservoirs from the global database (GRanD) (Lehner et al. 2011b, Section 7.2.1). In addition, I added DIP accumulation in sediments (Chapter 5). This model parameter varies between drier (Hai, Huai and Yellow) and wetter (Yangtze and Pearl) rivers (Strokal et al. 2016a).

Human activities result in point and diffuse sources of dissolved N and P in rivers. Most of these sources are the same as in Global *NEWS*-2 (see Section 7.2.1). Thus, I used the approaches of Global *NEWS*-2 to quantify most of the nutrient sources (Mayorga et al. 2010). In addition, the *MARINA* model accounts for direct inputs of animal manure to rivers and for uncollected human waste. Direct inputs of manure were quantified using provincial information of the NUFER model (Ma et al. 2012). Uncollected human waste is modeled according to the approaches and information of Morée et al. (2013) for urban people and of NUFER (Ma et al. 2012) for rural people (see Chapter 5).

The model parameters and inputs were taken from different sources, including Global *NEWS*-2 (Mayorga et al. 2010; Seitzinger et al. 2010). For example, Bouwman et al. (2009) and Van Drecht et al. (2009) prepared gridded model inputs of 0.5 by 0.5 degree cell using the IMAGE (Integrated Model to Assess the Global Environment) model. These inputs are for diffuse and point sources of nutrients such as agriculture, land use, population and economic activity. Fekete et al. (2010) provided gridded inputs of 0.5 by 0.5 degree cell for hydrology and climate using the WBM (Water Balance Model plus) model. All these gridded inputs are available for 1970, 2000 and 2050 for the *MARINA* model. For 2050 the inputs are based on the MEA storylines on socio-economic development, livestock and crop production (Alcamo et al. 2005; Seitzinger et al. 2010). In this study, the gridded inputs were aggregated to sub-basins. Some information was also used from other sources that are mentioned in paragraphs above. Literature was reviewed to justify some of the model parameters in combination with expert knowledge (see Section 7.2.5).

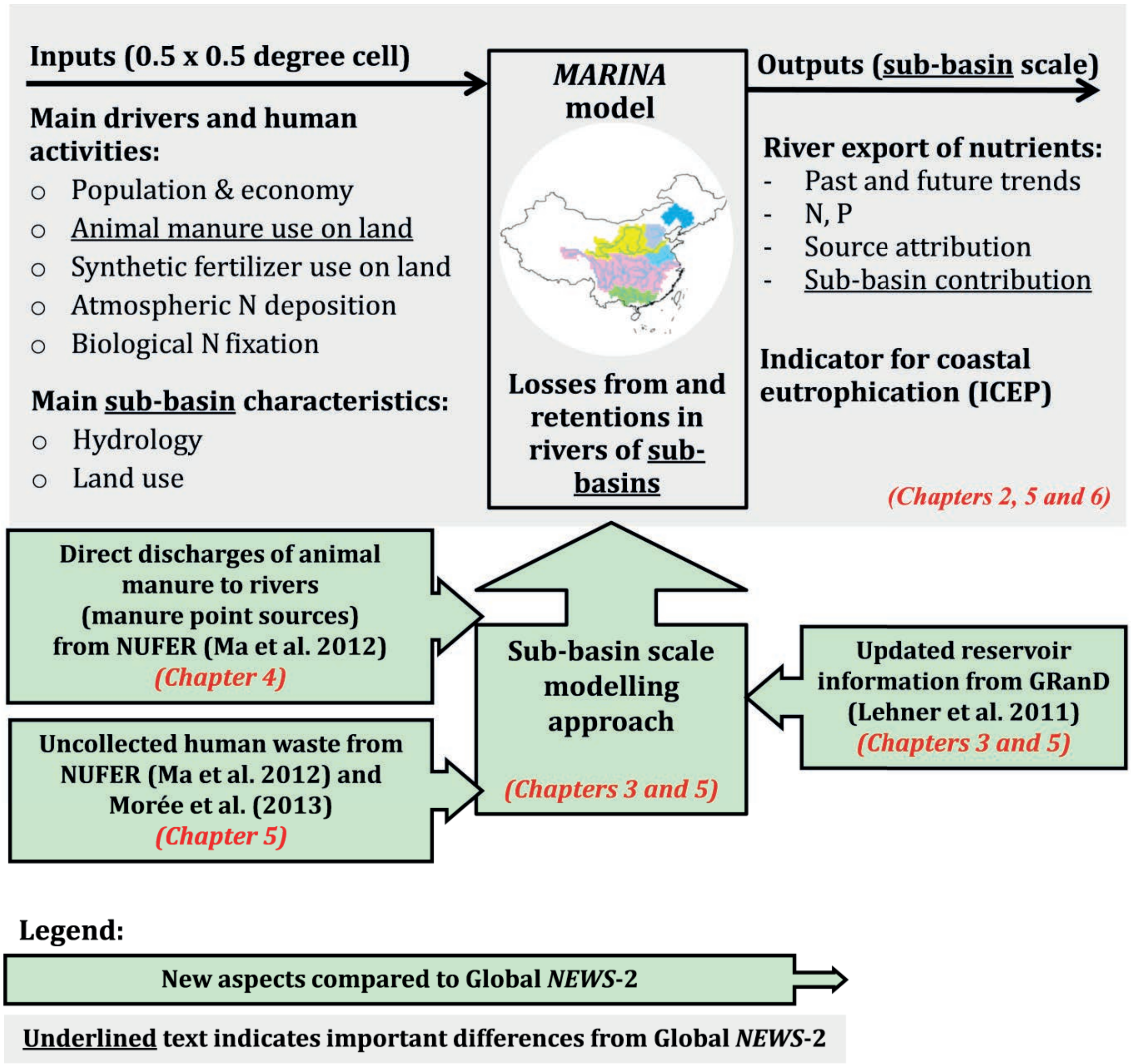

Figure 7.1. *Summary of how the MARINA model (Strokal et al. 2016a) differs from the Global NEWS-2 model (Mayorga et al. 2010; Seitzinger et al. 2010). Chapter 2 provides details on Global NEWS-2 and Chapter 5 on the MARINA model. Table 7.1 and Table 7.2 summarize descriptions of MARINA and Global NEWS-2 as well as the other selected nutrient models for China.*

7.2.3 Strengths of the *MARINA* approach

o Comprehensiveness

MARINA is the first comprehensive model for China (Table 7.3). Its comprehensiveness is reflected by various outputs to assess water pollution in China and to explore solutions. *MARINA* is a river export model for multiple nutrient forms and for many sources of these nutrients in Chinese rivers. The model provides river export of nutrients by sub-basin and the indicator for coastal eutrophication (ICEP). It does all of this for past and future years (Figure 7.1, Table 7.3). The combination of all these outputs makes *MARINA* unique among nutrient models for China (Tables 7.1-7.3). For example, *MARINA* is available for dissolved inorganic and dissolved organic N and P, whereas most of the selected nutrient models are generally limited to TN and TP (e.g., IMAGE-GNM and NUFER). *MARINA* covers the period of 1970-2050 while IMAGE-GNM currently covers the period of the 20th century. *MARINA* uses ICEP as Global *NEWS*-2 whereas the other selected models do not (Tables 7.1-7.3).

- Sub-basin scale

I consider the sub-basin approach as the most novel aspect of the *MARINA* model. This is because this approach allows to analyze the relative share of sub-basins in the total river export of nutrients at the river mouth. To my knowledge, no other models today exist that are able to do this. Moreover, the *MARINA* model does this by source (the source attribution) meaning it can be used to quantify the relative share of individual pollution sources in the impact on the coastal seas. This makes it a powerful tool to support effective nutrient management. This is especially helpful for large basins that are often poorly documented, but which can contribute largely to coastal water pollution by river export of nutrients. I demonstrated this in Chapters 4 and 5. The few existing studies with sub-basin analyses of nutrient fluxes in China are not comprehensive as *MARINA*. There are a few studies of single rivers that analyzed nutrient fluxes, but mainly for areas dominated by agriculture and for only TN and TP (e.g., Bao et al. 2006; Liu et al. 2008a). A few other studies addressed effects of the Three Gorges Dam on DIN and TP export by the Yangtze from up- to downstream areas (Sun et al. 2013a; Sun et al. 2013b), and effects of water consumption on TN and TP fluxes by the Yellow (Tong et al. 2016). However, none of these studies account for source attribution of sub-basins as detailed and complete as *MARINA* does.

Existing models for river export of nutrients are implemented typically at the basin or grid scales (Tables 7.2 and 7.3). However, whether these scales are appropriate depends on the study area and the objective of the study. Clearly, basin scale models like Global *NEWS*-2 are too coarse to account for the spatial variability in human activities within the basin. Gridded scale modeling has an advantage here because it accounts for this variability. Another advantage is the opportunity to analyze local pollution problems. IMAGE-GNM, SWAT and AGNPS are examples of such gridded models. These models take grids in different sizes (Tables 7.1-7.3). However, the demand for gridded model inputs and parameters is often high; and the accuracy of gridded model results is often difficult to check. This is especially the case for large basins because water quality data are often scarce. Another constraint of the gridded models is increasing computation time. I believe that the sub-basin *MARINA* approach has advantages here. It takes only a few seconds to run, does not require extensive datasets, and provides the needed information to comply with research objectives as formulated in this thesis. In addition, the performance of the *MARINA* model was evaluated. Results of this evaluation provided enough confidence in using the model for sub-basin analyses of nutrient fluxes in China (see Section 7.2.5 on the model evaluation).

o Improved modeling of pollution by manure and human waste

MARINA is the first river export model that includes direct discharges of animal manure to rivers and uncollected human waste at the sub-basin scale simultaneously (Table 7.3). *MARINA* accounts for many other diffuse and point sources of nutrients in Chinese rivers. Table 7.2 lists these sources. Other river export models like IMAGE-GNM and Global *NEWS*-2 also account for many of these sources, but ignore direct inputs of manure to rivers which are the largest source of nutrients in many Chinese rivers (Table 7.2). Direct inputs of manure are modeled by NUFER for all of China, but this is not a river export model and cannot address impacts of this source on coastal water quality. The same holds for uncollected human waste in NUFER. IMAGE-GNM accounts for the impact of urban human waste on river export of nutrients, but not at the sub-basin scale and not for the different nutrient forms and future years yet.

Direct discharges of manure to rivers seem to be unique for China (see Section 7.3.2 for the comparison with other studies). These direct discharges of manure to rivers have happened because animal production has been industrializing since the 1990s with rather poor manure management. Before the 1990s, traditional farms were dominant where most animal manure was recycled on land. Today, industrial farms dominate meat production. This holds especially true for pork and poultry production. For example, in 2010, 40-80% of the pigs and poultry in China were grown in industrial farms. Considerable amounts of manure in industrial farms are directly lost to nearby water systems. Some manure can be collected and composted, but it is hardly applied on land (Chadwick et al. 2015). One of the reasons is the location of industrial farms. Industrial farms are often constructed in semi-urban areas to meet high food demand. However, these semi-urban areas are often far from cropland. Consequently, this increases the cost for manure transportation and the demand for labor. Another reason is outdated techniques for manure application (Bai et al. 2015; Chadwick et al. 2015; Ma et al. 2013b). Thus, arable farmers prefer synthetic fertilizers because of their lower prices and lower labor demand (e.g., Ju et al. 2005; Li et al. 2013). However, limited knowledge on when and how much to apply, for both manure and synthetic fertilizers, has led to inefficient use of nutrients in crop production (e.g., Ju et al. 2009).

Another source of nutrients in rivers is uncollected human waste, an input that is ignored in Global *NEWS*-2, but important for China. Particularly in rural areas, people lack sewage connections (WHO/UNICEF 2014). Therefore, uncollected human waste may enter rivers via leaching or runoff from soils (diffuse sources) or be directly discharged as untreated influents (point sources). In Chapter 5, I showed the importance of uncollected human waste in water pollution especially for the past when most of the Chinese population was without sewage connections.

- Coupling with NUFER

Another important feature of *MARINA* is that it is coupled with NUFER (Table 7.3). I did this by using information from NUFER to improve modeling of animal manure and of human waste from the rural population (Section 7.2.2). Although NUFER is a food chain model for China, it has comprehensive information on nutrient management in agriculture at the scale ranging from the county (Wang et al. in preparation), to the province (Ma et al. 2012), and to the country (Ma et al. 2010). In addition, this information relies largely on local sources (see Section 7.2.1). Thus, for nutrient management aspects in Chinese agriculture NUFER has advantages over, for example, global river export models that are often based on national assessment reports like the FAO to generate gridded information for model inputs (e.g., Bouwman et al. 2009).

- Updated reservoir information

Another strong point of *MARINA* is the updated information for reservoirs (Section 7.2.2, Table 7.3). This is an update of Global *NEWS*-2 (Fekete et al. 2010; Vörösmarty et al. 2003). *MARINA* uses the recent GRanD database (Lehner et al. 2011b). This database is more complete than the information in Global *NEWS*-2. For example, GRanD provides information for around hundred dams in China and their associated reservoirs having water discharges larger than or equal to 0.5 km^3. The number for dammed reservoirs is 80 in Global *NEWS*-2 (Fekete et al. 2010; Vörösmarty et al. 2003).

- Transparency

Finally, I consider the *MARINA* model transparent (Table 7.3). This makes the model easy to understand by other users. Thus, the model has the potential to be applied to other large river basins in the world to analyze causes of water pollution and solutions (see Section 7.5 on future outlook).

7.2.4 Weaknesses of the *MARINA* approach

- The spatial and temporal level of detail for local or inter-annual water pollution problems

In Section 7.2.3, I showed how the sub-basin scale is useful to study large basins. On the other hand, this scale seems too coarse for local water pollution problems. For example, cities like Beijing, Shanghai and Shenzhen are densely populated with a lot of human waste, creating local pollution. These cities are not geographically specified in the model, but they are part of the downstream sub-basins (see Figures 5.1 and 5.2 for locations of the sub-basins). Thus, the current version of *MARINA* cannot provide city-oriented information. Models such as SWAT and AGNPS are, perhaps, better tools for in-depth analyses of such local areas, but these models are more developed for rural areas (Tables 7.1-7.3). IMAGE-GNM can also be a good tool because it covers both rural and urban areas at the grid scale. Nevertheless, I believe that *MARINA* can also be useful here: (i) it accounts for urban and rural population and (ii) sub-basin information from *MARINA* can serve as a first step in identifying areas that contribute largely to coastal water pollution. This information can also indicate where monitoring of water quality is more needed. Results of such monitoring can be used to reduce uncertainties in the model.

The *MARINA* model uses the ICEP approach to quantify the potential for coastal eutrophication (Garnier et al. 2010). However, this indicator is based on annual totals of riverine nutrient fluxes (Tables 7.1-7.3). Annual values are useful to analyze trends in nutrient fluxes, but may be less sufficient to analyze eutrophication problems such as blooms of harmful algae. This is because algal blooms often last for certain periods within a year: days, months or season (Davis et al. 2009; Tang et al. 2006a). This depend on various factors such as light, oxygen, nutrients and the presence of algal consumers in aquatic systems (Conley et al. 2009; Paerl et al. 2001). N and P are among the controlling factors for development of algal blooms, which are often undesirable (Conley et al. 2009). An example is the shift from siliceous diatoms to non-siliceous, often harmful algae like N_2-fixing cyanobacteria. This happens when aquatic systems have too much N or P in relation to Si (N:P:Si ratio). This limits the growth of diatoms and gives a chance for the development of non-diatom algae (Garnier et al. 2010). The Redfield ratio of N:P:Si (16:1:20) is often used as an indication of the diatom requirements for growth. This Redfield ratio is the basis of the ICEP indicator (Garnier et al. 2010). This is an advantage of ICEP.

Furthermore, ICEP can be a useful indicator, but we must interpret it with care. ICEP values above zero should be interpreted as an indication that the potential for harmful algae is high. Negative ICEP indicates a rather low potential for algal blooms. However, it does not mean that events of harmful algae do not occur even though the annual total river export of nutrients is low compared to Si levels. An important advantage of ICEP is its transparency. Thus, it is easy to use this indicator. In fact, ICEP has been applied in various studies to analyze coastal eutrophication problems (e.g., Dupas et al. 2015; Kroeze et al. 2013; Sattar et al. 2014; Strokal & Kroeze 2013).

- Missing local nutrient sources

The *MARINA* model includes sources of nutrients in rivers that are important for China (Section 7.2.3, Table 7.3). Nevertheless, there are still some additional local sources of nutrients that the model ignored. These are, for example, aquaculture, industry and direct atmospheric N deposition on seas. Aquaculture and industry are modeled by IMAGE-GNM for TN and TP (Tables 7.1-7.3). In general, this model indicates the minor contribution of these sources to the total nutrient loads in surface waters globally (Beusen et al. 2015a). However, in China, the demand for fish products has been increasing (Biao & Kaijin 2007; Cao et al. 2007). Cities have been developing rapidly along with industries (Maimaitiming et al. 2013). Thus, local pollution may occur, such as polluted fish ponds from aquaculture and lakes in cities. In addition, direct N deposition on the sea was found to be an important polluter for specific coastal areas according to an experimental study of Xu et al. (2015). Thus, I realize that these local sources of water pollution are important to consider in local assessments. However, I believe that including them in the *MARINA* model will not change the major conclusions for China (see Section 7.4).

- Missing reservoir retentions

The impact of reservoirs on dissolved inorganic N and P export is considered in *MARINA*. However, this is not done for dissolved organic N and P. To my knowledge none of the selected models for China (Tables 7.1-7.3) account for dissolved organic N and P retentions in reservoirs. A few studies for the Yangtze addressed the side-effects of the largest dam on nutrient flows from up- to downstream (Sun et al. 2013a; Sun et al. 2013b), but mainly for TP and DIN. Global *NEWS*-2 does it only for dissolved inorganic N and P and IMAGE-GNM for TN and TP (Tables 7.1-7.3). I followed the approach of Global *NEWS*-2 when I developed *MARINA* for China. I, however, realize that this weakness may lead to an overestimation of river export of dissolved organic N and P, but it will not affect largely the conclusions of the study on pollution causes (see Section 7.2.5 for the model evaluation). Furthermore, the model evaluation results are promising. This provides a confidence in using the model for China (Section 7.2.5).

- Steady state approach

The *MARINA* model takes a steady state approach for nutrient cycling in terrestrial and aquatic systems, following Global *NEWS*-2 (Tables 7.1-7.3). Other models like IMAGE-GNM and SWAT take a dynamic approach. Both approaches have pros and cons. Cleary, a dynamic approach better represents processes of N and P in soils and in rivers. In IMAGE-GNM the spiraling approach is better at accounting for N and P retentions in rivers (Beusen et al. 2015a). The steady state approach is, however, much more transparent and flexible with modeling the source attribution for multiple nutrient forms. Furthermore, a model with a steady state approach is generally easier and faster to run. I realize that ignoring dynamics / spiraling in *MARINA* for China may introduce uncertainties in the model results. This may hold especially for future projections to account for time-delay responses in management options as concluded by Strokal and de Vries (2012) for world rivers. Nevertheless, my aim was to have a transparent model for other users, so that it can be easily applied to other world basins. And my choice is justified by the model evaluation results (Section 7.2.5).

o Calibration

The *MARINA* model is based largely on model parameters from Global *NEWS*-2 (Mayorga et al. 2010). A few of them are calibrated at the global scale. I did not re-calibrate Global *NEWS*-2 parameters because I did not have enough water quality data. One may argue that it is not appropriate to apply a calibrated parameter from a global model to China. However, the evaluation results show that the *MARINA* model performs better than the Global *NEWS*-2 model for China. This builds trust in the *MARINA* model (see discussion in Section 7.2.5). An interesting future exercise could be to have an uncalibrated, process-based version of *MARINA* as IMAGE-GNM is (Tables 7.1-7.3). Comparing the results of both approaches may help better understand uncertainties in both approaches to model nutrient export by rivers.

7.2.5 Building trust in nutrient models

Nutrient models such as IMAGE-GNM, SWAT, Global *NEWS*-2 (Tables 7.1-7.3) are often checked for their accuracy against measurements. Such validation is important, and widely used in modeling studies. It reveals information on the model performance in terms of indicators such as R_P^2 (the Pearson's coefficient of determination, 0–1), R_{NSE}^2 (the Nash-Sutcliffe efficiency, 0–1) or ME (Model Error) (Moriasi et al. 2007). However, these indicators are highly dependent on the number of measurements and their quality, which is often difficult to check. Furthermore, such model validation can be a challenge for regions like China where measurements at the sub-basin scale are not readily available. As a result, it is difficult to justify the model performance.

However, this type of model validation is only one of the possible options to build trust in the performance of nutrient models (Figure 7.2). Other options exist to evaluate different model aspects, and are not necessarily limited to the evaluation of only model outputs. In Figure 7.2, I indicate six possible options and model validation is only one of them. In Chapter 5, I used these six options to evaluate the *MARINA* model. Below I discuss briefly these options. I believe that the options can also be applied to evaluate other nutrient models, especially models that lack measurements.

Option 1 is the most common and aims to validate a model with observed nutrient *concentrations* at the river mouth (Figure 7.2). This option can serve as the first step to gain insights into the model performance if water quality data are available. For example, nutrient fluxes of the *MARINA* model were compared with available measurements from open literature (see Chapter 5 for references). The comparison of model results with the available measurements indicates a good performance of *MARINA*: R_P^2 is 0.84, R_{NSE}^2 is 0.78 and ME is 8%. Comparing these measurements with the original Global *NEWS*-2 values give lower R_P^2 (0.82) and R_{NSE}^2 (0.72) and higher ME (26%). In Chapter 2 I also evaluated Global *NEWS*-2, but only for DIN and DIP export by Yellow, Yangtze, Liao, Fuchunjiang (not part of the *MARINA* model) and Pearl rivers using the number of measurements available at that time. The results of Chapter 2 indicate lower R_{NSE}^2 (0.42) and higher ME (18%) compared to *MARINA*. All this indicates that the *MARINA* model performs better for Chinese basins than Global *NEWS*-2.

Option 2 is to compare modeled nutrient *trends* with available empirical studies (Figure 7.2). This comparison added to the confidence in the model performance. For example, nutrient export by Chinese rivers has increased since 1970 according to the *MARINA* model. This is in agreement with, for example, Li et al. (2014b), Tao et al. (2010), Ti et al. (2012), Wang et al. (2006), Li et al. (2007), and Liu et al. (2008a). I discuss this comparison in Section 7.3.1.

Option 3 is to perform *sensitivity analyses* (Figure 7.2). These indicate which model inputs and parameters influence largely river export of nutrients. For example, the outputs of the *MARINA* model are sensitive to changes in animal manure production, direct losses of manure to rivers of sub-basins (fractions), use of synthetic fertilizers and water discharges (see Chapter 5). This means that these model inputs and parameters are important to consider in assessing water quality in China.

Option 4 is to compare *model inputs* with other independent datasets (Figure 7.2). This provides an indication on the quality of model inputs. For example, I compared inputs used in *MARINA* with available county information for animal manure, synthetic fertilizers, human population and sub-basin areas (RESDC 2014). Results of this comparison increase the confidence in using gridded databases for sub-basin modeling in China: R_P^2 is in between 0.87 and 0.99 for compared model inputs (Chapter 5).

Option 5 is to account for *expert knowledge* (Figure 7.2). This option can be useful to get more insights into the accuracy of model parameters that are, for example, based on limited literature. For example, I asked for feedback from local experts from the Chinese Academy of Sciences and Peking University of China to help me to adjust some of the model parameters that are highly uncertain, but important in nutrient modeling.

Examples are parameters that I used to partition nutrient forms in animal manure and uncollected human waste. Furthermore, presenting results to experts, and potential users of the models can also increase confidence in the model. Indeed, I did this at various times during my PhD research.

Option 6 is to compare model results with other *modeling studies* (Figure 7.2). This option gives an indication of the model performance in comparison to other models. I did this for the *MARINA* model through Chapters 2-6 and I discuss this comparison in Section 7.3.1 of this chapter. An important component of this option is also to account for modeling studies that already evaluated uncertainties in modeling approaches. For the *MARINA* model these are existing studies of Global *NEWS*-2 (see Chapter 5 for details).

Combining the results of these six options increases trustworthiness when using *MARINA* to quantify past and future trends in river export of nutrients by source from sub-basins. Thus, I argue that comparing model results with observations is an important option, but it should be combined with the other possible options to obtain a better understanding of the model performance.

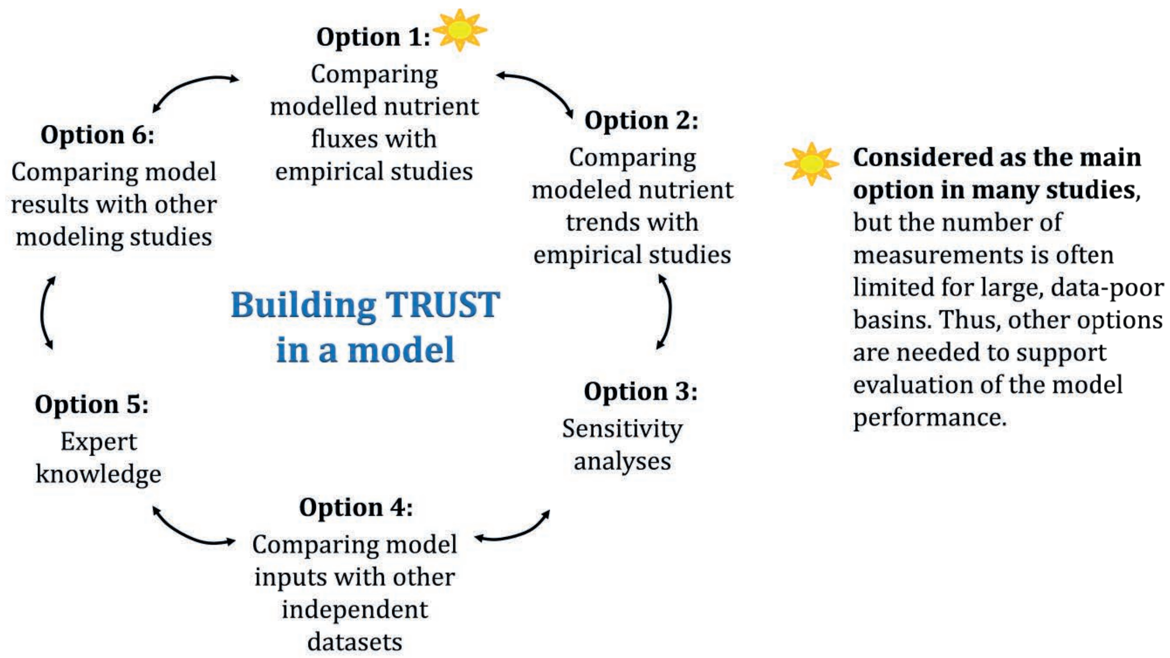

***Figure 7.2**. The cycle of building trust in nutrient modeling. This is based on my modeling experience (see Chapter 5 and Section 7.2.5).*

7.3 *MARINA* results in comparison with other studies

7.3.1 Comparison with other studies for China

The focus of this comparison is large-scale modeling of trends in river export of nutrients, sources of nutrients in Chinese rivers, impacts of nutrient exports and options to reduce nutrients in Chinese rivers. I pay particular attention to studies of Global *NEWS*-2 since this model formed the basis of the *MARINA* model.

- Trends in river export of nutrients

The *MARINA* results show increasing trends in nutrient export by Chinese rivers since 1970 (Section 7.4.2). These increasing trends agree with various modeling (e.g., Bao et al. 2006; Liu et al. 2008a; Qu & Kroeze 2010; Ti et al. 2012; Xing & Zhu 2002; Yan et al. 2010) and empirical studies (e.g., Li et al. 2014b; Li et al. 2007; Sun et al. 2013a; Tao et al. 2010; Wang et al. 2015). For example, studies of Global *NEWS*-2 quantify a factor of 2-4 increase in DIN and DIP export by the Yangtze and Pearl rivers between 1970 and 2000 (e.g., Qu & Kroeze 2010; Strokal et al. 2014b). This increase is in line with estimates of *MARINA* (see Chapter 5).

Other studies show similar increasing trends in DIN and DIP exports. Ti et al. (2012) report a factor of 1.4 increase in river export of DIN for the Yangtze between 1985 and 2005. This increase is a factor of 1.6 for the Yellow and Pearl rivers. *MARINA* quantifies similar increasing trends for the Yangtze (a factor of 1.9) and Pearl (a factor of 1.8), but higher for the Yellow river (a factor of 7) between 1970 and 2000 (Chapter 5). Tao et al. (2010) indicate an increase especially in N inputs to downstream surface waters of the Yellow since the 1970s. Wang et al. (2015) calculate a factor of 4-7 increase in DIN and DIP export by the Yangtze between 1970 and 2013. On the other hand, Powers et al. (2016) indicate somewhat a declining trend in P export by the Yangtze since 1970. This is not in line with *MARINA*, nor with the other aforementioned studies. I argue that Powers et al. (2016) may underestimate P pollution in the Yangtze, in particular from missing point sources of manure.

- Sources of nutrients in Chinese rivers

The *MARINA* model shows the large contribution of manure point sources to coastal water pollution in China (Section 7.4.1). This is a novel insight of this PhD thesis. Many earlier studies on river export did not come to this conclusion, because they did not consider manure point sources. This holds for Global *NEWS*-2 studies (e.g., Li et al. 2011b; Qu & Kroeze 2010; Yan et al. 2010) as well as for other individual studies (e.g., Bao et al. 2006; Liu et al. 2008a; Xing & Zhu 2002).

7

For example, in studies of Global *NEWS*-2 point sources are generally responsible for N and P pollution in the Bohai Gulf and diffuse sources for N and P (except for DIP) pollution in the Yellow Sea and South China Sea. In Global *NEWS*-2, point sources only include sewage effluents. Diffuse sources are, for example, mainly synthetic fertilizers and animal manure applications, and leaching of organic matter from agricultural land (Chapter 2 and Qu and Kroeze 2010). However, the *MARINA* model shows that manure point sources are important causes of N and P pollution in the three Chinese seas (Chapter 5). The other examples are individual studies for the Yangtze (e.g., Bao et al. 2006; Liu et al. 2008a) including studies of Global *NEWS*-2 (Li et al. 2011b; Yan et al. 2010). In general, these studies indicate the importance of synthetic fertilizers in N or P pollution of the river. The *MARINA* model recognizes this too, but mainly for DIN pollution of the river and emphasizes on the contribution of manure point sources for most of the dissolved nutrient forms.

Ignoring manure point sources may lead to an underestimation of river and coastal water pollution. *MARINA* estimates generally higher inputs of N to the Yangtze than the other studies. For example, Bao et al. (2006) and Liu et al. (2008a) quantified 2 Tg of N that was transported to waterbodies of the Yangtze in 1980. This is comparable to the estimates of *MARINA* for 1970 (1.7 Tg N) when the contribution of manure point sources was very small. However, estimates of *MARINA* are higher than that of Bao et al. (2006) and Liu et al. (2008a) for 2000. This is because 30-45% of N in animal manure was estimated to discharge to surface waters of the Yangtze sub-basins in 2000, causing nutrient pollution (Chapter 5). Estimates of *MARINA* are also higher than of Xing and Zhu (2002) for N inputs to waterbodies of the Yangtze, Yellow and Pearl rivers, likely for the same reason (see Chapters 4 and 5). It has to be noted that *MARINA* results are in agreement with observed concentrations of nutrients at the river mouth (see Table S5.11 for references and Section 7.2.5).

Estimates of *MARINA* for river export of nutrients differ from Global *NEWS*-2 for individual rivers. For some rivers *MARINA* values are lower which may seem surprising considering the fact that *MARINA* accounts for manure point sources as additional nutrient inputs to Chinese rivers. And, *MARINA* also considers uncollected human waste that may add more nutrients to rivers compared to Global *NEWS*-2. Nevertheless, lower values can be explained by the revised calculation of nutrient retentions during transport towards the river mouth. In general, *MARINA* calculates higher fractions of nutrient retention in the river systems than Global *NEWS*-2. This is because *MARINA* accounts for more dams and considers DIP retentions due to accumulation in sediments of rivers (details are in Chapter 5).

NUFER analyses (e.g., Bai et al. 2015; Ma et al. 2012; Ma et al. 2013a) agree with the *MARINA* model on manure as a source of nutrients in the environment. This is not surprising since *MARINA* was coupled with NUFER to account for manure point sources in river exports. This was done because NUFER provides nutrient losses from manure point sources. However, NUFER provides this information only for provinces or country and does not indicate in which river the manure is discharged. In addition, NUFER only quantifies nutrient losses from agriculture, but it does not quantify nutrient flows through rivers to seas (Section 7.2.1). Thus, coupling the river export *MARINA* model with NUFER was needed to assess impacts of manure point sources on Chinese rivers and seas at the sub-basin scale (see Section 7.2.3).

Another novel insight of *MARINA* is related to the sub-basin information on the impact of urbanization on water quality. It is a well-known fact that urbanization activities pollute Chinese aquatic systems (e.g., Liu & Diamond 2005; Qu & Kroeze 2010). The degree of pollution depends largely on how many people are connected to sewage systems and on technologies that treat human waste (Jin et al. 2014; Ju et al. 2005; Qu & Kroeze 2010). However, none of the existing studies quantified the effects of urbanization activities on water quality at the sub-basin scale for the main Chinese rivers. For example, *MARINA* shows that in 1970 direct inputs of untreated human waste to most Chinese rivers were more important than other sources. This holds especially for P in rivers of most sub-basins and for N in rivers of the northern sub-basins. This was a result of poor sanitation in 1970. However, by 2000 urbanization had expanded. *MARINA* shows that the share of urbanization in river pollution is generally higher in downstream sub-basins than in upstream. This is because downstream sub-basins are often densely populated river deltas with large cities.

- Impacts of nutrient exports

Both *MARINA* and Global *NEWS*-2 use the ICEP indicator to assess coastal eutrophication. In general, the *MARINA* model calculates higher ICEP values than Global *NEWS*-2 (Figure 7.3). This means that the potential for coastal eutrophication, and thus for the development of non-siliceous, harmful algae is higher in *MARINA* results. This can be explained by differences in N and P loads and stoichiometry. For example, the Yangtze and Yellow are identified as N limited rivers by *MARINA* and P limited by Global *NEWS*-2 for 1970 and 2000. This is because the new sources of nutrients considered in *MARINA* increase P inputs more than N inputs. In contrast, the Liao is P limited from 2000 according to *MARINA,* but N limited in Global *NEWS*-2. Results of *MARINA* correspond reasonably to some experimental studies. For example, some studies indicate P limitation for the South China Sea as also modeled by *MARINA* (Huang et al. 2003; Yin et al. 2004). Other studies indicate the importance of both N (Zhengyan et al. 2003) and P

(Xu et al. 2010b) for phytoplankton growth in the Bohai Gulf. For 2000, *MARINA* indicates N limitation for the Yellow and P limitation for the Liao draining into the Bohai Gulf (Figure 7.3).

MARINA results are generally in line with observed occurrences of harmful algae in Chinese seas (e.g., Li et al. 2014b; Tang et al. 2006a; Wang et al. 2008; Xiao et al. 2007). For example, Tang et al. (2006a) indicated that the reported number of occurred blooms along the coasts of the Yellow Sea increased from less than five before the 1970s to over a hundred during 2000-2004. Most of these events happened during May-June in the period of 2000-2004. A similar situation is indicated by Xiao et al. (2007) for Chinese coastal waters. However, not only coastal seas have been developing towards eutrophic states, but also inland surface waters (Huang 2015; Le et al. 2010; Xu et al. 2014), which is in line with *MARINA* results for river pollution in China (see Chapter 4).

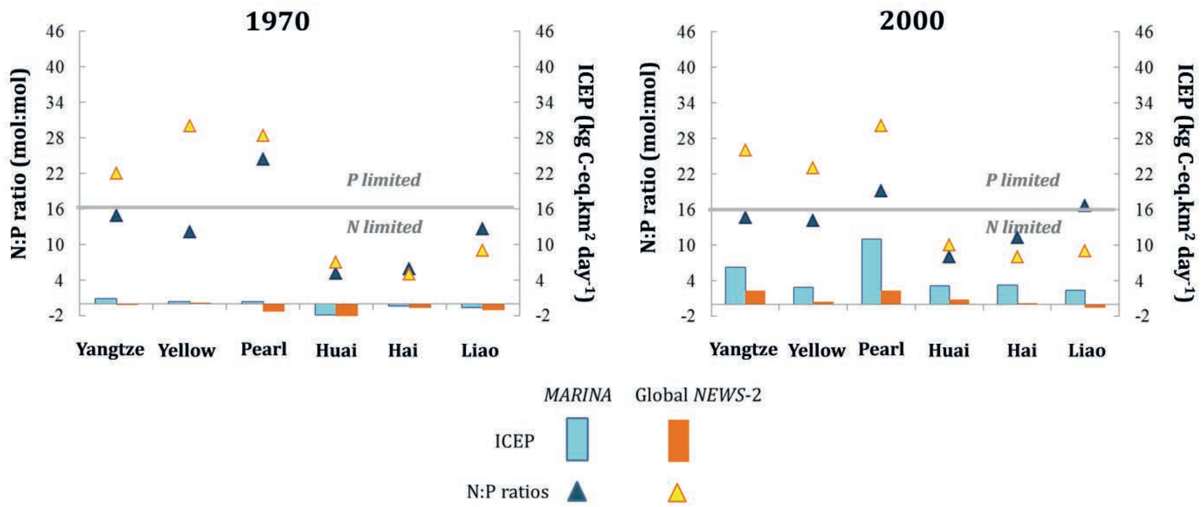

Figure 7.3. *Comparison of the MARINA model with Global NEWS-2 for N:P ratios (mol:mol) and ICEP (kg C-eq.km^{-2} day^{-1}) for 1970 and 2000. ICEP is the Indicator for Coastal Eutrophication Potential, with estimates based on Garnier et al. (2010). P limited rivers occur when their N:P ratio is above 16, and N limited when their ratio is below 16 according to the Redfield ratio of N:P. 2050 is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment. MARINA is short for Model to Assess River Inputs of Nutrients to seAs. Global NEWS-2 is short for Nutrient Export from WaterSheds. Source: the MARINA (Chapter 5) and Global NEWS-2 models (Chapter 2).*

o Options to reduce water pollution

From the *MARINA* model we now know how important the manure point sources are for river and coastal water pollution in China. We also have a better understanding of the relative importance of sewage point sources in water pollution of urbanized areas. Thus, the *MARINA* results indicate that effective policies should focus on point sources to reduce coastal eutrophication. This is different from many earlier studies (see below).

Many earlier studies recommended to focus on diffuse sources when reducing agricultural-related water pollution problems. These are, for example, Global *NEWS*-2 studies for river export of nutrients (e.g., Li et al. 2011b; Qu & Kroeze 2010; Yan et al. 2010) and experimental studies for crop production (Chen et al. 2011; Chen et al. 2014; Ju et al. 2009; Miao et al. 2010). In general, those earlier studies recommend options to reduce overuse of synthetic fertilizers. For example, the integrated soil-crop system management has been suggested (Chen et al. 2011). Recycling of manure is generally not considered in these options. According to *MARINA*, however, manure recycling is essential to improve river quality (see below). Only the NUFER scenarios account for both nutrient losses from fertilized fields and animal production, but these scenarios are limited to the food chain and do not account for nutrient flows in the environment (e.g., Bai et al. 2015; Ma et al. 2013a).

I recommend to avoid manure point sources by recycling most available manure in crop production, instead of using synthetic fertilizers. I incorporated this option in my optimistic scenarios. Results show the high effectiveness of this option to clean rivers and reduce coastal eutrophication by 2050 (Chapter 6). This insight is useful for developing effective environmental policies for Chinese agriculture. In fact, several environmental policies in China were introduced to control manure-related pollution since the 2000s (e.g., MOA 2011a; Zheng 2013). The most recent policy is "Zero Growth in Synthetic Fertilizers after 2020" with plans to recycle 60% of available manure in 2020 (MOA 2015). These policy plans are a good start in reducing manure losses to rivers, but may not be enough to reduce coastal eutrophication in 2050, according to my study. More manure recycling is needed as shown in my optimistic scenarios (Section 7.4.2). To optimize this, integrated nutrient management needs to be combined with advanced technologies as assumed in my optimistic scenarios. Examples of such combinations can be balanced fertilization with technologies to inject the required amount of manure into the soil (e.g., Oenema et al. 2012). In addition, many technologies exist to reduce gaseous emissions from animal houses and to improve manure processing (like composting or anaerobic digestion) (e.g., Loyon et al. 2016; Magrí et al. 2013; Oenema et al. 2009). These can contribute to close nutrient cycles in agriculture.

My recommendations for sewage point sources are to improve wastewater treatment. In my optimistic scenarios, I show the effectiveness of increasing nutrient removal up to 80-90% to reduce coastal eutrophication in 2050 (see Section 7.4.2). The effectiveness of better wastewater treatment was also shown in Global *NEWS*-2 studies, but not at the sub-basin scale (Qu & Kroeze 2012). Jin et al. (2014) emphasize a strong need to upgrade current wastewater treatment plants (WWTPs) with more advanced technologies. This is because many current WWTPs rely on outdated technologies that are often ineffective for nutrient removal, yet require considerable energy for aeration and produce a lot of sludge that needs proper management (see Jin et al. (2014) for an overview). Today, advanced technologies are able to remove most of N and P from waste during treatment and produce less sludge at the lower cost for energy (e.g., Kartal et al. 2010; Khiewwijit 2016). Combining them with technologies that allow nutrient recovery and recycling can facilitate to close nutrient cycles (e.g., Cai et al. 2013; Jaffer et al. 2002; Tervahauta et al. 2014).

In optimistic futures for China, we need implementation of best available today technologies (see examples above). A challenge can be to upscale emerging technologies (e.g., Chen et al. 2015; Tervahauta et al. 2014). Nevertheless, the economy in China develops quickly. This opens up opportunities to explore the implementation of the technologies to reduce water pollution from animal and human waste in the future.

7.3.2 Comparison with Global *NEWS*-2 studies for other regions

Coastal eutrophication has not only been observed in Chinese seas, but also in many other coastal waters of the world (Diaz & Rosenberg 2008; Garnier et al. 2010; UNEP 2016). Many events of harmful algal blooms have been reported in coastal waters of the Americas, Europe, and South Asia (Diaz & Rosenberg 2008). The Global *NEWS*-2 model is a widely used tool for global and regional analyses as I indicated in Section 7.2.1. To my knowledge, 35 studies have used the model since 2010 (Figure 7.4). Most of these are for regional analyses in China, Indonesia, the Bay of Bengal, Europe, North and South America and Africa (Figure 7.4). I contributed to approximately one-third of these global and regional analyses (Figures 7.4 and 7.5).

Figure 7.5 distinguishes between four types of Global *NEWS*-2 studies. *The first type* is typically studies that used the original version of Global *NEWS*-2. In 2010, this version was published with applications of MEA scenarios for future projections (Mayorga et al. 2010; Seitzinger et al. 2010). This version was used in around 20 studies for different regions (Figure 7.5). Half of these studies developed alternative scenarios to MEA. These studies belong to *the second type* in Figure 7.5 and are for different regions. They focus on different aspects such as effects of alternative agricultural scenarios including energy crops on coastal eutrophication or how people experience coastal eutrophication (the lived experience) (Table 7.4).

The third type includes the studies that developed new formulations of Global *NEWS*-2 since 2010. For example, for Indonesia new model formulations have been developed for dams (Suwarno et al. 2014b) and for direct sewage inputs to rivers (Suwarno et al. 2014a). For the Bay of Bengal, new model formulations are developed for open defecation (Nurul et al. under review) and aquaculture (Sattar et al. 2014). Two global studies include new formulations; the first is a version of Global *NEWS*-2 that includes seasonality for DIN (McCrackin et al. 2014) and the second includes soil P dynamics (Strokal & de Vries 2012). But these formulations for global analyses are not integrated into one new model system (Table 7.4). *The fourth type* includes the development of a new model. This is what I did: I developed a new *MARINA* model for China based on Global *NEWS*-2 (this PhD thesis). I consider this as a new model because it differs more from Global *NEWS*-2 in terms of model inputs and formulations than any of the other studies. In particular, it differs with respect to the spatial scale (sub-basin), aquatic retentions and sources of nutrients in rivers (see Section 7.2).

The results of applying the models (both original and improved) reveal similarities and differences between China and other regions (see Table 7.4 for examples). Similarities are mainly associated with trends in river export of nutrients since 1970 and the effects of these trends on coastal eutrophication. For example, the overall conclusion of the studies from Table 7.4 is that nutrient export by many world rivers has been increasing since 1970 (Mayorga et al. 2010), resulting in coastal eutrophication. This holds particularly true for rivers in North America (McCrackin et al. 2013), Europe (Blaas & Kroeze 2016; Strokal & Kroeze 2013), Asia (Sattar et al. 2014; Strokal et al. 2014b; Suwarno et al. 2013) and for some rivers in South America (Van der Struijk & Kroeze 2010), according to the original Global *NEWS*-2 model. Similar conclusions can be drawn for Chines rivers (Qu & Kroeze 2010; Strokal et al. 2014b).

However, there are differences in nutrient pollution levels between the original and improved versions of Global *NEWS*-2. For example, new model formulations for the Bay of Bengal (Nurul et al. under review), Indonesia (Suwarno et al. 2014a), and the *MARINA* model for China (this PhD thesis) suggest higher levels of nutrients in their rivers than the original Global *NEWS*-2. This has to do with missing sources of nutrients in the original model (e.g., Section 7.3.1 for China). Furthermore, the original Global *NEWS*-2 was found to underestimate the effects of dams on nutrient export by Indonesian rivers. Similar conclusions are drawn in this PhD thesis for Chinese rivers.

An important difference between China and other regions is in causes of water pollution (Table 7.4 as an example). For China the large contribution of manure point sources may be unique, but in other regions, this source could be unimportant. This pollution is largely from industrial farms with inadequate manure management (Chapter 4, Sections 7.2.3 and 7.3.1). Animal production has also been industrializing in other world regions such as North America and Europe (e.g., Herrero et al. 2013; Steinfeld et al. 2006). However, management of animal manure in those regions is apparently more effective than in China. For example, discharges of manure to water bodies are strictly controlled by permits in North America (Hribar 2010) and recycling of manure is regulated in

Europe (Oenema et al. 2007). In China, new environmental policies for animal production have been recently introduced but require time and efforts for their effective implementation (Section 7.3.1).

In contrast, open defecation is probably not an important source of water pollution in China, but it may be for other Asian rivers, e.g. draining into the Bay of Bengal (Nurul et al. under review) and the Java Sea (Suwarno et al. 2014a). This is because in China less than five percent of the human population practiced open defecation in 2000 (WHO/UNICEF 2014). This is not the case for countries including India, Bangladesh and Pakistan which contain rivers draining into the Bay of Bengal. This holds especially for rural populations in those countries as over two-thirds, half and one-fifth of rural populations in India, Pakistan and Bangladesh practice open defecation, respectively (WHO/UNICEF 2014). For Indonesia, this number was one-fifth, but for the urban population in 2000 (Suwarno et al. 2014a). As a result, open defecation became a large contributor to nutrient pollution in rivers of those countries. This also contributes to coastal eutrophication (Nurul et al. under review; Suwarno et al. 2014a).

Urban waste is today an important source of P in Chinese rivers especially in urbanized deltas with large cities such as Shanghai (Yangtze river delta), Shenzhen and Guangzhou (Pearl river delta). Sewage from the urban population is also an important driver of P pollution in many other world rivers (Harrison et al. 2010; Mayorga et al. 2010; Strokal & de Vries 2012). However, for the rivers of Indonesia this source may be one of the most important causes of N and P pollution in the future (Suwarno et al. 2014a). For example, Suwarno et al. (2014a) calculated a factor of 20-40 increase in nutrient inputs to Indonesian rivers between 2000 and 2050 using the improved version of Global *NEWS*-2. This increase is a result of rising urban population connected to sewage systems with relatively low nutrient removal efficiencies.

Future trends in river export of nutrients differ among regions and depend on scenarios (Figure 7.5 and Table 7.4). In general, MEA scenarios with globalized socio-economic trends and with a reactive approach for environmental management (the GO scenario) project higher exports of dissolved nutrients by world rivers in the coming years (Seitzinger et al. 2010). However, these trends still differ among the regions. The difference is especially visible between Europe and Asia. European rivers may become cleaner in the future as a result of effective environmental policies. In contrast, Asian rivers may be more polluted due to a lack of environmental policies based on the GO scenario. Examples are rivers in China (this PhD thesis), Indonesia (Suwarno et al. 2013) and Bay of Bengal (Sattar et al. 2014).

However, the results of alternative scenarios differ from the results of the MEA scenarios and among regions (Figure 7.5 and Table 7.4). For example, in European seas, the potential for coastal eutrophication may become higher in the future if large-scale production of micro-algae for biodiesel increases in European countries, according to Blaas and Kroeze (2014). This is a result of nutrient losses to water systems during micro-algae production on land. Strokal et al. (2014c) showed that increasing nutrient removal during treatment is more effective to reduce coastal eutrophication in the southern Black Sea and increasing nutrient efficiencies in agriculture is more effective to reduce coastal eutrophication in the northern Black Sea. Biogas production may be a solution to reduce water pollution in Indonesian rivers by reducing losses of human waste (Suwarno 2015). Zinia and Kroeze (2015) analyzed expectations of people to future changes and their lived experience in Bangladesh draining into the Bay of Bengal. For China, I developed different alternative scenarios with the focus on optimistic perspectives. I also explored the effectiveness of the recent environmental policy on reducing water pollution. I discuss and present the results of my scenarios in Sections 7.3.1 and 7.4.2.

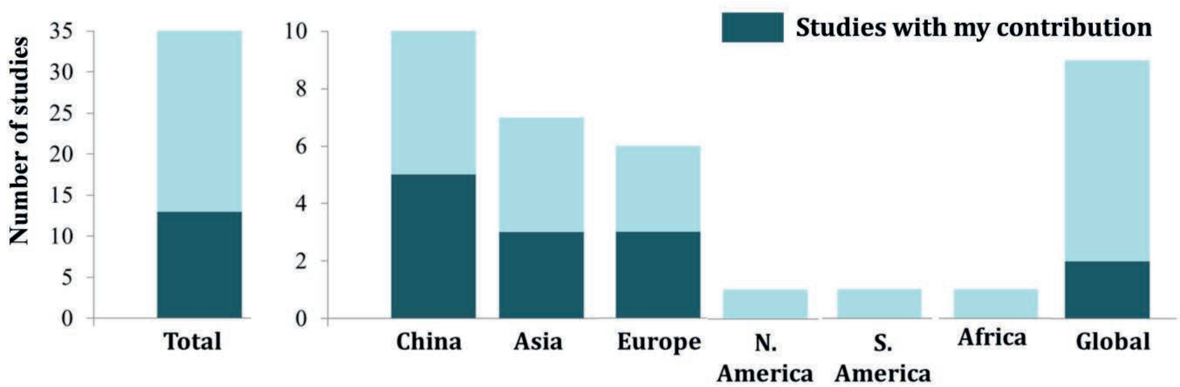

Figure 7.4. Relevant studies on river export of nutrients that used the original Global NEWS-2 model or the MARINA model since 2010. Details and references are given in Figure 7.5. Asia includes rivers draining into the Bay of Bengal and Indonesia. Europe includes rivers draining into the Black Sea and other European Seas such as the Mediterranean, Baltic and North seas. N and S stand for North and South.

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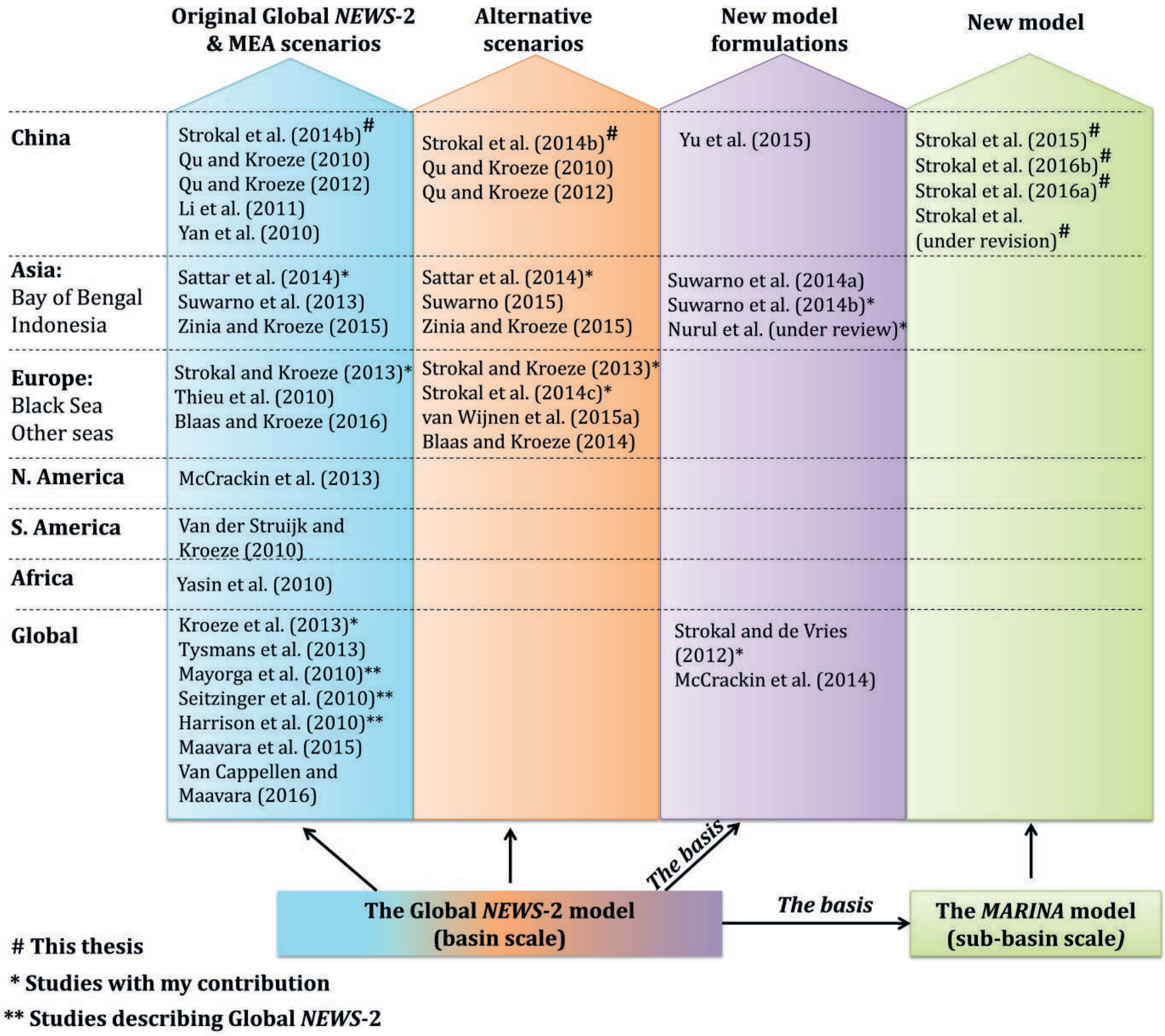

Figure 7.5. *Details on Global NEWS-2 and MARINA shown in Figure 7.4. See Table 7.4 for aspects that new model formulations differ from the original Global NEWS-2 model and for alternative scenarios. MEA = Millennium Ecosystem Assessment.*

Table 7.4. *Comparison of selected studies using Global NEWS-2 and MARINA.*

Region	New developments and alternative scenarios	Dominant sources of nutrients in rivers	References
China	o *MARINA* (new model), differs from Global *NEWS*-2 in: - sub-basin scale - dams and P accumulation in rivers - manure point sources - uncollected human waste o Alternative optimistic scenarios	o Point inputs of manure for N and P o Sewage for P in urbanized areas	This study, Chapters 2-6
Asia:			
The Bay of Bengal	o New model formulations for: - open defecation - aquaculture o Analysis of lived experience of future trends	o Open defecation for N and P o Agriculture for N	Zinia and Kroeze (2015) Nurul et al. (under review) Sattar et al. (2014)
Indonesia	o New model formulation for: - dams - direct inputs of sewage o Alternative scenarios for biogas	o Direct sewage inputs for N and P	Suwarno et al. (2014a) Suwarno et al. (2014b)
Europe:			
The Black Sea	o Original Global *NEWS*-2 o Alternative scenarios for sewage and agriculture	o Agriculture in the north o Sewage in the south	Strokal and Kroeze (2013) Strokal et al. (2014c)
European seas	o Original Global *NEWS*-2[(a)] o Alternative scenarios for biodiesel	o Agriculture or sewage depending on limiting nutrient	Blaas and Kroeze (2016) Blaas and Kroeze (2014)
North America	o Original Global *NEWS*-2[(b)] o Original MEA scenarios	o Agriculture N (fertilizers, manure and crop N fixation)	McCrackin et al. (2013)
South America	o Original Global *NEWS*-2 o Original MEA scenarios	o Agriculture for DIN[(f)] o Sewage for DIP[(f)] o Leaching of organic matter for DON and DOP[(f)]	Van der Struijk and Kroeze (2010)
Africa	o Original Global *NEWS*-2 o Original MEA scenarios	o Agriculture, especially manure for N o Sewage for P	Yasin et al. (2010)
Global	o New model formulations[(c)] for: - P soil dynamics[(d)] - DIN seasonality[(e)]	o Sewage for P globally o Agriculture for N globally o Vary among regions	Strokal and de Vries (2012) McCrackin et al. (2014)

(a) The study area includes 48 rivers draining into the North Sea, Mediterranean Sea, Black Sea, Baltic Sea, Atlantic Ocean. **(b)** Includes the study area of 113 river basins. **(c)** Global analyses with the 2010 Global *NEWS*-2 version include over 6000 rivers. **(d)** Global analyses include around 500 large rivers. **(e)** Global analyses include 77 rivers. **(f)** DIN = dissolved inorganic nitrogen. DIP = dissolved inorganic phosphorus. DON = dissolved organic nitrogen. DOP = dissolved organic phosphorus.

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7.4 Conclusions of the PhD thesis

7.4.1 Main lessons for nutrient modeling

From my study I can draw five main lessons for nutrient modeling at the sub-basin scale.

Lesson 1: The MARINA model can help to better understand causes of water pollution and explore solutions

The *MARINA* model has been developed to better understand trends in river inputs of nutrients to Chinese seas and their sources in a spatially explicit way. The most important difference between the *MARINA* model and the other models is that it quantifies the relative importance of human activities to coastal water pollution in a better way for China and by sub-basin (see Lesson 2 for sub-basins). This helps to identify the main causes of coastal water pollution by nutrients. For example, with this model we now better understand the impact of animal production and uncollected human waste on water quality in China (Chapters 4 and 5). The model is also useful to explore solutions to reduce coastal water pollution by 2050. I show this in Chapter 6 for China.

Lesson 2: Sub-basin scale modeling is useful support for effective nutrient management in large basins

My sub-basin scale modeling approach is meant for large basins such as the Yangtze, Pearl and Yellow. It identifies areas with human activities that pollute coastal waters with nutrients (see Lesson 1). This information can help to allocate nutrient management and thus effectively reduce coastal water pollution from large basins. This makes the sub-basin scale approach of the *MARINA* model a useful tool to support decision making (see also Section 7.2.3). I believe that the sub-basin scale modeling is also an appropriate alternative to, for example, basin or gridded scale modeling to address nutrient flows from large basins. This is because it provides the useful information with minimum efforts in terms of model inputs and computation time. In contrast, basin scale modeling does not account for the spatial variation in human activities within a basin. Meanwhile, gridded scale modeling requires more inputs and computation time, and may also need more simplifications and assumptions. This can increase uncertainties, particularly in data scarce regions. Another important feature of the sub-basin scale *MARINA* model is its transparency. This makes the model easy to understand by other users and thus to apply it to the other large basins.

Lesson 3: Preferred sizes of sub-basins depend on the research objectives and resource availability

What is a preferred or appropriate size of a sub-basin? I think part of this answer is related to the research objective and another part to the availability of resources such as time, finances and data. For *MARINA*, many model inputs were available at 0.5 latitude by 0.5 longitude grid. These gridded data were easily aggregated to sub-basins, making modeling of nutrients fluxes at the sub-basin scale possible. The *MARINA* sub-basins were delineated using the river network of the WBM model (STN-30 at 0.5 latitude by 0.5 longitude). The intention was to cover the primary tributaries of a large river. This resulted in 22 sub-basins of the Yellow, Yangtze and Pearl rivers with drainage areas ranging from 31 and 460 thousands km^2. Classifying up-, middle- and downstream sub-basins allowed us to account for upstream and downstream relationships (see research objectives in Chapter 1). This delineation and classification was supported by literature (see Chapter 5). My research shows that my choices on the sizes and delineation of sub-basins provide the information needed to meet thesis' research objectives (see Lessons 1 and 2).

Lesson 4: Basin scale approaches can serve as a basis for sub-basin scale modeling of nutrient fluxes

The *MARINA* model has been developed based on basin scale approaches from Global *NEWS*-2. Implementing these basin approaches for sub-basins proved to be a good choice for China. Some of the Global *NEWS*-2 approaches for animal manure and human waste were improved to include missing sources of nutrients in the studied rivers. Evaluation results of *MARINA* justify the choice of applying basin scale approaches for sub-basin modeling of nutrient fluxes. This is shown by the evaluation results of *MARINA*, indicating its better performance over Global *NEWS*-2 for China (Lesson 5, Section 7.2.5). This study can also serve as an example for other large basins in the world. However, it has to be noted that the improvements of Global *NEWS*-2 approaches for animal manure and human waste were specific for China. Other basins may require improvements in modeling approaches for the other specific sources (e.g., open defecation for the Bay of Bengal and Java Sea; Section 7.3.2).

Lesson 5: Building trust in nutrient models is more than just comparing modeled and measured data

Results of nutrient models are often validated against measurements. This type of validation depends largely on the quality and availability of such water quality data. When these data are scarce, model validation may not provide enough confidence in the model performance. However, such model validation is not the only option to build trust in nutrient models. In this PhD thesis, I explored several options (including validation based on empirical data), a combination of which can increase our trust in the use of models for nutrient analyses. I started with comparing modeled nutrient fluxes with available measurements as many studies do. Furthermore, I compared modeled trends in river exports of nutrients with available monitoring studies. Comparison of model inputs with an independent dataset, sensitivity analyses and expert knowledge helped to justify model inputs and parameters. Finally, I looked at previous modeling studies on river export of nutrients for China and compared them to *MARINA* results to get insights into differences and similarities. Results of all these options build adequate trust in using the *MARINA* model for its purpose. I believe that the proposed options for the *MARINA* model can also be useful for other studies to evaluate their nutrient models. This holds especially for studies modeling large basins for which water quality data are often limited.

7.4.2 Main findings of the results

In the following I summarize the six main findings of the *MARINA* model results. The conclusions of the chapters are summarized in Figure 7.6.

Finding 1: Dissolved N and P export by Chinese rivers increased by a factor of 2-8 between 1970 and 2000

River export of dissolved N and P to the Bohai Gulf increased by a factor of 6-8, to the Yellow Sea by a factor of 2-5 and to the South China Sea by a factor of 2-4 from 1970 to 2000 (ranges are for nutrient forms). The *MARINA* model includes six rivers draining into three major Chinese seas. The Yellow, Hai and Liao rivers are located in the northern part of China and drain into the Bohai Gulf. The Yangtze and Huai drain into the Yellow Sea, and the Pearl drains into the South China Sea. The Yellow, Yangtze and Pearl rivers have large drainage areas that are subdivided into sub-basins. These large rivers are dominant exporters of the nutrients to the Bohai Gulf, Yellow Sea and South China Sea.

Finding 2: The potential for coastal eutrophication was low in 1970 and high in 2000 in China

Increased river export of dissolved N and P led to higher potential for coastal eutrophication in 2000. I evaluated the potential for coastal eutrophication using the ICEP indicator that is based on ratios of N:P:Si in coastal waters (see Chapter 5). In 1970, all studied rivers had an ICEP close to zero (the range is from -1.8 to 0.8 kg C-eq. km^{-2} $year^{-1}$). This indicates a low potential for coastal eutrophication and thus for blooms of harmful algae. However, by 2000 this potential increased because rivers transported more dissolved N and P from human activities and less Si due to river damming.

Finding 3: Most dissolved N and P in Chinese seas is from middlestream and downstream activities

The Yangtze and Pearl rivers exported over half of the nutrients from middle- and downstream human activities in 2000 (see Finding 4 on human activities). The Yellow River exported up to 70% of dissolved inorganic N and P from downstream and of dissolved organic N and P from middlestream activities. Sub-basins that are located closer to coastal waters are generally more urbanized than upstream sub-basins. In general, the amount of nutrients reaching coastal waters is a net-effect of human activities on land and nutrient retentions and losses during transport to the coastal waters.

Finding 4: Manure point sources are responsible for 20-80% of dissolved N and P in Chinese rivers

Manure point sources contributed 60-80% of dissolved N and P to the Bohai Gulf and to 20-75% to the Yellow Sea and South China Sea in 2000. These manure point sources occurred as a result of agricultural transitions, which started in the 1990s. These transitions include changes in animal production from traditional-oriented farming systems to industrial-oriented farming systems. However, industrial farms are largely disconnected from crop production. Thus animal manure is often not applied on land, which is different from the past with traditional farms. Furthermore, management of animal manure is poor. All this has resulted in direct discharges of manure to water systems in China (thus point manure sources). The share of these manure point sources is larger for rivers of the Bohai Gulf than for the other rivers. This is because the other rivers receive more nutrients from diffuse sources as they have higher runoff from land than rivers of the Bohai Gulf. These diffuse sources include synthetic fertilizers (for dissolved inorganic N), leaching of organic matter (for dissolved organic N and P) and weathering of P-contained mineral (for dissolved inorganic P). Sewage is an important contributor to dissolved P in coastal waters of urbanized areas.

Finding 5: In the future river export of nutrients may increase, and current policy plans may not be sufficient to avoid this increase

Rivers may transport up to 90% more nutrients to the Chinese seas in 2050 than in 2000 (except for DIN export by the Hai). Thus, the potential for coastal eutrophication may increase (Figure 7.7). These projections are based on the GO scenario of the MEA. This scenario projects the consequences of globalization trends in socio-economic development with a reactive management of environmental problems.

This scenario assumes that animal production will keep industrializing along with urbanization. However, inadequate nutrient management in agriculture and sewage will lead to large losses of nutrients to the rivers, causing nutrient pollution in 2050. Thus, this GO scenario is the worst case for the environment.

Similar conclusions hold for the CP scenario (Figure 7.7). This CP scenario is an alternative to GO, but incorporates the current plans (CP) from the recent "Zero Growth in Synthetic Fertilizers after 2020" policy. These plans aim at 60% recycling of manure on land and at zero growth in synthetic fertilizers after 2020. The *MARINA* results for the CP scenario show that many rivers may still export more nutrients by 2050 (up to two-thirds more than in 2000). This indicates that the current plans are a good start, but they may not be enough to reduce coastal eutrophication by 2050.

Finding 6: In an optimistic world the potential for coastal eutrophication is low in 2050 as a result of assumed full implementation of (1) high recycling rates of animal manure and (2) high efficiencies of nutrient removal in sewage systems

In 2050, river export of most N and P is projected to be at around 1970 levels in my optimistic scenarios (OPT-1 and OPT-2). Thus, a low potential for coastal eutrophication is projected (Figure 7.7). I developed OPT-1 and OPT-2 using the GO scenario as a basis for socio-economic development. Efficient nutrient management in agriculture (OPT-1 and OPT-2) and sewage (OPT-2) are important strategies to reduce coastal eutrophication. In particular, maximum recycling of manure on land is the most effective strategy. This is because direct discharges of manure to rivers are avoided (Figure 7.7). This resulted in zero inputs of nutrients from manure point sources, decreasing river export of nutrients by 45-90% to the Bohai Gulf between 2000 and 2050 (the ranges are for nutrient forms and rivers). This decrease is 7-93% for the Yellow Sea and South China Sea. The efficient use of synthetic fertilizers in cropland is more effective to reduce dissolved N inputs to the Yangtze (draining into the Yellow Sea) and Pearl (draining into the South China Sea) rivers. In my OPT-2 all human waste is collected by centralized systems from urban people and by decentralized systems from rural people. High nutrient removal rates during sewage treatment are effective to reduce dissolved P export by rivers from urbanized areas (Figure 7.7). The optimistic scenarios are considered as the best cases for the environment.

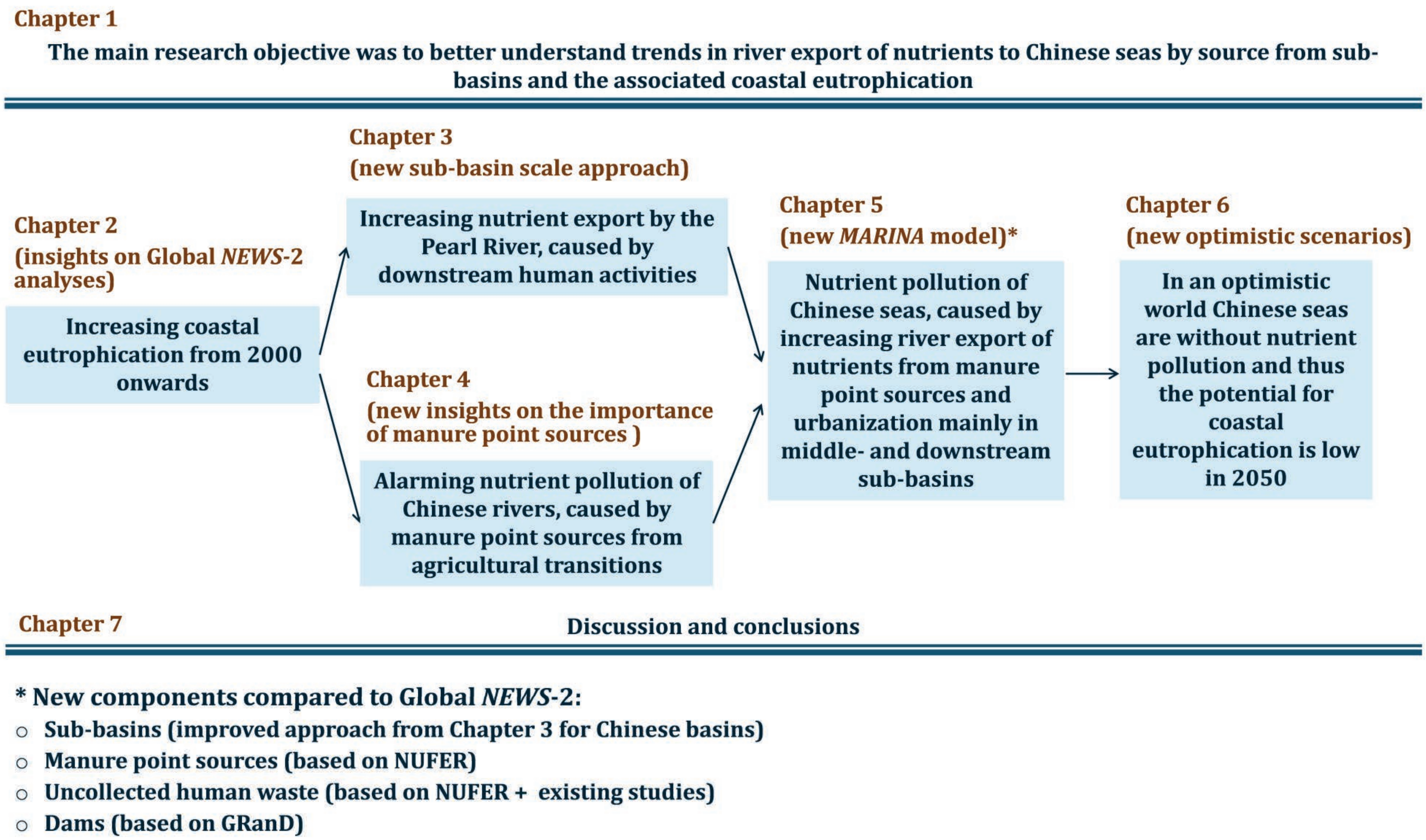

Figure 7.6. *Summary of conclusions and novelties of Chapters 2-6. Note that the detailed conclusions of Chapters 2-4 differ to some extent from the conclusions of the final MARINA model results. For example, the source attributions differ among the chapters because the modeling approach improved when the research evolved.*

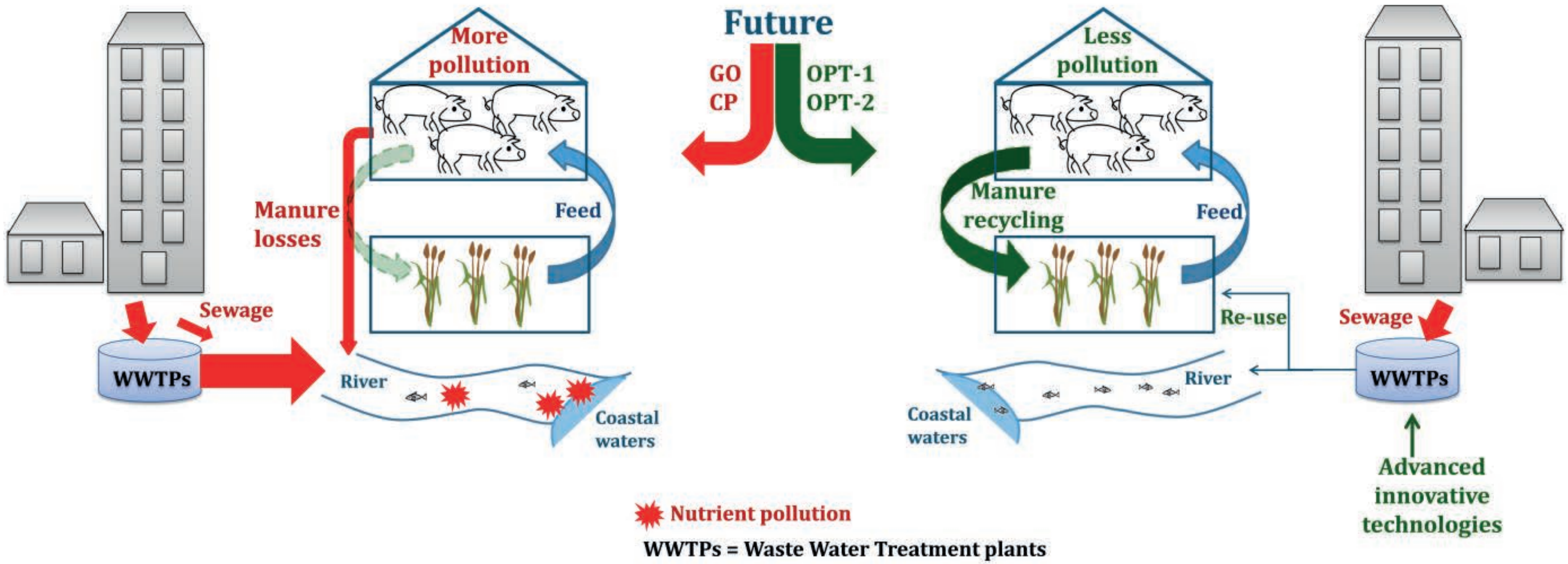

Figure 7.7. *Illustration of future scenarios for coastal water quality in China. GO is Global Orchestration of the Millennium Ecosystem Assessment and assumes environmental actions that are either absent or ineffective in reducing water pollution. CP is based on GO, but incorporates the "Zero Growth in Synthetic Fertilizers after 2020" policy. OPT-1 and OP-2 are optimistic scenarios that assume high nutrient use efficiencies in agriculture (OPT-1, OPT-2) and sewage (OPT-2).*

7.5 Future outlook

o Improving *MARINA* for river export of nutrients in China

MARINA, as any other model, has room for improvements. From the model limitations that are discussed in Section 7.2.4 we can identify the following points for improvement:

- Improving spatial and temporal level of detail for local water pollution assessments;
- Including local sources of nutrients in rivers such as aquaculture, industry, and atmospheric N deposition on sea;
- Including reservoir retentions for dissolved organic N and P;
- Improving modeling of nutrient cycles by accounting for dynamics;
- Recalibrating model parameters, which will require more water quality data.

In addition, a next version of the *MARINA* model should also account for sub-basin modeling of particulate forms of N and P. *MARINA*, so far, accounts for dissolved forms of N and P at the sub-basin scale. It is the first sub-basin model version for China's past and future trends in river export of dissolved inorganic and organic N and P. Thus, the model can be further expanded to particulate forms and to account for current trends. Approaches of Global *NEWS*-2 can be used as a starting point to include particulate N and P in a next version of *MARINA* as it was done similarly for dissolved N and P. Sources like Chinese Yearbooks (e.g., MOA 2006) and county datasets (Wang et al. in preparation) can provide some information for model inputs. This gives the opportunity to update the model for the current year. Alternatively, scenarios from the Shared Socioeconomic Pathways (SSPs, O'Neill et al. 2014) and Representative Concentration Pathways (RCPs, Van Vuuren et al. 2011) can be implemented instead of MEA scenarios.

o Using the sub-basin approach of *MARINA* for transboundary rivers

The sub-basin scale approach of the *MARINA* model can also be useful for transboundary river basins. There are over two hundred transboundary rivers in the world. These rivers have often large drainage areas because of crossing more than one country (UNEP-DHI & UNEP 2016). These river basins accommodate less than half of the world population and are important habitats for a variety of living organisms. These rivers account for approximately half of the global water discharge (UNEP-DHI & UNEP 2016). Thus, these rivers may bring considerable amounts of pollutants to coastal waters from human activities taking place in different countries. Applying the sub-basin scale approach of the *MARINA* model will allow to identify "hot" spots (i.e., sub-basins) within the large transboundary river basins. This information can give more insights on where reduction of water pollution is required and from which human activities. This information may be helpful for cooperation between countries when formulating effective management options to reduce river and coastal water pollution.

o Modeling other pollutants with *MARINA*

In this PhD thesis, the focus was on nutrients as pollutants of rivers and coastal waters in China. However, rivers in China and also rivers in other parts of the world can contain other pollutants from human activities (UNEP 2016) such as heavy metals, pathogens (Hofstra et al. 2013; Vermeulen et al. 2015a), plastics (Koelmans et al. 2016; Siegfried et al. 2016) and chemicals (van Wijnen et al. in preparation). Similar to nutrients, these pollutants can be transported to coastal waters, causing pollution (Schwarzenbach et al. 2006). Integrating nutrients with other pollutants in modeling can contribute to a comprehensive water quality assessment. This assessment can support the development of effective pathways to meet Sustainable Development Goals (SDGs) of the United Nation (Griggs et al. 2013).

The *MARINA* model can serve as a basis to develop a multi-pollutant model. This is because human activities that contribute to nutrients in rivers can also contribute to other pollutants (e.g., heavy metals, toxic chemicals, micro-plastics). For example, sanitation (e.g., open defecation) and urbanization are shared sources of nutrients, and pathogens in aquatic systems (Vermeulen et al. 2015a; Vermeulen et al. 2015b). Agriculture can also contribute to pesticides in rivers via leaching and runoff in addition to nutrients (e.g., Alavanja et al. 2003; Chau et al. 2015). Mining of, for example, P-rocks to produce synthetic P fertilizers can add not only P to nearby water systems, but also heavy metals (Akcil et al. 2015). Some industrial influents contain a lot of heavy metals. Rivers receive large amounts of heavy metals especially when wastewater treatment is poor to remove these pollutants from sewage and industrial influents (e.g., Akcil et al. 2015; Wu et al. 2016). Wastewater can also contain residues of consumer products (e.g., soup, toothpaste) and thus be of a source of chemicals such as triclosan (van Wijnen et al. in preparation) and also micro-plastics in water systems (Siegfried et al. 2016).

However, the biogeochemistry (e.g., retention processes in soils and rivers) differs among pollutants. For example, retention of persistent heavy metals with high mobility in the environment (e.g., cadmium, zinc, lead, arsenic; cannot be degraded) differs from that of short-living pathogens. Micro-plastics can also accumulate in fish stock and in sediments for a long time. Thus, the differences in the biogeochemistry among pollutants should be accounted for in modeling approaches for river exports. Here an integration of the existing modeling approaches for individual pollutants is needed. In fact, several attempts have been made recently to model individual pollutants: export of pathogens to surface waters globally (Hofstra 2011; Hofstra et al. 2013; Vermeulen et al.

2015a), and export of micro-plastics (Siegfried et al. 2016) and chemicals (van Wijnen et al. in preparation) by rivers to coastal waters continentally. Approaches of the *MARINA* model can be used here to account for nutrients.

- Integrating water quality with water quantity

In addition to the integration of other pollutants in water quality assessments, water quantity issues (e.g., the demand for clean water) need to be accounted for. Over two-thirds of the world population lives in areas with water security problems (Vörösmarty et al. 2010). This holds in particular for densely populated areas with intensive human activities. These are, for example, large part of north-east China, India, North America and Europe. Water security is associated not only with the availability of water for human needs, but also with the quality of water taken to fulfill those needs. Multiple stressors such as agriculture and urbanization influence the quality of water (Seitzinger et al. 2010; Vörösmarty et al. 2010) whilst climate change influences the availability of water (Sherwood & Fu 2014). Integrating more research fields of water quality with quantity may facilitate the development of pathways for multiple stressors: mitigation pathways for effects of human activities and adaptation pathways for effects of climate change. These pathways may help to solve water security problems and thus to meet water- and society-related SDGs (Griggs et al. 2013). Involving stakeholders like policy-makers, users of water resources and polluters in research activities can increase the effectiveness of implementing pathways to meet SDGs. This can be done via participatory approaches. Scientists can advise different stakeholders on solutions from scenario studies. My PhD thesis can serve as an example of scenario analyses for China.

- This PhD thesis for impact

China develops very quickly in many aspects as illustrated by the fast industrialization of animal production and urbanization. Environmental concerns have also been growing fast. This is shown in the development of environmental policies since the 2000s. My PhD thesis is relevant here. It adds new insights on how to manage effectively water pollution in China. For this, I provide a comprehensive tool: the *MARINA* model. With this model we now better understand the causes of water pollution by nutrients and the impacts of upstream activities on downstream coastal water pollution. This new information is useful to develop effective solutions. In my optimistic scenarios, I showed that it is technically possible to have clean water by 2050. This is possible when most animal manure is recycled and wastewater treatment is based on advanced technologies.

These scientific insights from my PhD thesis can help policy-makers to prioritize their focus when developing environmental policies. Effective environmental policies will likely bring changes to society and current practices of people. Support is thus needed to help change these current practices. Policy support may facilitate manure application on land. Advisory services are highly recommended for knowledge exchange and communication. Hopefully, all this can help to ensure that people in China will have enough clean water for their current and future generations. I hope that my PhD research may serve as an example for other regions where water pollution control is needed for the future availability of clean water.

"*Clean, accessible water for all is an essential part of the world we want to live in.*"
Sustainable Development Goals, 2015

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Supplementary materials

Supplementary materials contain additional information to the following chapters of the PhD thesis:

- Chapter 3 (published as Strokal et al. 2015);
- Chapter 4 (published as Strokal et al. 2016b);
- Chapter 5 (published as Strokal et al. 2016a);
- Chapter 6 (is under revision as Strokal et al. under revision).

The text, figures and tables of the supplementary materials from published articles and the article under revision have been adjusted to the PhD thesis format (e.g., the numbering, formatting). This includes editorial changes for the consistency of presentation in the PhD thesis. The adjusted PhD thesis version of the supplementary materials is available on request (maryna.strokal@wur.nl). The published versions of the supplementary manterials are available online with the published articles.

Summary

Clean water is essential to support our daily needs and to maintain our and ecosystems' health. However, today the availability of clean water is threatened by nutrient pollution. Nutrients like nitrogen (N) and phosphorus (P) are present at increasing levels in many rivers worldwide. Rivers transport these nutrients to coastal waters and this can cause eutrophication and consequent symptoms like blooms of harmful algae and oxygen depletion. These environmental problems can damage living organisms in anoxic conditions and threaten human health through toxic algae.

China has experienced such environmental problems over past several decades. This is a result of the rapidly developing economy and population growth. These drivers have intensified crop and animal production and expanded urbanization. However, the relative importance of these human activities to river export of nutrients to Chinese seas is not well studied. There are two important issues that need further investigation. First, the relative importance of upstream pollution on downstream impacts is not well known. This is needed especially for rivers with large drainage areas as they supply large quantities of nutrients to Chinese coastal waters. Second, there are sources of nutrients in rivers that are typical for China, but are generally not well studied. These are point sources of N and P from animal production and uncollected human waste.

Models are tools that help assess causes and effects of river export of nutrients over time and explore solutions. Global *NEWS*-2 (Nutrient Export from WaterSheds) is an integrated, multi-element model used to quantify river export of different nutrients in different forms and the Indicator for Coastal Eutrophication Potential (ICEP) for over 6000 world rivers. The model quantifies the source attribution by accounting for diffuse and point sources of nutrients in rivers. Global *NEWS*-2 does all this for the past and future. Future trends are based on scenarios of the Millennium Ecosystem Assessment (MEA). However, the model underrepresents the abovementioned anthropogenic sources that are typical for China. This may underestimate nutrient pollution of Chinese rivers. Moreover, it does not account for sub-basin processes.

My PhD thesis, therefore, aims to *better understand trends in river export of nutrients to the coastal waters of China by source from sub-basins, and the associated coastal eutrophication*. To this end, I developed the *MARINA* model for China using Global *NEWS*-2 as a starting point. *MARINA* is short for Model to Assess River Inputs of Nutrients to seAs. I formulated five sub-objectives to achieve the main objective. These sub-objectives are:

1. To analyze the original Global *NEWS*-2 model for river export of nutrients and the associated coastal eutrophication (Chapter 2);
2. To develop a sub-basin scale modeling approach to account for impacts of upstream human activities on downstream water pollution, taking the Pearl River as an example (Chapter 3);
3. To quantify the relative share of manure point sources to nutrient inputs to rivers at the sub-basin scale (Chapter 4);
4. To quantify the relative share of sources to river export of nutrients at the sub-basin scale (Chapter 5);
5. To explore optimistic futures to reduce river export of nutrients and coastal eutrophication in China (Chapter 6).

I started my research from Global *NEWS*-2 analyses for sixteen Chinese rivers draining into the Bohai Gulf, Yellow Sea and South China Sea (Chapter 2). Then, I developed a sub-basin modeling approach with updated information for reservoirs and implemented it to Global *NEWS*-2 (Chapter 3). I applied this sub-basin model to the Pearl River (Zhujiang) to quantify dissolved inorganic N (DIN) and P (DIP) export to the South China Sea. Next, I included manure point sources to the sub-basin model, but extended it to the six main rivers of China (Chapter 4). These rivers include the Yangtze (Changjiang), Yellow (Huanghe), Pearl, Huai, Hai and Liao covering 40% of China whith most densely populated areas and intensive economic activities.

Then, I developed the *MARINA* model (Chapter 5). It quantifies river export of nutrients from different sources and sub-basins, and the associated coastal eutrophication for the past and future (1970-2050) (Chapter 5). *MARINA* is applied to the six abovementioned main Chinese rivers. It integrates the new model approaches of Chapters 3 and 4. In addition, it includes (1) uncollected human waste from urban and rural population, (2) an improved modeling of nutrient retentions and losses during river transport, (3) dissolved inorganic (DIN, DIP) and organic (DON, DOP) N and P forms and (4) the ICEP indicator. In the *MARINA* model, the drainage areas of the large Yangtze, Yellow and Pearl rivers are divided into up-, middle- and downstream sub-basins. The principle of the sub-basin approach of *MARINA* is that nutrients from human activities are transported by tributaries to outlets of sub-basins and then to the river mouth (coastal waters) through the main channel. The model takes into account nutrients that are partly lost or retained during transport towards the river mouth. Finally, I performed a scenario analysis using the *MARINA* model (Chapter 6).

I formulated the main conclusions of my PhD thesis in terms of five main lessons for nutrient modeling, and six main findings for China from *MARINA* results.

The five lessons from the PhD thesis for nutrient modeling are:

Lesson 1: The *MARINA* model can help better understand causes of water pollution and explore solutions;
Lesson 2: Sub-basin scale modeling is useful support for effective nutrient management in large basins;
Lesson 3: Preferred sizes of sub-basins depend on research objectives and resource availability;
Lesson 4: Basin scale approaches can serve as a basis for sub-basin scale modeling of nutrient fluxes;
Lesson 5: Building trust in nutrient models is more than just comparing modeled and measured data.

The new *MARINA* model provides insights into trends, causes and effects of water pollution by nutrients and opens up the opportunity to discuss the effects of solutions (Lessons 1 and 2). A novel, sub-basin modeling approach allows to quantify the relative importance of upstream human activities on downstream water quality to support effective policies for large basins. The delineation of the sub-basins was determined taking into account the research objective and the availability of resources such as time and data for model inputs (Lesson 3). In developing the sub-basin *MARINA* model, I used basin scale approaches of Global *NEWS*-2 with further improvements for China. Evaluation results of the *MARINA* model justify this choice (Lesson 4). This evaluation included several options to build trust in the model. Comparision of modeled results with observations is only one of them (Lesson 5). Other options are sensitivity analyses, comparison of model inputs with other independent datasets, comparison of model results with other modeling studies, and expert knowledge.

The main six findings of the *MARINA* results for China are:

Finding 1: Dissolved N and P export by Chinese rivers increased by a factor of 2-8 between 1970 and 2000;
Finding 2: The potential for coastal eutrophication was low in 1970 and high in 2000 in China;
Finding 3: Most dissolved N and P in Chinese seas is from middlestream and downstream human activities;
Finding 4: Manure point sources are responsible for 20-80% of dissolved N and P in Chinese rivers;

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Finding 5: In the future, river export of nutrients may increase, and current policy plans are not sufficient to avoid this increase;
Finding 6: In optimistic scenarios, the potential for coastal eutrophication is low in 2050, mainly as a result of assumed full implementation of: (1) high recycling rates of animal manure, and (2) high efficiencies of nutrient removal in sewage systems.

The *MARINA* results indicate that Chinese rivers had much more nutrients in 2000 than in 1970. As a result, the Bohai Gulf received a factor of 6-8 more dissolved N and P in 2000 compared to 1970. For the Yellow Sea this increase is a factor of 2-5 and for the South China Sea, a factor of 2-4 (the ranges are for nutrient forms). This explains why the potential for coastal eutrophication was higher in 2000 than in 1970 (Findings 1 and 2). Most of dissolved N and P was exported from middle- and downstream human activities (Finding 3). An important polluter is animal production directly discharging manure to Chinese rivers and thus causing coastal eutrophication (Finding 4). These direct manure discharges are a result of inadequate manure management in combination with industrialization trends in animal production. These industrialization trends have disconnected crop from animal production. This explains partly why farmers have been using considerable amounts of synthetic fertilizers while often ignoring animal manure. Another important polluter is urbanization activities discharging sewage from wastewater treatment facilities. This pollutes urban aquatic systems in particular with P when treatment is ineffective.

By 2050 China may continue experiencing eutrophication problems. This is according to the Global Orchestration (GO) scenario of the Millennium Ecosystem Assessment where effective environmental policies are not assumed (Lesson 5). GO was used for the baseline scenario here. In this baseline scenario, Chinese rivers are projected to export more nutrients than in 2000 as a result of poor manure management in industrial animal farms and ineffective wastewater treatment. Therefore, the potential for coastal eutrophication remains high. This high potential is only somewhat lower in the scenario, incorporating the "Zero Growth in Synthetic Fertilizers after 2020" current policy plans (the CP scenario). These policy plans were recently adopted by the Chinese government and aim at recycling 60% of manure on land and zero growth in use of synthetic fertilizers after 2020. These policy plans are a good start, but they alone may not be enough to reduce coastal eutrophication.

In contrast, my optimistic scenarios (OPT-1 and OPT-2) show that the potential for coastal eutrophication can be low in 2050 (Finding 6). This is a result of two main strategies to avoid river pollution. The first strategy is for animal production to recycle almost all available manure. In this way direct discharges of animal manure to rivers are avoided. This consequently reduces the dominant source of river pollution and thus largely improves water quality. The second strategy is for human waste that is assumed to be collected from the whole population. This strategy accounts for the implementation of the latest advanced technologies to remove almost all nutrients during treatment in sewage systems. This strategy is especially important for urban rivers.

My PhD thesis reveals novel insights for effective environmental policies in China. It shows the importance of manure point sources in water pollution by nutrients. Clearly, managing this source will likely reduce coastal eutrophication in the future. Furthermore, the implementation of advanced technologies is essential when dealing with urban pollution. Hopefully, my PhD thesis is also useful for other world regions that have similar environmental problems to China. The new, sub-basin scale *MARINA* model is rather transparent and thus can be applied to other large, data-poor basins that may benefit from the allocation of effective management options. With this I hope to contribute to increase the future availability of clean water for the current and next generations, not only in China, but also in other world regions.

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Furthermore, I would like to thank the Global *NEWS* – China team, especially to Mengru (who is one of my paranymphs) and Ang. I enjoyed very much to be part of this team. We had wonderful and cheerful trips in China, which I will never forget! I am very grateful for your support throughout of my research and for taking care of me in China. I am happy that this team is growing. Chen Xi will join the team soon. Hopefully, there will be many more people to come.

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Environmental Assessment Agency, PBL) and abroad (e.g., China, Ukraine). I benefited from this collaboration in many ways. For example, I improved the quality of my research and extended my network largely. I thank all who contributed to this! I would like to thank people from the Environmental Technology group of Wageningen University & Research for inviting me to participate in the trip to China. I will never forget that trip! I also would like to acknowledge the Water Systems and Global Change group for welcoming me in their group!

I was also involved in (co)supervision of many MSc students during my PhD period. My students were my inspiration. I enjoyed working with them. I thank all of them for their hard work!

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My PhD research would have never been successful without support of my family: Olga (my mother), Petro (my father) and Vita (my twin sister). I dedicate my PhD thesis to them. All my words will not be enough to express my appreciation for your support during my PhD period and for your understanding about my choices to study and work abroad! You always encouraged me and told me “everything will be fine and you will manage” when I had difficult times. I realize how difficult it is for you to communicate with me over two thousands kilometers. We see each other twice per year on average. Nevertheless, I feel your support and love every day! Thank you for this!

Now I am happy to say:

My PhD thesis is finished and new challenges are starting!

About the author

Maryna Strokal was born on 7 December 1984 in Brovary town in Ukraine. She was born about half an hour later after her twin sister, Vita Strokal. After high school Maryna studied Applied Ecology at the college of Ukraine for four years (2000 - 2004). Next, she studied Environmental Sciences at the National University of Life and Environmental Science of Ukraine (NuBiP). In 2007, she received a BSc degree and in 2008 an MSc degree (both with distinction, Cum Laude) in Ecology and Environmental Protection. During her study Maryna focused on chemical contamination of water systems in rural areas of Ukraine. For this she did a lot of experimental work. In 2009, she started an MSc program at Wageningen University & Research, The Netherlands. In 2011, she received an MSc degree (with distinction, Cum Laude) in Environmental Systems Analysis (ESA). After her graduation, Maryna worked as a teaching assistant at Wageningen University & Research for a few months, co-organized the Sixth International Symposium on Non-CO_2 Greenhouse Gases (NCGG-6) and collaborated with the NuBiP on water quality in the Black Sea. In October 2012, she received a PhD position at the ESA group of Wageningen University & Research. Her PhD thesis is about modeling water pollution in China as affected by human activities. She focuses on nutrient pollution that causes coastal eutrophication problems. For this she developed an integrated *MARINA* model, in short for Model to Assess River Inputs of Nutrient to seAs. This model aims at identifying causes of nutrient pollution and exploring solutions to reduce this pollution in a spatially explcit way. During the PhD period Maryna was active in education and supervision activities. Maryna contributed largely to extending the network with China on nutrient management. She also attended various international conferences and symposia. Now, Maryna is looking forward to new challenges in her scientific career.

Awards

- In 2009, Maryna received a NUFFIC scholarship for an MSc study at Wageningen University & Research in The Netherlands;
- In 2012, Maryna received a WIMEK fellowship to conduct a joint research between the NuBiP and Wageningen University & Research for six months;
- In 2015, Maryna was awarded by the Storm-van der Chijs Fund for most promising female PhD candidates of Wageningen University & Research.

List of publications

Published peer-reviewed articles

2016

Strokal M, Kroeze C, Wang M, Bai Z, Ma L (2016a) The *MARINA* model (Model to Assess River Inputs of Nutrients to seAs): model description and results for China. Science of the Total Environment 562: 869-888*

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Strokal MP, Kroeze C, Kopilevych VA, Voytenko LV (2014c) Reducing future nutrient inputs to the Black Sea. Science of the Total Environment 466–467(0): 253-264*

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Sattar MA, Kroeze C, **Strokal M** (2014) The increasing impact of food production on nutrient export by rivers to the Bay of Bengal 1970–2050. Marine Pollution Bulletin 80: 168-178**

Suwarno D, Löhr A, Kroeze C, Widianarko B, **Strokal M** (2014) The effects of dams in rivers on N and P export to the coastal waters in Indonesia in the future. Sustainability of Water Quality and Ecology 3-4: 55-66**

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Kroeze C, Hofstra N, Ivens W, Löhr A, **Strokal M**, van Wijnen J (2013) The links between global carbon, water and nutrient cycles in an urbanizing world—the case of

coastal eutrophication. Current Opinion in Environmental Sustainability 5(6): 566-572**

Published reports

Strokal M, de Vries W (2012) Dynamic modeling of phosphorus export at river basin scale based on Global NEWS. In. Alterra Wageningen UR, Report 2321, Wageningen, the Netherlands. p 100*

Conference proceedings

Strokal M, Kroeze C, Li L, Luan S, Wang H, Yang S, Zhang Y (2014a) Modeling nitrogen and phosphorus export by the Pearl River in China 1970–2050. In: Proceedings of the 2014 12th International Conference on Modeling, Monitoring and Management of Water Pollution. WIT Press, UK, Algarve, Portugal*

Strokal M, Kroeze C, Wang M, Ma L (accepted) Increasing nitrogen use efficiency in agriculture reduces future coastal water pollution in China. In: Proceedings of the 2016 7th International Nitrogen Initiative Conference, "Solutions to improve nitrogen use efficiency for the world". Melbourne, Australia*

In preparation or submitted articles

Strokal M, Kroeze C, Wang M, Ma L (under revision) Reducing future coastal water pollution in China in optimistic scenarios. This manuscript is under revision after positive review comments. It will be resubmitted soon to the Science of the Total Environment*

Nurul MA, Kroeze C, **Strokal M** (under review) Human Waste: An Underestimated Source of Nutrient Pollution in Coastal Seas of Bangladesh, India and Pakistan. Marine Pollution Bulletin**

Wang M, Ma L, **Strokal M**, Chu Y, Kroeze C (under review) Exploring Nutrient Management Options to Increase Nutrient Use Efficiencies in the Food Chain of China. Agricultural Systems**

Wang M, Ma L, **Strokal M**, Ma W, Liu X, Liu Z, Kroeze C (in preparation) Low nutrient use efficiencies in Chinese agriculture call for region-specific nutrient management. To be submitted to Nutrient Cycling in Agroecosystems**

Li A, **Strokal M**, Bai Z, Kroeze C, Ma L, Zhang F (in preparation) ‘Double High Agriculture’ reduce nutrient export by rivers in China. To be submitted to Regional Environmental Change**

*My contribution to the articles of which I am the first author

I am responsible for the research. In most of the articles, I focused on global and regional modeling of nutrient pollution in aquatic systems, the causes and effects of this pollution on coastal waters. In one article, I analyzed global and continental N_2O emissions from human waste. All publications present new approaches for analyzing nutrient export by rivers. I developed new models for world (Strokal & de Vries 2012) and Chinese (Strokal et al. 2015; Strokal et al. 2016a; Strokal et al. 2016b) rivers. Several articles present new alternative scenarios to reduce water pollution in the Black Sea (Strokal et al. 2014c) and Chinese seas (Strokal et al. under revision). I took the lead in developing and applying the new modeling approaches and scenarios. My work for nutrient export analyses was supervised by prof dr W de Vries for the world rivers (Strokal & de Vries 2012), prof dr C Kroeze for the Black Sea rivers (Strokal & Kroeze 2013; Strokal et al. 2014c) and by prof dr C Kroeze, prof dr L Ma and prof dr S Luan for the Chinese rivers (Strokal et al. 2014a; Strokal et al. 2015; Strokal et al. 2016a; Strokal et al. accepted; Strokal et al. under revision; Strokal et al. 2016b; Strokal et al. 2014b). The other co-authors advised on the underpinning of the studied areas and gave expert opinion on modeled water pollution problems. They also provided local information for developing models and assisted with interpreting and analyzing the results. All co-authors read and commented on the text.

**My contribution to the articles that I co-authored

I supervised the work for the Bay of Bengal (Nurul et al. under review; Sattar et al. 2014) and China (Li et al. in preparation). In particular, I assisted modeling of nutrient flows from land to sea. I also assisted modeling the effects of dams on nutrient export by Indonesian rivers (Suwarno et al. 2014b). I helped with the structure and advised on the graphics (all articles). I also contributed to interpreting the results and finalizing them for publication (all articles).

Netherlands Research School for the
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D I P L O M A

For specialised PhD training

The Netherlands Research School for the
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(SENSE) declares that

Maryna Strokal

born on 7 December 1984 in Brovary town, Ukraine

has successfully fulfilled all requirements of the
Educational Programme of SENSE.

Wageningen, 13 December 2016

the Chairman of the SENSE board	the SENSE Director of Education
Prof. dr. Huub Rijnaarts	Dr. Ad van Dommelen

The SENSE Research School has been accredited by the Royal Netherlands Academy of Arts and Sciences (KNAW)

KONINKLIJKE NEDERLANDSE
AKADEMIE VAN WETENSCHAPPEN

The SENSE Research School declares that **Ms Maryna Strokal** has successfully fulfilled all requirements of the Educational PhD Programme of SENSE with a work load of 53.5 EC, including the following activities:

SENSE PhD Courses

- Complex dynamics in human-environment systems (2013)
- Scales and dimensions of sustainability: A system perspective (2013)
- Governance, sustainable development and climate change in China (2013)
- Environmental research in context (2013)
- Research in context activity: 'Organising an international workshop on 'Pollution management in China: nutrient export from land to sea', Wageningen' (2013)
- Pitch training (2014)

Other PhD and Advanced MSc Courses

- Information literacy PhD including introduction to EndNote, Wageningen University (2012)
- i-GIS*:* A practical post-graduate GIS course, Wageningen University (2013)
- Greencycles II training workshop, Max Planck Institute for Biogeochemistry (2013)
- Programming in Python, Wageningen University (2013)
- Techniques for writing and presenting a scientific paper, Wageningen University (2013)
- Reviewing a scientific paper, Wageningen University (2013)
- Data management planning, Wageningen University (2015)
- Introduction to R for statistical analysis, Wageningen University (2015)
- Teaching and supervising thesis students, Wageningen University (2016)

External training at a foreign research institute

- Field trips to rural and urban areas, Peking University and Chinese Academy of Sciences (2014-2016)

Management and Didactic Skills Training

- Co-organising the International Non-CO_2 Greenhouse Gas Emissions (NCGG6) conference, 2-4 November 2011, Amsterdam, The Netherlands
- Supervising ten MSc students (2012-2016)
- Organising the Global NEWS-2 (Global Nutrient Export from WaterSheds 2) model training for MSc students and staff (2012-2016)
- Guest lectures for the Open University (Utrecht, The Netherlands), the National University of Life and Environmental Sciences of Ukraine (Kyiv, Ukraine), the Peking University (Shenzhen, China) and the Chinese Academy of Sciences (Shijiazhuang, China) (2013-2016).
- Supervising in the MSc course 'Introduction to global change' (2014-2015)

Selection of Oral Presentations

- *Modelling nitrogen and phosphorus export by the Pearl River in China 1970-2050.* 12th International Conference on Modelling, Monitoring and Management of Water Pollution, 26-28 May 2014, The Algarve, Portugal
- *Sub-basin scale modelling of nutrient export by rivers to coastal waters of China.* International Interdisciplinary Conference on Land Use and Water Quality: Agricultural Production and the Environment, 21-24 September 2015, Vienna, Austria

SENSE Coordinator PhD Education

Dr. ing. Monique Gulickx

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