

Invitation

You are kindly invited to attend the public defence of my PhD thesis, entitled:

Nutrients and Pesticides in Chinese Waters:

Future Pollution and Solutions

on Tuesday 23 March 2021 at 11.00, in the Aula of Wageningen University & Research, Generaal Foulkesweg 1, Wageningen

Following the defence you are welcome to join the reception

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Propositions

- Global socio-economic and climatic change will increase water pollution by food production in China. (This thesis)
- Nutrient pollution in waters can be reduced to sustainable levels without full-scale implementation of available technologies.
 (This thesis)
- 3. The first step to overcome a crisis, is to develop a vision of our desired future.
- 4. To move the environmental modeling community as a whole forward, natural and social scientists should collaborate and communicate more.
- 5. Healthier diets are needed to meet SDG6 (clean water for all).
- 6. The COVID-19 crisis also is a PhD crisis.

Propositions belong to the thesis, entitled

Nutrients and Pesticides in Chinese waters: future pollution and solutions

Ang Li

Wageningen, 23 March 2021

Nutrients and Pesticides in

Chinese Waters:

Future Pollution and Solutions

Ang Li

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This research was conducted under the auspices of the Graduate School for Socio-Economic and Natural Sciences of the Environment

Nutrients and Pesticides in

Chinese Waters:

Future Pollution and Solutions

Ang Li

Thesis

Submitted in fulfilment of the requirements for the degree of doctor at Wageningen University
by the authority of the Rector Magnificus,
Prof. Dr A.P.J. Mol,
in the presence of the
Thesis Committee appointed by the Academic Board
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Chapters 3 and 5 have been published as peer-reviewed scientific articles. Chapter 4 will be resubmitted after revision based on positive reviews. Chapter 2 will be submitted for publication soon. The text, figures, and tables of the published and submitted articles have been adjusted to the PhD thesis format. Editorial changes were made for reasons of uniformity of presentation. Reference should be made to the original articles.

Chapter 1

General introduction

1.1 Background

Feeding the global population in a sustainable way is a grand challenge (Gerten et al. 2020). The population has been increasing relatively rapidly worldwide since the 1960s worldwide (Sutton et al. 2013). Consequently, the demand for food has increased. This is also true for China, where food production has been increasing since the Green Revolution around the 1960s. As a result, the use of agrochemicals has also increased (Wu et al. 2018). During the 1990s and 2000s, China consumed almost 47% of the global pesticides (Miao 2019) and about one-third of synthetic N fertilizers (Gao et al. 2006) on 22% of the agricultural land to produce sufficient food for the Chinese population (Wu et al. 2018). Increasing amounts of nutrients and pesticides entered the environment (Erisman et al. 2013; Liu et al. 2012). Particularly, large amounts of nitrogen (N), phosphorus (P) and pesticides are lost to surface waters and negatively affect aquatic ecosystems (Grung et al. 2015; Jin et al. 2019; Le et al. 2010; Liu et al. 2013; Yan et al. 2010). China is one of the countries facing this problem (Ma et al. 2020). Increasing number of algal blooms has been observed in Chinese waters in response to excess nutrients (SOA 2010). Toxic pesticides have been detected in drinking water sources, rivers and lakes in China (Grung et al. 2015; Huang et al. 2018; Jin et al. 2019).

1.1.1 Sources of nutrients and pesticides in Chinese waters

Nutrients enter waters from point and diffuse sources (Galloway et al. 2004). In China, major point sources of N and P for waters include the direct discharge of animal manure to surface water, sewage effluents, and uncollected human waste (Wang et al. 2018a). Diffuse sources of N in surface waters are synthetic fertilizers, animal and human excretion on land, biological N fixation, atmospheric N deposition, and weathering of P-containing minerals (Galloway et al. 2008). Because of the poor air quality, direct deposition of N on surface waters is also an important source (Liu et al. 2011b). Part of

the N and P entering rivers is retained in sediments. The remainder of the N and P is further transported to coastal seas by rivers (Dumont et al. 2005).

Pesticides used in agriculture typically reach the targeted insects or weed to a small extent (Wauchope 1978). This is because of the widely used application methods (Matthews 2008; Wauchope 1978). Consequently, large amounts of pesticides are dispersed through the air, bounded with organic carbon in the soil, or deposited in nearby surface waters (Beketov et al. 2013). Surface runoff after heavy rainfall transports some of the pesticides from soils to the surrounding surface waters (Lewan et al. 2009). In urban regions, pesticide runoff is collected by the sewage system and reaches the surface water as point sources (Wittmer et al. 2010). In rural areas, pesticides also enter the sewage system when farms wash equipment for pesticide application. The above mentioned pathways lead to large pesticide losses and losses through surface runoff after rainfall represent the major pathway (Berkowitz et al. 2014; Huang et al. 2018; Wauchope 1978). Without appropriate pollution control strategies, the agricultural sectors with intensive use of pesticides and nutrients will continue to negatively impact Chinese waters.

1.1.2 Modelling water pollution caused by nutrients and pesticides

Models are useful tools for improving understanding of water pollution problems. During the 1990s, large-scale water quality models for nutrients were developed (Leon et al. 2001; Reckhow et al. 1990; Seitzinger and Kroeze 1998; Srinivasan and Arnold 1994). Today, several spatially explicit models are available to quantify nutrient flows from land to Chinese waters (Gu et al. 2012; Ma et al. 2010; Strokal et al. 2016a; Wang et al. 2016). Strokal et al. (2016a) developed the Model to Assess the River Inputs of Nutrient to seAs (MARINA) 1.0 to quantify the river export of nutrients from land to seas for Chinese rivers at a sub-basin scale for 1970, 2000 and 2050. This model considered human activities on land and nutrient retention in soil and waters. Future scenarios are based on the Millennium Ecosystem Assessment (MEA) scenarios (Alcamo et al. 2005). Several versions of the MARINA have been developed in recent years (Chen et al. 2020; Chen et al. 2019; Strokal et al. 2016a; Wang et al. 2020). The updated version, MARINA 1.1 was developed to account for seasonality of nutrient losses to rivers from sub-basins (Chen et al. 2020). This version has been applied to the Yangtze basin and to 2012. Wang

et al. (2020) developed the MARINA 2.0 version by adding new sources of nutrients in rural areas, improving model inputs with local information, and updating global change scenarios for future trends in China. The global change scenarios are based on the recently published storylines of Shared Socio-economic Pathways (SSPs) and Representative Concentration Pathways (RCPs). MARINA 3.0 uses a multiscale approach to quantify the dissolved N and P inputs to rivers on grid, sub-basin and country scales, and this version is for 2012 (Chen et al. 2019). Additionally, a MARINA-Lakes model has been developed that quantifies nutrient losses to Chinese lakes (Li et al. 2019b; Wang et al. 2019; Yang et al. 2019).

Some other large-scale nutrient models include China. For example, Global NEWS-2 (Global Nutrient Export from Watersheds) (Mayorga et al. 2010) and IMAGE-GNM (Integrated Model to Assess the Global Environment-Global Nutrient Model) (Beusen et al. 2015; Liu et al. 2018). Moreover, some nutrient models such as SWAT (Soil and Water Assessment Tool) are applied to small Chinese rivers (Yang et al. 2008). Large-scale models are widely used to better understand the causality and sources of nutrient pollution in Chinese waters.

The MARINA model family provides valuable insights into nutrient pollution in Chinese waters (Chen et al. 2020; Strokal et al. 2016a; Wang et al. 2020; Wang et al. 2019; Yang et al. 2019). Strokal et al. (2016a) indicate that the total dissolved N (TDN) and total dissolved P (TDP) export by six large rivers to Chinese seas increased by twofold and eightfold from 1970 to 2000, respectively. According to the results of MARINA 2.0, in 2012, 18553 kton of TDN and 2143 kton of TDP were lost to six large rivers in China (Wang 2020). Point sources including the direct discharge of animal manure and discharge of human waste account for one-third of the TDN and 90% of the TDP inputs to rivers (Wang 2020). Diffuse sources, including synthetic fertilizers, animal manure and human waste, are responsible for one-third of TDN to coastal seas. Under global change, river export of nutrients is projected to increase in the future (Strokal et al. 2017; Wang 2020).

Conversely, modelling studies on pesticide pollution for all of China are rare. Some studies have reported on pesticides in Chinese waters (Grung et al. 2015; Zhang et al. 2019; Zheng et al. 2016). However, large-scale modelling of pesticide flows is not as extensively conducted as for nutrients. During recent decades, interest in using models

to better understand the distribution of pesticide pollution in Chinese waters has increased. Consequently, various pesticide models are available; however, these are mostly used to quantify single types of pesticide losses in small watersheds (Liu et al. 2017; Zhang et al. 2020; Zheng et al. 2016). Few studies use integrated approaches to analyze pesticide pollution for China as a whole. Ouyang et al. (2017) used an empirical based approach to estimate the pesticide losses to Chinese waters by crop type from 1990 to 2011. This approach used pesticide losses coefficients based on farm field experiments in America and Norway. Sun et al. (2019) used a pesticide emission model to estimate pesticide losses from agricultural land to Chinese waters as diffuse sources for 2004, 2008 and 2013. This approach strongly depended on emission factors for pesticides and was based on monitoring plots in only 232 monitoring sites in China. The pesticide losses estimated by these two approaches differed largely. The pesticide losses to waters estimated by Ouyang et al. (2016) were 10 times larger than those estimated by Sun et al. (2019). This illustrates the uncertainties in estimates, and the need for improved estimation of pesticide pollution.

For Europe, some other modelling approaches were used to estimate pesticide losses to waters (OECD 1998). A runoff-based model was applied in Europe (Kattwinkel et al. 2011; Schriever and Liess 2007; Schriever et al. 2007). Schriever and Liess (2007) describe how their pesticide model estimates the potential for pesticide runoff rather than actual flows. Their approach provides the temporal and spatially explicit potential for pesticide pollution in waters, with relatively simple model inputs. Therefore, it is suitable for modelling water pollution by pesticides for data-poor regions on larger scales. Hence, the potential for pesticide runoff can be used to identify potential hotspots of pesticide pollution. Based on this approach, Ippolito et al. (2015) developed a global model to estimate potential insecticide losses into waters through surface runoff in response to single heavy events. This study mapped the potential for agricultural insecticide runoff to surface waters worldwide, including China. However, based on a sensitivity analysis (Ippolito et al. 2015), they concluded that the application rate caused high uncertainty in the model outputs. For China, they used one country-based insecticide application rate in their model. Therefore, the estimates for pesticide pollution for China can potentially be improved. Additionally, because the data demand for this model is low, scenario analyses can be applied to the model. Using scenarios

analyses can improve understanding of the impact of global change on pesticide pollution. This type of study is lacking in China.

Food production is a large contributor to both nutrient and pesticide pollution in Chinese waters. However, other water pollutants exist. Moreover, under global change, losses of all pollutants associated with food production may change (Savary et al. 2014). These pollutants are interrelated, because they may have common sources or affect the same sectors. Moreover, they may interact in the environment. However, current modelling studies often analyze water quality for nutrients and pesticides separately for China, especially for large-scale water quality assessment. For air pollution problems, models are available that adopt a multi-pollutant perspective (Amann et al. 2004; Wagner et al. 2013). Such integrated models have successfully supported the international negotiation on air pollution control. For water pollution such integrated approaches are scarce. Therefore, a large-scale water quality model taking multi-pollutant perspectives will be the next focus of water quality assessments.

1.1.3 Future trends of water pollution caused by nutrients and pesticides

Global change may strongly impact water pollution in the future (Kanter et al. 2020; Mateo-Sagasta et al. 2018; Mekonnen and Hoekstra 2018). China is projected to experience rapid socio-economic development including rapid population growth and increasing urbanization in the future (Guo et al. 2019; Jones and O'Neill 2016). Therefore, the food demand in China will increase further. Synthetic fertilizer and pesticides are likely to increase in the future to meet the increasing food demand, thus, losses of nutrients and pesticides may also increase. Increasing inputs of nutrients and pesticides to surface waters may worsen water quality in the future. Human activities may accelerate climate change, which could alter the rainfall patterns and temperature (Hempel et al. 2013a). These changes in weather conditions might largely affect land use (Hurtt et al. 2020a), leading to changes in inputs of nutrients and pesticides to waters in China.

To better understand the impact of global change on water pollution in China, many studies applied scenario analyze with water quality models. Scenario-based forecasting is widely used in studies on water pollution. Some studies forecast future trends of

nutrient pollution in China under different scenarios (Ma et al. 2013b; Strokal et al. 2017; Wang et al. 2017). Other studies quantify the river export of nutrients in the future under the MEA scenarios (Alcamo et al. 2005a; Qu and Kroeze 2012; Strokal et al. 2014). Wang et al. (2019) model the impact of global change on river export of nutrients under scenarios based on combinations of SSPs (O'Neill et al. 2014) and RCPs (Van Vuuren et al. 2011). The above-mention studies indicate increasing nutrient pollution in Chinese waters in the future and they all recommend exploring solutions for nutrient pollution in China. To our knowledge, no studies exist on exploring pesticide pollution under global change for China.

Scenario-based back-casting is another method that can be combined with water quality models to better understand the future of water pollution. Back-casting is often used in searching for a pathway to reach a certain desired future and earlier studies applied back-casting on energy future (Gomi et al. 2011; Haslauer 2015; Van Berkel and Verburg 2012; Van Vliet and Kok 2013). Such application in water pollution studies is rare.

1.1.4 Possible strategies to reduce nutrient pollution

Excess nutrients enter surface water as point and diffuse sources. In China, point sources contribute more than diffuse sources to surface waters (Ma et al. 2012; Strokal et al. 2017; Wang 2020). Therefore, strategies to reduce nutrient pollution from point sources might be more efficient. Several studies state that direct discharge of animal manure from industrialized animal farms (point sources) is the most important source of nutrient pollution in Chinese waters (Strokal et al. 2016b; Wang 2020). Management options to reduce nutrient losses to rivers from animal production are suggested in previous studies (Li et al. 2017b; Ma et al. 2013b; Strokal et al. 2017; Wang et al. 2018a). These options include improving nutrient use efficiency (Wang et al. 2018a; Zhang et al. 2015), reducing direct discharge of animal manure (Ma et al. 2013a), recycling animal manure to replace synthetic fertilizer in crop production (Jie et al. 2017; Sun et al. 2012), and improving animal feed to reduce nutrient content in animal manure (Lesschen et al. 2011). Sewage effluent is another important point source of nutrient losses. It is technologically possible to reduce nutrient losses by increasing N and P removal during wastewater treatment (Kartal et al. 2010; Shi et al. 2010; Winkler et al. 2012) and by increasing the population connected to sewage systems in rural areas in China.

Some studies use scenario analyses to quantify the technological potential of the above-mentioned strategies to reduce river pollution from food production (Li et al. 2017b; Strokal et al. 2017; Wang et al. 2018a). Some studies applied integrated models to explore options to close nutrient cycles in agriculture. For example, Strokal et al. (2017) forecast the nutrient pollution in Chinese rivers in the future from an optimistic perspective. This study indicates that nutrient loads in Chinese rivers for 2050 can be reduced to 1970 levels by maximizing manure recycling and improving sewage systems. However, large efforts are needed to implement these technologies. Our knowledge of environmental targets for nutrient pollution is lacking for China. Thus, a better understanding of environmental target and pollution control strategies to reach the target is certainly needed.

1.2 Knowledge gaps and research objectives

Based on the above, I formulated the following knowledge gaps for water pollution from pesticides and nutrients in China:

Knowledge Gap 1: The effects of global change on water pollution caused by pesticides in China are poorly understood.

Knowledge Gap 2: Combined pollution control strategies for nutrients and pesticides to reach future targets for water quality in China have not been studied to a large extent.

Knowledge Gap 3: We have little knowledge of how to develop multi-pollutant modelling approaches to quantify the water pollution caused by multiple pollutants.

Therefore, the main objective of this PhD thesis is, therefore, to explore control strategies to reduce future water pollution in China, caused by nutrients and pesticides.

To this end, four specific research objectives are formulated and form the four research chapters of this thesis:

- 1. To analyse future potential pesticide losses to Chinese waters under global change (related to Knowledge Gap 1). A forecasting scenario approach is applied.
- 2. To explore the possibilities to avoid coastal eutrophication in the North China Plain (related to Knowledge Gap 2). A combination of forecasting and backcasting approaches is applied.

- 3. To quantify desired nutrient pollution levels for sub-basins of the Yellow, Yangtze and Pearl Rivers from both an environmental and equality point of view (related to Knowledge Gap 2). A combination of forecasting, back-casting and optimization approaches is applied.
- 4. To draw lessons from air pollution control for large-scale water quality assessments, where multi-pollutant approaches are more common (related to Knowledge Gap 3).

1.3 Research approaches and thesis outline

This thesis has six chapters (Figure 1.1). Chapter 1 presents the background and provides the information on the knowledge gaps and research objectives. The above-mentioned four sub-objectives correspond to Chapters 2 to 5. The methods and sub-objectives of all chapters are presented in Figure 1.1.

In Chapter 2, I analyzed the potential for pesticide runoff in China under global change (sub-objective 1). To achieve this, I first developed a pesticide model based on an existing global insecticide model and available local information in this chapter. I then developed three scenarios: Sustainability (SSP1-RCP2.6), "Middle of the Road" (SSP2-RCP4.5) and Economy First scenario (SSP5-RCP8.5), using combined SSPs and RCPs. The potential for pesticide runoff was quantified for China at 30 by 30 arcminutes grid for 2000, 2010, 2050 and 2099.

In Chapter 3, I explored the possible combination of pollution control strategies for the North China Plain (NCP, sub-objective 2). To achieve this, I combined the back-casting approach with the MARINA and Indicator for Coastal Eutrophication Potential (ICEP). I developed 54 scenarios based on different combinations of three pollution control strategies. Using back-casting and ICEP, targets for the dissolved N and P exported by the Yellow, Huai and Hai rivers that flow through the NCP were quantified. I then identified scenarios that could reach the targets for the three rivers.

Chapter 4 considers both the environment and equality in pollution control in quantifying the desired level for river export of dissolved N and P for sub-basins of the Yellow, Yangtze and Pearl Rivers (sub-objective 3). To minimize the socio-economic inequality in pollution control, I applied the Gini optimization approach with the ICEP and MARINA. Additionally, I identified possible sources that could help to reduce the

river export of nutrients to the desired level. This is the first study combining the Gini optimization and MARINA and it is also the first study considering equality in pollution control for the Yangtze, Yellow and Pearl Rivers.

In Chapter 5, I drew lessons on modeling water pollution by multi-pollutants for large regions (sub-objective 4). These lessons were learned from air pollution models that calculate multiple pollutant emissions for large regions. In this chapter, a modeling approach for assessing water pollution caused by multiple pollutants is proposed. Hence, I also introduced examples for including multiple targets with water quality models to explore solutions for water pollution.

In Chapter 6, I discussed the main findings and modeling approach and drew the main conclusion of this thesis.

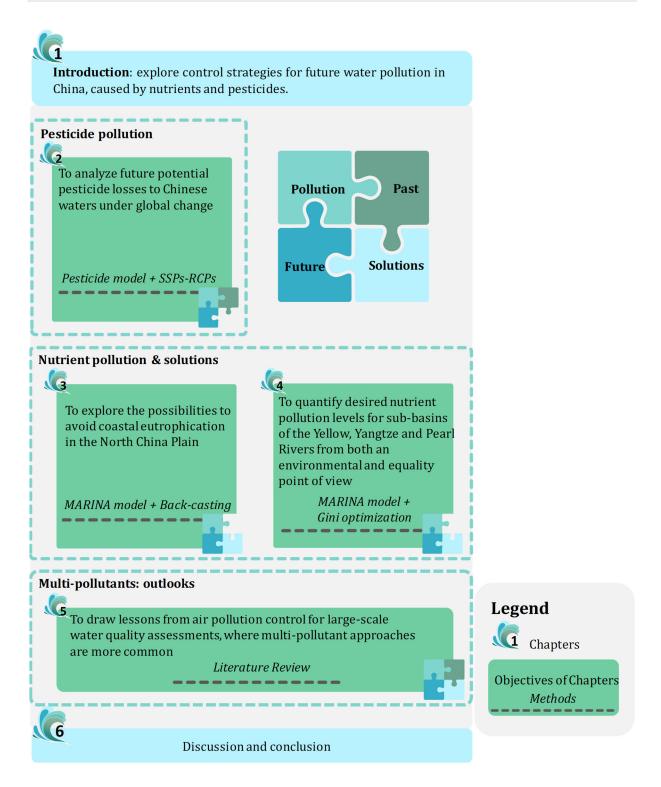


Figure 1.1 The structure of the PhD thesis. Chapters are presented with their research objectives. Main methods that have been used are given for Chapters 2-5. SSPs-RCPs refers to the combination of Shared Socioeconomic Pathways (O'Neill et al. 2014) and Representative Concentration Pathways (Van Vuuren et al. 2011). MARINA is short for the Model to Assess River Inputs of Nutrients to seAs (Strokal et al. 2016a).

Chapter 2

Past and future pesticide losses to Chinese

waters under global change

Abstract

Increasing pesticide use pollutes Chinese surface waters. Pesticides often enter waters through surface runoff from agricultural fields. This occurs especially during heavy rainfall events. Global change may accelerate future loss of pesticides to surface waters as due to increasing food production and climate change. The main objective of this study is to analyze past and future pesticide losses to Chinese waters under global change. To this end, we developed a pesticide model with local information to quantify the potential pesticide runoff from near-stream agriculture to surface waters after heavy rainfall. We project future trends in potential pesticide runoff. For this, we developed three scenarios: Sustainability, "Middle of the Road" and Economy-first. These scenarios are based on combined Shared Socio-economic Pathways and Representative Concentration Pathways. We identified hotspots with high potentials pesticide runoff. The results show that the potential pesticide runoff increased by 45% from 2000 to 2010, nationally. Over 50% of the national pesticide runoff in 2000 was in five provinces. Over 60% of the Chinese population lived in pesticide polluted hotspots in 2000. For the future, trends differ among scenarios and years. The largest increase is projected for the Economy-first scenario, where the potential pesticide runoff is projected to increase by 85% between 2010 and 2099. Future pesticide pollution hotspots are projected to concentrate in the south and south-east of China. In our global change scenarios, 58% to 84% of the population is projected to live in pesticide polluted hotspots from 2050 onwards. These projections can support the development of regional management strategies to control pesticide pollution in waters in the future.

To be submitted:

Ang Li, Carolien Kroeze, Mengru Wang, Lin Ma, Maryna Strokal. Past and future pesticide losses to Chinese waters under global change.

2.1 Introduction

Pesticides are often used in agriculture to protect crops from pests, weeds and fungal disease. China became the largest consumer of pesticides in the 1990 (FAO 1990). In China, the use of pesticides increased since the 1970s to ensure food security. In 1995, the total pesticide use in China reached one million ton and has been increasing since then (NBSC 2018). According to national statistics, 90 million tons of cereals and 78 million tons of vegetables were produced that required pesticides in 2006 (NBSC 2007). However, using large amounts of pesticides in cropland can threaten for the environment and human health (Fang et al. 2017; Hernández et al. 2013; Morrissey et al. 2015; Zhang et al. 2019).

Existing studies indicate that pesticides and their related metabolites have been detected in soil and waters in China (Gao et al. 2009; Huang et al. 2018; Liu et al. 2016). Many pesticides are toxic and are not easily degraded in the environment (Grung et al. 2015; Huang et al. 2018; Yang et al. 2016). Despite regulations controlling the use of pesticides, they are still detected in the environment. For example, dichlorodiphenyltrichloroethane has been banned since 1983 (Tao et al. 2007). Nevertheless, this pesticide has been detected in the Bohai Sea and Taihu Lake in the past decades (Hu et al. 2009; Nakata et al. 2005; Ta et al. 2006). Some studies reported contamination of waters with pesticides at a river basin or national scale in China in the past (Jin et al. 2019; Liu et al. 2016; Liu et al. 2015; Mai et al. 2002; Wauchope 1978). For example, Ouyang et al. (2016) estimated that 4.39×10³ tons of pesticides were lost to the environment nationally in 2011. These losses mainly occurred in the eastern and southern provinces, such as Shandong, Jiangxi and Guangxi. Zheng et al. (2016) reported that 82 types of pesticides in the Jiulong River basin were detected and the concentrations of 14 out of 82 types of pesticides were exceeded 100 ng L⁻¹. Many other studies exist on studies the pesticide contamination in China (Gao et al. 2009; Grung et al. 2015; Wu and Chen 2013; Zhou et al. 2008; Zhou et al. 2006). Therefore, the Chinese government launched the National Agricultural Diffuse Pollution Action Plan in 2015 to prevent water pollution from pesticides (MOA 2015).

Changes in society and the environment may influence future losses of pesticides into waters (Delcour et al. 2015; Kattwinkel et al. 2011). In the coming years, the Chinese population and urbanization are projected to increase (Ding et al. 2018; Guo et al. 2019).

Consequently, the food demand will increase especially in cities. However, the Chinese government prefers not to expand or reduce agricultural land (Zhao et al. 2011). Therefore, current Chinese agricultural practices are intensifying and using many resources (Shen et al. 2013). This also holds for pesticide use. This could increase pesticide pollution in Chinese waters. However, our knowledge about the effects of intensified agricultural practices on pesticide pollution in waters is limited. Additionally, climate conditions affect the amount of pesticides lost to surface waters. Many pesticides are applied to cropland through drifting or spraying (Matthews 2008). After application, pesticides enter the nearby waters through surface runoff generated after heavy rainfall events (Berkowitz et al. 2014; Ritter and Shirmohammadi 2000; Wauchope 1978). Climate change can influence future rainfall patterns affecting pesticide losses into surface waters (Lee et al. 2014; Wang and Chen 2014; Zhou et al. 2014). However, studies focusing on the influence of climate change on pesticide losses into surface waters in China are lacking.

The main objective of this study is to analyze the past and future potential pesticide losses to Chinese waters under global change. To achieve this, we develop a pesticide model based on an existing global insecticide model developed by Ippolito et al. (2015) with local information to quantify the potential pesticide runoff in China in the past. We then developed three scenarios reflecting future food production and climate change in China. These scenarios are based on combined Shared Socio-economic Pathways (SSPs) and Representative Concentration Pathways (RCPs) (O'Neill et al. 2014; Van Vuuren et al. 2011). Finally, we analyze hotspots with high potentials for pesticide runoff (see Section 2.2 for the definition of the pollution hotspots). Our study contributes to a better understanding of future spatial patterns in pesticide pollution in Chinese waters.

2.2 Material and methods

2.2.1 Model description and inputs

We develop a pesticide model based on the existing insecticide model by Ippolito et al. (2015), and used local information to run the model (see Section 2.2). The model is applied to the past and future. For future years, we develop scenarios and incorporate them into the pesticide model for China. The model quantifies the potential for pesticide runoff from the near-stream agriculture to surface waters in 2000, 2010, 2050 and 2099

at 0.5° . These calculated potential pesticide runoff can reflect the upper bound of pesticide losses to surface water in response to the maximum daily rainfall within the year.

We modify the global insecticide model developed by Ippolito et al. (2015) for the Chinese situation. The main equations for quantifying the potential for pesticide runoff (gLOAD) and the generic indicator of the gLOAD (RP) are provided in Figure 2.1. The RP is calculated as the logarithm of gLOAD (Eq.1 in Figure 2.1). gLOAD is calculated as a function of nine major factors that affect pesticide losses to surface waters (Eq.2 in Figure 2.1). These nine factors are the area of the near-stream agricultural land (A, km2), pesticide application rate (D, g km-2), soil carbon content (OC, %), soil texture (T, classified into fine and coarse soil), average slope (s, %), the proportion of agricultural land (p, %), maximum daily precipitation (P, mm), soil organic carbon-water partitioning coefficient of the pesticide (Koc, ml g-1), and plant interception (I, %). Model outputs are gLOAD and RP per grid of 0.5° for China.

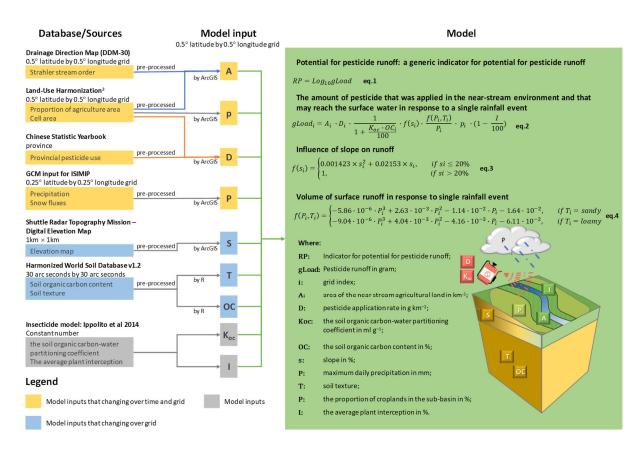


Figure 2.1 Model inputs and model description for the pesticide model. See Table S1 in Appendix I for inputs sources and pre-processing methods.

The model inputs are preprocessed or derived from different existing databases (Figure 2.1 and Table S1 in Appendix I). We calculate the near-stream agricultural land for a 0.5° grid as follows. Based on the drainage direction map (DDM-30), grids are classifies as those with a main channel or tributaries (Döll and Lehner 2002). We assume that grids with a main channel have three segmented streams. Grids with tributaries have two segmented streams. A segmented stream in the grid is assumed to be 1500 m in length. The near-stream area is defined as the area along the stream within 100m. Agricultural land in the near-stream area is defined as near-stream agricultural land. If the near-stream area is less than the agricultural area, the near-stream agricultural land is set at 0.45 km2 for a grid with the main channel and 0.30 km2 for a grid with tributaries. If the near-stream area is larger than the agricultural land area, then the near-stream agricultural area equals the agricultural land in the grid.

The proportion of agricultural land in the grids of 0.5° (p, %) is derived from the Landuse Harmonization2 database (LUH2) (Hurtt et al. 2020b). The agricultural land of the LUH2 database includes 175 different crops, which are aggregated and re-classified into five crop functional types based on their photosynthetic pathways (C3 or C4), lifespan (annuals or perennials) and whether they are a nitrogen fixers. In this study, we focus on cropland as a whole. Therefore, the agriculture land is the sum of five types of cropland in LUH2. The pesticide application rate (D, g km-2) is estimated based on the pesticide use in provinces (g) and the agricultural land area in those provinces (km2). The pesticide use for provinces for 2000 and 2010 is from the Chinese Statistical Yearbook (NBSC 2001, 2011). We allocated the provincial pesticide use to grids of 0.5° using the area-weighted method, which is based on the share of agricultural land area in each grid over the total agricultural land of the province (Figure S1 in Appendix I).

The organic carbon content (OC, %) and soil texture data (T) for grids of 0.5° are derived from the Harmonized World Soil Database (Fischer et al. 2000). The average slope of the 0.5° grid is calculated based on the Shuttle Radar Topography Mission- Digital Elevation Map using ArcGIS (RESDC 2003). The model considers the influence of the slope on pesticide runoff. The influence of the slope on surface runoff is calculated using Eq. 3 in Figure 2.1. We calculate the surface runoff generated after a single rainfall event using the maximum daily precipitation (P, mm). This is calculated as a function of the soil

texture and the precipitation (see Eq. 4 in Figure 2.1 and Ippolito et al. (2015)). Pi in Eq.4 of Figure 2.1 refers to the daily rainfall and snow fluxes.

The maximum daily precipitation (P, mm) is retrieved from the Global Climate Model (GCM) inputs from the Inter-Sectoral Impact Model Intercomparison Project (ISIMIP) project (Hempel et al. 2013b). We calculate the pesticide runoff using precipitation from four different climate models, GFDL-ESM2M, HadGEM2-ES, IPSL-CM5A-LR, MIROC-ESM-CHEM, to account for uncertainties of the modelled precipitation. The pesticide runoff in this study is the averaged gLOAD value over these four GCMs.

The plant interception (I, %) in the planting and growing period is assumed to be 50% based on the approach developed by Ippolito et al. (2015). The plant interception depends on the plant species, growing periods and pesticide application time. However, this information is not available for China. Therefore, we used 50% for each grid as assumed by Ippolito et al. (2015). The soil organic carbon-water partitioning coefficient of the pesticides (Koc, ml g-1) is a substance specific value. However, this information is not readily available for specific pesticide substances for a 0.5° grid in China. We assume to be Koc of 100 following the approach by Ippolito et al. (2015).

We define hotspots for the potential pesticide runoff based on the RP results. We classify the RP values into classes based on the approach developed by Ippolito et al. (2015). We then convert values of gLOAD to a logarithmic scale (RP). The RP results are classified as very low (below -3), low (-3 to -2), medium (-2 to -1), high (-1 to 1), very high I (above 1), very high II (2 to 3), and very high III (above 3). Grids of 0.5° are assigned to these classes. Grids classifies as very high (I, II and III) are considered as pollution hotspots for high potential pesticide runoff to surface waters. We calculate the population in pesticide pollution hotspots. The Chinese population is from Jones and O'Neill (2016).

2.2.2 Scenario description

We develop three scenarios based on the combined SSPs-RCPs: the Sustainability (SSP1-RCP2.6), "Middle of the Road" (SSP2-RCP4.5) and Economy-first (SSP5-RCP8.5) scenarios. SSPs are the latest global storylines that indicate future socio-economic development in population, urbanization and gross domestic products (O'Neill et al.

2014). SSPs are often used alongside RCPs. RCPs describe different pathways for radiative forcing reaching 2.6 to 8.5 W m⁻² by 2100 (Van Vuuren et al. 2011).

The Sustainability scenario follows SSP1 and assumes rapid socio-economic development towards an environmentally friendly future with a high productivity on land. Based on this, global population growth will slow down and more people will be concentrated in urban areas. The Chinese population is projected to decrease to 1,254 million in 2050. By 2099, the Chinese population is projected to further decrease to 664 million (Table 2.1). Cropland is predicted to decrease between 2010 and 2050, and then increase to above 1,400 thousand km² in 2099 (Table 2.1). In the agriculture sector, it is assumed that the best pest-control technologies would be adopted to replace pesticides. Therefore, we assume that the pesticide application rate will be reduced by 10% in 2050 and 30% in 2099 compared to 2010 (Table 2.1). In this scenario, the greenhouse gas emission will be largely controlled to achieve the maximum warming of 2 °C. Thus, the maximum daily precipitation is predicted to be relatively low (Table 2.1).

The "Middle of the Road" scenario assumes that socio-economic development will follow the same historical trends as SSP2. The total population is projected to decrease slightly between 2010 and 2050 and largely decrease between 2050 and 2099. By 2050, the Chinese population is projected to reach 1,293 million, which is the highest value of the three scenarios. The population is projected to decrease to 790 million in 2099 (Table 2.1). Cropland area is projected to increase in 2050 and then decrease in 2099. In this scenario, development and implementation of technologies are slow in the agricultural sector. Thus, we assume that the pesticide application rate would not change in the future and would remain at the 2010 level. This scenario assumes that efficient climate mitigation strategies against climate change will be implemented (Van Vuuren et al. 2011). Thus, the projected maximum daily precipitation is slightly increased in 2050 compared to 2010. Compared with the Sustainability scenario, some climate models, such as GFDL-ESM2M and IPSL-CM5A-LR, indicate that precipitation will increase in 2099 in this scenario (Table 2.1) (Hempel et al. 2013b).

The Economy-first scenario assumes that the world will develop rapidly with intensive use of resources as in SSP5. The total population in China is projected to decrease slightly between 2010 and 2050. After 2050, the total population is projected to decrease largely by 2099. This would result in 1,259 million people in 2050 and 676

million people in 2099 (Table 2.1). However, the food demand would double nationally between 2010 and 2099. This is associated with an increasing preference for a protein-rich diet. The doubled food demand would require high productivity in the agricultural sector with intensive use of resources. Therefore, it is assumed that the pesticide application rate would increase by 10% in 2050 and by 30% in 2099 compared with 2010 (Table 2.1). Cropland area is also projected to increase to 1,360 thousand km² in 2050 and 1,362 thousand km² in 2099 due to increasing food demand (Table 2.1). RCP8.5 is the scenario with the highest greenhouse gas emission. This will result in a large increase in the maximum daily rainfall in China in the future (Table 2.1). For example, MIROC5 projects the median (min.–max.) of the maximum daily precipitation for China at 29 mm (2–155 mm) in 2050 and 40 mm (2–267 mm) in 2099.

2.2.3 Model evaluation

Our pesticide model for China is based on the existing approach developed and evaluated by Ippolito et al. (2015). They evaluate the model performance by conducting sensitivity and uncertainty analyses. Their results show that the model outputs are sensitive to changes in the slope and precipitation. They also show that the insecticide application rates, interception and precipitation are model inputs contributing largely to uncertainties in the model outputs. To reduce the uncertainty associated with the pesticide application rate, we use the pesticide application rate specific to the Chinese provinces (see Section 2.2.1). To reduce the uncertainty associated with precipitation, we calculate gLOAD using precipitation from the four GCMs and then average the gLOAD results over the four GCMs (see Section 2.2.1). Thus, we avoid the influence of outliers in precipitation on our results.

Table 2.1 Main characteristics of the three scenarios. See Figure S1 in Appendix I for more details of Cropland area.

			Past					
Year	Pesticide application rate (ton km ⁻²) median (min max.) ^a	Population (10 ⁶ person) ^b	Cropland area (10³ km2) c	Max. daily precipitation (mm) ^d median (minmax.) ^a				
2000	0.6 (0.1-4.1)	1280	1312	26 (1-248)				
2010	0.9 (0.3-5.7)	1370	1264	30 (1-297)				
			Future					
Scenario	Pesticide application	Population	Cropland area (10³ km²) c	Max. daily precipitation (mm) ^c median (minmax.) ^a				
	rate (ton km²)*	(10 ⁶ person) ^b		GFDL-ESM2M	HadGEM2-ES	IPSL-CM5A-LR	MIROC5	
			2050					
Sustainability	Decrease by 10%	1255	1095	36(2-291)	35(2-217)	32(2-260)	31(2-249)	
"Middle of the Road"	No change	1293	1287	34(1-267)	32(3-230)	32(2-296)	32(2-229)	
Economic first	Increase by 10%	1259	1360	42(3-194)	35(2-209)	28(2-204)	29(2-155)	
			2099					
Sustainability	Decrease by 30%	664	1411	36(1-400)	31(3-209)	29(3-187)	30(2-128)	
"Middle of the Road"	No Change	790	1192	38(2-266)	34(2-362)	38(3-382)	33(2-224)	
Economic first	Increase by 30%	676	1362	42(2-337)	40(2-241)	31(1-297)	40(2-267)	

^{*}comparing with 2010

^a The median (min. – max.) is the median (min.- max.) value of all grids in China.

^b Future population is derived from Jones and O'Neill (2016).

^c Future cropland area is derived from the Land-use Harmonization² database (Hurtt et al. 2020b)

^d Future precipitation is from four global climate models (GCMs) under each scenario (Hempel et al. 2013b).

Model validation is challenging. Our pesticide model calculates the potential pesticide runoff rather than concentrations. This value reflects the pesticide losses when the maximum daily rainfall event occurs immediately after applying the pesticides. Measurements for such potential pesticide runoffs do not exist for China. We compare our results with those of existing studies. We also compare the spatial patterns in pollution hotspots with two existing studies for the past years for the whole of China. Our model results are in line with those for the pollution hotspots from the two existing studies (Ouyang et al. 2016; Sun et al. 2019). Ouyang et al. (2016) quantified the pesticide losses to waters by provinces from 1990 to 2011. The top ten contributing provinces were Shandong, Guangdong, Anhui, Jiangxi, Henan, Hubei, Jiangsu, Hunan, Sichuan and Heilongjiang. Our results also indicate that these provinces are hotspots except for Anhui, Jiangsu and Heilongjiang. Sun et al. (2019) calculate a small increase in hotspots from 2004 to 2013. We also calculate an increase which is somewhat higher between 2000 and 2010 than the estimate of Sun et al. (2019). A possible reason for the differences between our study and others could be associated with differences in spatial and temporal aggregation levels. We also compare the trends in pesticide use with available studies for the past. Our study shows that the pesticide use in China increased nationally by 37% from 2000 to 2010. This increase is close to the estimate of Ouyang et al. (2016). Generally, our results are in line with those of existing studies. Therefore, we believe that our pesticide model identifies plausible pollution hotspots. We consider that our model reflects China's pesticide use situation because we use Chinese information on pesticides. Thus, we argue that the model can be used to analyze future pesticide pollution hotspots in China in our study. We discuss uncertainties in Section 2.3.3.

2.3 Results and discussion

2.3.1 Pesticide pollution in the past

Pesticide runoff to surface waters from the near-stream agriculture increased considerably in the past. **In 2000**, the potential pesticide runoff in China was calculated as 660 kg. Over half of this was concentrated in five provinces: Hebei, Fujian, Sichuan, Hubei, and Henan (Figure 2.2). We mapped the pesticide hotspots with classes as *very high I, II*, and *III* (see Section 2.2) in China for the past. Pesticide hotspots were in densely populated areas in the north, east, south, central and southwest of China. Additionally, the hotspot areas were distributed in large parts of the Yangtze, the North China and the Chengdu Plains in 2000. The hotspot area covered 26% of China and 55% of the cropland area. Moreover, 62% of the Chinese population lived in pesticide hotspots in 2000 (Figures 2.5 & S6 in Appendix I). This applies to the north, central, south and east part of China (Figure 2.6).

In 2010, the total pesticide runoff in China had increased to 960 kg (Figure 2.2). The largest increases between 2000 and 2010 were calculated at grids of 0.5° in six agroecological zones including north, northeast, south, southeast, east and central China (Figure 2.4, indicated by orange and red). The hotspot area increased to 34% of China (Figure 2.4). The hotspot area expanded in all agro-ecological zones except for the plateau. Some hotspot areas classified as *very high III* in 2000 increased to *very high II* and *I* in 2010 (Figure 2.5). The cropland area in the hotspots increased to 69% (Figure S7 in Appendix I), and the number of people living in hotspots increased to 72% in 2010 (Figures 2.6 and S7 in Appendix I).

2.3.2 Future pesticide pollution under global change

Under the Sustainability scenario, the total pesticide runoff in China is projected to decrease to 780 kg in 2050 and then increase to 950 kg in 2099 (Figure 2.2). In 2050, over 50% of the national pesticide runoff is projected to be in the provinces of Hubei, Sichuan, Hebei, Henan, Hunan and Fujian. This differs from the past and 2099. In 2000 and 2010, Hunan was not among the top five pollution contributors (Figures 2.2 and S2 in Appendix I). In 2099, over 50% of the national pesticide runoff is projected to occur in Fujian, Guangdong, Hubei, Guangxi, Jiangxi and Yunnan provinces. Some southern

provinces, such as Sichuan, Fujian and Hubei, are projected to use less pesticide but still have high potentials for pesticide runoff. This is associated with the steepness of their terrain, heavy rainfall and/or intensive crop production in these provinces.

From 2010 to 2050, many grids in all agro-ecological zones are projected to show a decreased potential pesticide runoff (blue and dark blue in Figure 2.3) or to remain unchanged (yellow in Figure 2.3). Grids showing an increased potential pesticide runoff are projected to be concentrated in two agro-ecological zones including southwest and central China, where the Chengdu Plain is located. In the north, which covers the North China Plain with intensive agriculture, increases in pesticide runoff are projected for fewer grids (Figures 2.3 and S5 in Appendix I). From 2050 to 2099, increases are projected for most grids in the south whereas decreases are projected for most grids in the north (Figure 2.3).

The hotspot areas may shrink to 32% and 30% in 2050 and 2099 compared to the past. The projected spatial distribution of the hotspots in 2050 is similar to that in 2000. Hotspot areas with potentials for high pesticide runoff (very high I and II classes in Figure 2.4) are projected to expand in three agro-ecological zones including the south, southwest and east, and to shrink in two agro-ecological zones including the north and northwest in 2050 compared to 2010 (Figures 2.4 and S6 in Appendix I). The cropland area in hotspot areas is projected to decrease to 65% by 2050 and 67% by 2099. The Chinese population is projected to decrease slightly to 1.25 billion in 2050 and further decrease to 0.66 billion in 2099 (Figure 2.5). The population in the hotspot areas is projected to increase to 76% by 2050 and then decrease to 65% by 2099 (Figure S7 in Appendix I). The hotspot area in the Sustainability scenario is smaller than in the other scenarios. Yet, over half of the Chinese population is projected to live in grids with high potentials for pesticide pollution in the future.

Under the "Middle of the Road" scenario (SSP2 RCP4.5), the pesticide runoff is projected to increase to 1060 kg in 2050 and then decrease to 980 kg in 2099 (Figure 2.2). In 2050, the top five contributing provinces are the same as in 2010: Hubei, Sichuan, Hebei, Henan and Hunan. Over half of the pesticide runoff is projected to occur in Guangdong and the top five contributing provinces (Figures 2.2 and S3 in Appendix I).

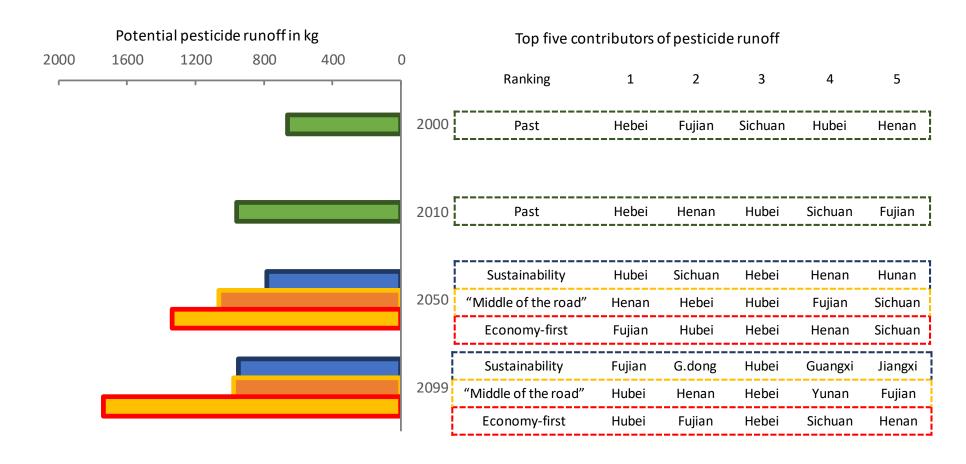


Figure 2.2 Potential pesticide runoff that is generated after heavy rainfall event in China and the top five polluting provinces in 2000, 2010, 2050 and 2099 under the Sustainability (SSP1-RCP2.6), "Middle of the Road" (SSP2-RCP4.5) and Economy-first scenarios. SSPs is short for Shared Socioeconomic Pathways. The ranking starts from the most polluting provinces (left) and ends with the 5th polluting province. Most polluting provinces are the provinces that contribute the most to the national potential pesticide runoff. RCPs is short for Representative Concentration Pathways. Source: See Section 2.2 in the main text for the model and scenario description.

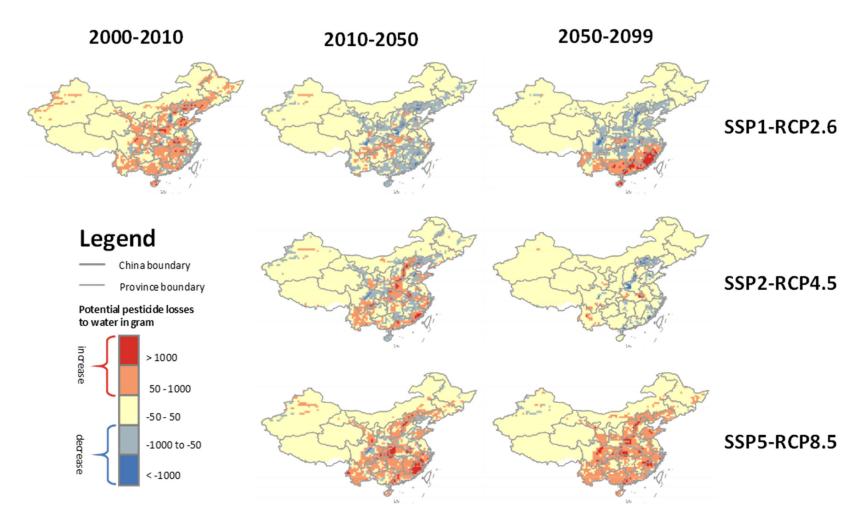


Figure 2.3. Changes in potential pesticide runoff to surface water that are generated after maximum daily rainfall event during the periods of 2000-2010, 2010-2050, and 2050-2090 under the Sustainability (SSP1-RCP2.6), "Middle of the Road" (SSP2-RCP4.5) and Economy-first (SSP5-RCP8.5) scenarios. SSPs is short for Shared Socio-economic Pathways. RCPs is short for Representative Concentration Pathways. Source: see Section 2.2 in the main text for the model and scenario description.

A large spatial variation is projected in future pesticide runoff trends during the period of 2010-2050 (Figures 2.3 and S5 in Appendix I). In the south and southeast, we project many grids of 0.5° to show an increased pesticide runoff between 2010 and 2050. Increases are also projected for some grids in north, central and east China. In the other agro-ecological zones, the pesticide runoff is projected to decrease during 2010-2050 (Figures 2.3 and S5 in Appendix I). From 2050 onwards, the spatial differences in the potential pesticide runoff are smaller. We calculate that the potential pesticide runoff would stabilize in the future in most areas of China. Decreases are projected for some grids of 0.5° in the north, central, southeast and southwest of China from 2010 to 2050 (Figures 2.3 and S5 in Appendix I). In central, south and southwest China, the grids showing a decreased pesticide runoff outnumbered those showing an increased pesticide runoff (Figures 2.3 and S5 in Appendix I).

The pesticide pollution hotspots are projected to cover 33% of China in 2050 and 2099 (Figures 2.5, 2.6 and S7 in Appendix I). In 2099, we projected a larger hotspot for China in the "Middle of the Road" scenario than in the Sustainability scenario (Figures 2.5 and 2.6). However, we projected a smaller hotpot in the northeast of China under the "Middle of the Road" scenario than under the Sustainability scenarios in 2099 (Figures 2.5 and 2.6). In the "Middle of the Road" scenario, we projected that 77% of the Chinese population would live in the pesticide hotspots in 2050 (Figures 2.5 and S7 in Appendix I). For 2099, this percentage drops to 58% which is the lowest of the three scenarios. The percentage of the population in the hotspots in the "Middle of the Road" scenario is lower than that in the Sustainability scenario (Figures 2.4, 2.5 and S6 in Appendix I). This is because grids with high populations in the northeast of China are not classified as hotspots in the "Middle of the Road" scenario (Figure 2.6). The cropland area in the pesticide hotspots is higher than in the Sustainability scenario and stabilized at 71% from 2050 (Figure S7 in Appendix I).

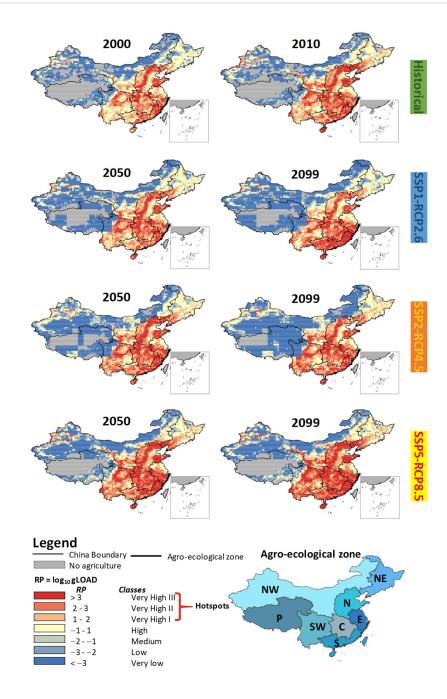


Figure 2.5. Pesticides polluting hotspots map in China in 2000, 2010, 2050, and 2099 under the Sustainability (SSP1-RCP2.6), Middle of the road (SSP2-RCP4.5) and Economy-first (SSP5-RCP8.5) scenarios. Hotspots are defined based on the generic indicator of potential pesticide runoff (RP). We convert values of gLOAD into a logarithmic scale (RP). Results of RP are classified as *very low* (below –3), *low* (–3 to –2), *medium* (–2 to –1), *high* (–1 to 1), *very high I* (above 1), *very high II* (2 to 3), and *very high III* (above 3). Grids of 0.5° are assigned to those classes. Grids classifies as *very high I, II* and *III* are considered as the pollution hotspots for high potential pesticide runoff to surface waters. SSPs is short for Shared Socio-economic Pathways. RCPs is short for Representative Concentration Pathways. Source: see Section 2.2 in the main text for the model and scenario description.

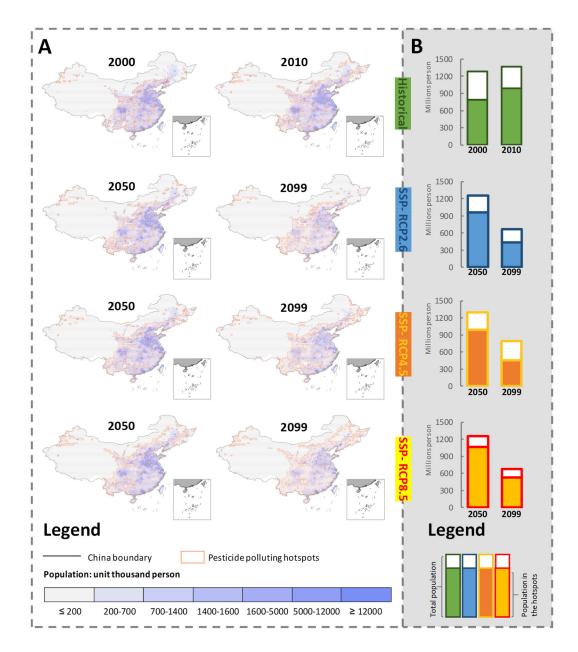


Figure 2.6 Population living in pesticides polluting hotspots in China in 2000, 2010, 2050 and 2099 under the Sustainability (SSP1-RCP2.6), "Middle of the Road" (SSP2-RCP4.5) and Economy-first scenarios. Hotspots are defined based on the generic indicator of potential pesticide runoff (RP). We convert values of gLOAD into a logarithmic scale (RP). Results of RP are classified as *very low* (below –3), *low* (–3 to –2), *medium* (–2 to –1), *high* (–1 to 1), *very high I* (above 1), *very high II* (2 to 3), and *very high III* (above 3). Grids of 0.5° are assigned to those classes. Grids classifies as *very high I*, *II* and *III* are considered as the pollution hotspots for high potential pesticide runoff to surface waters. Total population in hotspots is the sum of population in hotspots grid of 0.5°. Chinese population is the sum of population in grids of 0.5° in China. SSPs is short for Shared Socio-economic Pathways. RCPs is short for Representative Concentration Pathways. Source: the data of population was from Jones and O'Neill (2016). See Section 2.2 in the main text for the model and scenario description.

The Economy-first scenario (SSP5 RCP8.5) can be considered the scenario with the highest environmental impact. Pesticide runoff is projected to reach 1,340 kg in 2050 and 1,740 kg in 2099. These values are much higher than the other scenarios. The top five contributing provinces of 2010 and Guangdong and Hunan together are projected to contribute over 50% of the national pesticide runoff in China in 2050 and 2099 (Figures 2.2 and S4 in Appendix I).

Future trends in the potential pesticide runoff differ considerably among agro-ecological zones and in the other two scenarios. During 2010–2050, a 50 to 1,000 g increases in potential pesticide runoff are projected for most grids of 0.5° in four agro-ecological zones including north, east, central and southwest China (Figure 2.3). Grids with a 50 to 1,000 g decreases in potential pesticide runoff during this period are scattered for all parts of China, except for the plateau (Figure 2.3). From 2050 onwards, the pesticide runoff is projected to increase further (Figures 2.2, 2.3 and S5 in Appendix I).

The hotspot area is projected to be the largest among the three scenarios. The hotspot areas are projected to cover 38% of China by 2050 and 40% by 2099 (Figures 2.5, 2.6 and S7 in Appendix I). The cropland area in the pesticide hotspots is projected to cover 75% by 2050 and 79% by 2099 (Figure S7 in Appendix I). More people are projected to live in the hotspot areas in 2050 (84%) and 2099 (78%) in the Economy-first scenario (Figures 2.6 and S7 in Appendix I). These percentages are higher in this scenario compared to the other two scenarios.

2.3.3 Uncertainties and strengths of this study

Uncertainties are associated with model inputs, approaches, sources and scenario assumptions. We derived model inputs from existing studies. Some model inputs were processed to match the spatial level of detail in our study. Examples are the near-stream agriculture (corridor area) and the pesticide application rates (see Section 2.2). Processing model inputs may generate uncertainties in model results. Some model inputs are fixed such as the Koc value, while some model inputs are area specific. Examples are the pesticide use, precipitation and slope. We used the best available datasets to us to reflect the situation of China (see Section 2.2).

Another source of uncertainties is the modelling approach. We used the approach of Ippolito et al. (2015). This approach was applied globally and regionally. However, it has limitations. An example is that the model treats pesticides as a group and does not distinguish between types and varieties of pesticides. This approach was applied to Europe (Kattwinkel et al. 2011; Schriever and Liess 2007). Schriever and Liess (2007) validated the model for Finland, France and Germany. They modelled the RP for streams. They compared polluted streams (with high potential pesticide runoff) with streams where they observed negatively affected invertebrate communities in Finland, France and Germany. Schriever and Liess (2007) concluded that the modelling approach provides appropriate estimates of potential pesticide runoff. Additionally, Ippolito et al. (2015) evaluated the model performance. All this builds trust in using the modelling approach for analyzing the potential pesticide runoff.

The model accounts for agricultural sources. However, it does not account for pesticide losses from urban areas and sewage. It assumes that all pesticides are applied to agricultural land. However, studies indicated that pesticide losses from urban areas and sewage account for small proportions of the total losses of pesticides to waters. Generally, the largest pathway of pesticide losses is surface runoff. Furthermore, agriculture is a large user of pesticides nationally in China. During heavy rainfall events, it is likely that more pesticides enter nearby waters. Our approach accounts for the most important source (agriculture) and pathway (runoff). Our study focuses on the impact of agriculture on water pollution. We compared our results with Ouyang et al. (2016) and Sun et al. (2019). To our knowledge, these are the only existing studies estimating pesticide losses from agriculture to waters at a national level using an empirical model. The comparisons show that our results are in line with the pollution hotspots from the two existing studies (Ouyang et al. 2016; Sun et al. 2019). Therefore, we believe that the major conclusions of this study are not affected by these uncertainties. Future studies can improve the model by distinguishing between different pesticide varieties. Local studies are required to validate the model for local assessments and to add missing sources such as sewage.

Our future projections are based on SSPs for socio-economic developments and RCPs for climate and hydrology. Our scenarios need projections for cropland, precipitation and the pesticide use in agriculture for 2050 and 2099. We took the projections from

existing studies for population, cropland and precipitation. We made our own projections for the pesticides use on cropland following the storylines of SSPs (Table 2.1). The future is uncertain. Thus, the projections for these variables are also uncertain. Nevertheless, we believe that the projections are useful for exploring future trends in the potential pesticide runoff.

The model offers a first insight into the spatial variability in pesticide pollution in China. It can be used to analyze pesticide pollution in data-poor regions. We perform scenario analyses to better understand the trends of potential pesticide runoff under global change. This is the first study analyzing the effects of global change on pesticide pollution for the whole of China at the 0.5° grid scale. We projected future pollution hotspots for potential pesticide runoff in three scenarios. Our study identifies hotspots in north, east, central and southwest China. This indicates that effective policies to reduce pesticide losses should focus more on these regions. This study provides scientific insights into future changes in pesticide pollution in Chinese waters. These insights can help policymakers to identify pollution areas requiring their attention and develop region-specific policies for those areas to reduce future pesticide pollution under global change.

2.4 Conclusion

This study aims to analyze past and future potential pesticide losses to Chinese waters under global change. To calculate the potential pesticide runoff, we developed a pesticide model based on the existing insecticide model. We used available data from Chinese Statistical Yearbooks as input to the model. To model the impact of socioeconomic developments and climate change on the potential pesticide runoff, we selected and interpreted three combinations of the SSPs and RCPs. These combinations are SSP1-RCP2.6 (Sustainability), SSP2-RCP4.5 ("Middle of the Road"), and SSP5-RCP8.5 (Economy-first).

The potential pesticide runoff increased nationally by 45% from 2000 to 2010. Five out of 31 provinces (Hebei, Hubei, Sichuan, Fujian and Henan) contributed over 50% to the national pesticide runoff in China in 2000 and 2010. In some southern provinces, such as Sichuan, Fujian, and Hubei, pesticide use was less; however, the potential pesticide runoff was relatively high in 2000 and 2010. This is due to steep terrains, heavy rainfall

and/or intensive crop production in these provinces. The pollution hotspots of potential pesticide runoff were concentrated in the agro-ecological zones including the south, southeast, and east, and they cover most parts of the Yangtze, North China and Chengdu Plains in the past. Over 60% of the Chinese population lived in pesticide hotspots in 2000. This value increased to 72% in 2010.

Under global change, 58% to 84% of the Chinese population is projected to live in pesticide pollution hotspots. Future trends in the potential pesticide runoff vary among scenarios. In the Economy-first scenario (SSP5-RCP8.5), the potential pesticide runoff increases relatively rapidly in the future. The top five provinces with the highest potential pesticide runoff in 2010 are projected to remain unchanged in the future. In these five provinces, the potential pesticide runoff is projected to increase by 85% from 2010 to 2099. Pesticide pollution hotspots are projected to expand particularly in the densely populated areas of southeast of China. The hotspots are calculated to accommodate 84% of the total population in 2050. However, we also show that pollution levels can be lower than in 2010 in the future. In the Sustainability scenario (SSP1-RCP2.6), the potential pesticide runoff can be largely controlled. This is due to the reduction in the pesticide application rate and agricultural land, and lower maximum daily rainfall over the year. We projected very different top five provinces with the highest potential pesticide runoff in 2050 and 2099. In 2050, the top five are Hubei, Sichuan, Hebei, Henan and Hunan. In 2099, the top five provinces change to Fujian, Guangdong, Hubei, Guangxi and Jiangxi. In the Sustainability scenario, hotspots for pesticide runoff are concentrated in the southeast of China where the population is projected to be high in 2099. The population living in the hotspots is projected to increase to 76% by 2050 and then decrease to 65% by 2099. In the "Middle of the Road" scenario, we show that pollution level may increase by 2050 but then decrease to the level of 2010 by 2099. This is due to the reduction in agricultural land from 2050 to 2099 in the "Middle of the Road" scenario. In this scenario, the projected top five provinces with the highest potential pesticide runoff are Henan, Hebei, Hubei, Fujian and Sichuan in 2050. In 2099, Yunnan is projected to replace Sichuan in the list of the top five polluting provinces. The distribution of hotspots in 2050 is similar to that in 2099. However, the population in the hotspots is projected to change between 2010 and the future years. The population living in the hotspots is projected to increase to 77% by 2050 and then decrease to 58% by 2099.

Our study shows that pesticide pollution may increase in the future, and that without additional measures, many people in China may be exposed to polluted waters in the future. Our study indicates areas with high potential pesticide runoff. This information can facilitate the formulation of effective, province-specific agricultural policies to reduce pesticide pollution in China in the future.

Acknowledgments

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Chapter 3

How to avoid coastal eutrophication - A backcasting study for the North China Plain

Abstract

Eutrophication is a serious problem in Chinese seas. We explore possibilities to avoid coastal eutrophication without compromising food production in the North China Plain. We used the Model to Assess River Inputs of Nutrient to seAs (MARINA 1.0) for back-casting and scenario analysis. Avoiding coastal eutrophication by 2050 implies required reductions in river export of total nitrogen (TN) and phosphorus (TP) by 50-90% for the Hai, Huai and Huang rivers. We analyzed the potential to meet these targets in 54 scenarios assuming improvements in manure recycling, fertilizer application, animal feed and wastewater treatment. Results indicate that combining manure recycling while reducing synthetic fertilizer use are effective options to reduce nutrient inputs to seas. Without such options, direct discharge of manure are important sources of water pollution. In the 7-25 scenarios with the low eutrophication potential, 40-100% of the N and P in untreated manure is recycled on land to replace synthetic fertilizers. Our results can support the formulation of effective environmental policies to avoid coastal eutrophication in China.

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3.1 Introduction

Chinese coasts have suffered from eutrophication for decades (Daoji and Daler 2004; Liu et al. 2010; Strokal et al. 2014). This also holds for the Bohai Gulf and Yellow Sea, where algae regularly bloom (Liu et al. 2010; Liu et al. 2013; Wu et al. 2010). These algae blooms are caused by increasing river export of nitrogen (N) and phosphorus (P) by the Huang, Huai and Hai rivers (Qu and Kroeze 2012; Strokal et al. 2016b; Yan et al. 2013).

The Huang, Huai and Hai rivers flow through the North China Plain which is the largest area for food production in China. It covers one of the three largest economically developed regions in China called the Jing-Jin-Ji region (Zhang et al. 2010; Zhao et al. 2006). The intensive agricultural practices in combination with the economic growth have resulted in increasing nutrient losses to the rivers (Bai et al. 2014; Li et al. 2017a; Strokal et al. 2017). Direct discharge of untreated animal manure to the rivers, which are regarded as a point sources of pollution, are likely major sources of nutrients in the rivers and thus in coastal waters (Ma et al. 2006; Strokal et al. 2017; Strokal et al. 2016b). In addition, inefficient wastewater treatment results in nutrient losses to the rivers (Huang et al. 2003; Wang et al. 2014). As a consequence, the risks of coastal eutrophication are high in the Bohai Gulf and Yellow Sea (Liu et al. 2010; Liu et al. 2011a).

If current trends continue, future nutrient losses to aquatic systems in China may be higher than today (Strokal et al. 2017; Strokal et al. 2016b). Over half of the nutrient losses to rivers are from food production (Strokal et al. 2016b). The losses would further increase if effective actions are not taken to improve management in agriculture (Wang et al. 2017). The Chinese national government launched several environmental policies, aimed at (1) Zero Fertilizer Growth, (2) Improvement of manure recycling and (3) Prevention and Treatment of Water Pollution ("Ten–Point Water Plan"), to reduce nutrient losses from agriculture and improve water quality (GOV 2015; MOA 2015, 2017). These policies provide opportunities for resolving water quality and offer guidelines to the local government for formulating regional environment protection plans.

Environmental models have been used to explore solutions for nutrient pollution. For instance, several studies modelled nutrient losses from agriculture to the Chinese rivers,

leading to recommendations on nutrient pollution control (Bai et al. 2017; McCrackin et al. 2018; Wang et al. 2018a). For example, Wang et al. (2018a) and Bai et al. (2017) applied the NUtrient flows in Food chains, Environment and Resources use (NUFER) model in China to show that nutrient use efficiencies in China are generally lower than in European countries; they conclude that improving nutrient use efficiencies in food production can lower nutrient losses to waters considerably. This can be achieved through improving animal feed, increasing manure recycling in crop production and improving storage of manure facilities. Yet, the impact of improving nutrient use efficiencies on coastal eutrophication is not well understood.

Other models focus on nutrient flows in the environment. For instance, the Global Nutrient Export from WaterShed (Global NEWS) model projects river export of nutrients to seas, and quantifies the impact on coastal waters worldwide. Global NEWS projections show increasing nutrients export by rivers to the Chinese seas (Qu and Kroeze 2010, 2012; Strokal et al. 2014). Strokal et al. (2017) developed a sub-basin version of Global NEWS for the Chinese rivers: the Model to Assess River Inputs of Nutrient to seAs (MARINA 1.0). MARINA 1.0 includes region-specific nutrient sources, such as the direct discharge of animal excretion to rivers (point sources of manure). MARINA 1.0 projections also show increasing export of nutrients by the Chinese rivers to seas from 2000 to 2050 (Strokal et al. 2016a). According to MARINA 1.0, over half of N and P inputs to the Chinese seas are from the agricultural sector, especially from the direct discharge of animal manure to rivers.

Most studies explore the future trends in coastal eutrophication through scenario analyzes, and first develop qualitative storylines about possible futures. These storylines are then quantitatively interpreted by using the models (Mayorga et al. 2010; Strokal et al. 2016a). These are forward looking scenarios, showing what would happen in the environment if socio-economic trends and policies develop along assumed pathways. Here we follow a back-casting approach to better understand how to realize desired futures. Back-casting was earlier applied in studies on energy futures (Gomi et al. 2011; Haslauer 2015; Holmberg and Robert 2000; Van Vliet and Kok 2013). We adapted this approach to the North China Plain.

In this study, we explored the possibilities to avoid coastal eutrophication in the North China Plain (NCP). To this end, we combine back-casting and forward looking scenario analysis. We used the MARINA 1.0 model and Indicator for Coastal Eutrophication Potential (ICEP) (Garnier et al. 2010; Strokal et al. 2016a). First, we quantified environmental targets for river export of nutrients, using the ICEP approach. We quantified nutrients exported by the Huang, Huai and Hai rivers for the year 2050 and used the Global Orchestration (GO) scenario from the Millennium Ecosystem Assessment as a baseline. We then explored how future improvements in agriculture and wastewater treatment (WWT) can achieve the environmental targets.

3.2 Materials and methods

3.2.1 Back-casting method

Our back-casting approach follows two steps (Figure 3.1). First, a desirable future is defined. In our manuscript, this is a future with a low potential for coastal eutrophication (see Section 3.2.5). Second, pathways or management options are explored to reach this desirable future (see Section 3.2.6).

In the first step, we set environmental targets for river export of nutrients to seas. These targets aim to avoid coastal eutrophication, and are derived from the Indicator for Coastal Eutrophication Potential (ICEP) (Billen and Garnier 2007). ICEP is calculated from river loads of nutrients (N and P) and dissolved silica, as explained below. The ratio of nutrients to Si indicates whether environmental conditions are favourable for algal growth. To avoid algal blooms, the ICEP value should be lower than 0 with an uncertainty range (-1 to +1). We took this as an indicator for low risks of coastal eutrophication. Based on this, we quantified maximum total N (TN) and total P (TP) inputs to the seas (see Section 3.2.4).

In the second step, we developed alternative scenarios to explore possible combination(s) of management options in agriculture and wastewater treatment that can reduce river exports of N and P to the maximum desired TN and TP levels. Our scenarios were based on a number of assumptions. We assumed that fertilizer use in China can be reduced without crop yield loss based on literature and expert knowledge.

Currently, synthetic fertilizers and animal manure that are applied on land are higher than crop requirements under the current practice in the NCP (Ju et al. 2009; Li et al. 2011). Therefore, we assumed that TN and TP inputs to land can be reduced in all

scenarios by 2050 (see below). Then we considered three groups of options to increase nutrient use efficiencies and thus further reduce nutrient inputs to waters. (1) We assumed that N and P in animal excretion will not be reduced (0%) or can be reduced (by 10% and 20%) by improving the quality of animal feed in combination with the precision feeding. (2) We assumed that N and P removal rate in wastewater treatment will not be improved (0%) or can be improved (to 50%, and 80%). (3) We assumed that recycling rate of point manure on land will not be increased (0%) or can be increased (to 20%, 40%, 60%, 80%, and 100%); and that recycled manure can be used to replace synthetic fertilizers.

These combinations of options for agriculture and sewage systems result in 54 alternative scenarios. We quantified river export of nutrients for these 54 alternative scenarios by the MARINA 1.0 model (Figure 3.1) and compare these with the desired maximum nutrient loads.

3.2.2 Study area

The Huang, Huai and Hai rivers flow through the NCP to the seas. The NCP is also therefore known as the Huang-Huai-Hai plain (Figure 3.2). These three rivers supply a large portion of water for agriculture in NCP. Huang is the second largest basin in China and has been divided into six sub-basins in the MARINA 1.0 model. Lanzhou and Toudaogual are two upstream sub-basins. Longmen, Wehe Jinghe and Huayuankou are the middle-stream sub-basins group. The Huang He Delta is the down-stream sub-basin.

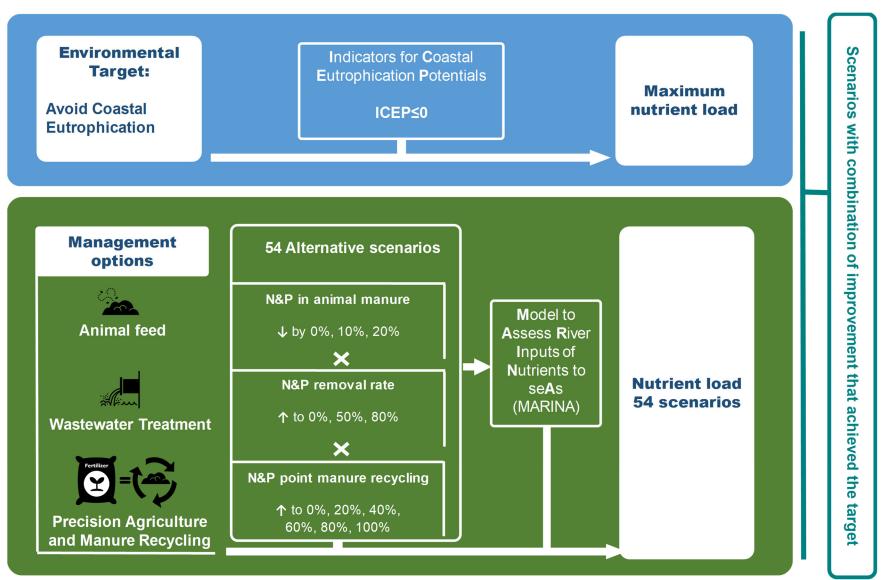


Figure 3.1. Research steps in the back-casting of this study.

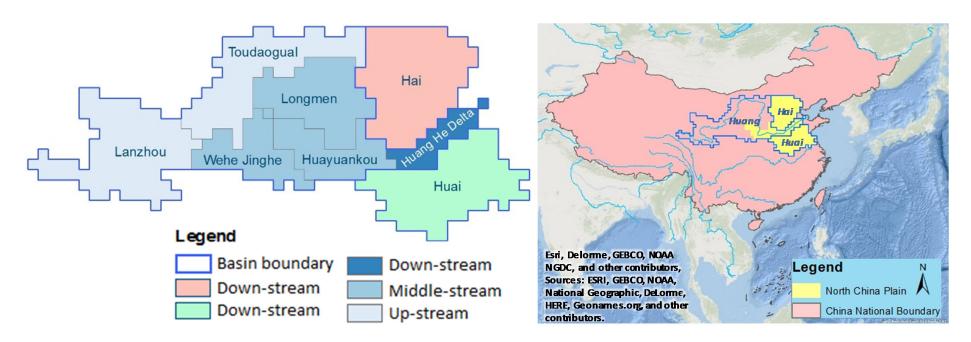


Figure 3.2 The overview of selected basins in the North China Plain.

3.2.3 MARINA 1.0 model

The MARINA 1.0 model calculates river export of dissolved inorganic and organic N and P (DIN, DIP, DON, DOP) for the year 1970, 2000 and 2050 for the six Chinese basins (Mg year⁻¹) (Strokal et al. 2016a). This model was developed on the basis of the Global NEWS model (Mayorga et al. 2010). The MARINA 1.0 model has been validated and applied in earlier studies (Strokal et al. 2016a; Strokal et al. 2017). Main equations of the MARINA 1.0 model can be found in Box A.1 in Appendix II.

The model quantifies river export of nutrients using the sub-basin approach for the six large rivers. In total, there are 25 sub-basins for the six rivers. For the Huang, Huai and Hai rivers, the MARINA 1.0 model distinguishes eight sub-basins (Figure 3.2) (Strokal et al. 2016a). The MARINA 1.0 model calculates the river export of nutrients as a function of anthropogenic activities on land, such as crop and animal production and wastewater discharge. The model also takes into account sub-basin characteristics (e.g., hydrology, land use) and nutrient retention or removal by water consumption along the transportation in the river network (see below).

The model calculates the attribution of point sources and diffuse sources for the river export of N and P (Strokal et al. 2016a). Sewage effluents, detergent use, direct discharge of animal manure and uncollected human waste to rivers are accounted as point sources. In sewage effluents, N and P are partly removed through wastewater treatment system and the detergent use is quantified as a function of population. The direct discharge of animal manure to rivers are calculated as: the total animal excretion times the share that is directly discharged to rivers. A similar approach is applied for uncollected human waste (Strokal et al. 2016a). The diffuse sources include synthetic fertilizers, animal and human excretion on land, atmospheric N deposition, biological N fixation, and weathering of P-contained minerals. The model also considers animal grazing, crop harvesting, retentions in soil, and leaching of organic matter. The total nutrient inputs from land to rivers are estimated as a function of the annual runoff (Strokal et al. 2016a).

Nutrient retention and losses along the river are calculated based on water consumption, denitrification, sedimentation, and dammed reservoirs along the river network. The MARINA 1.0 model follows the approach that is used in the Global NEWS model on quantifying the nutrient retention and losses (Strokal et al. 2016a), but with updated

information for reservoirs (Lehner and Döll 2004). The MARINA 1.0 model also considers sedimentation processes of DIP in rivers (Strokal et al. 2016a). Large basins are classified into the up-, middle- and downstream. Nutrients travel generally, longer from the upstream than from the middle- and down-stream sub-basins to seas.

In this study, we also included particulate forms of N and P. These particulate forms of N and P are calculated by the Global NEWS model based on a linear regression analysis, taking into account the total suspended solids and basin characteristics such as precipitation and slope (Beusen et al. 2005; Mayorga et al. 2010). We included direct N deposition on coastal waters as a pollution source for coastal eutrophication in this study (Luo et al. 2014).

3.2.4 Model performance

The performance of the MARINA 1.0 model was discussed in earlier studies (e.g., Strokal et al., 2016a). The model was validated by comparing measured DIN, DIP, TN and TP at the river mouth with their modelled values for the Huang, Changjiang and Zhujiang rivers. Measured values of the nutrients for each river were available from several studies (Tong et al. 2016; Tong et al. 2015) and are summarized in Strokal et al. (2016a). Validation results were shown by a number of statistical indicators: the Pearson's coefficient of determination (R²P, 0-1), the Nash-Sutcliffe efficiency (R²NES, 0-1), and Model Error (ME, %) according to Moriasi et al. (2007). Results of these statistical indicators were 0.84 for R^{2}_{P} , 0.78 for R^{2}_{NES} and 8% for ME (Strokal et al., 2016a). Calculated indicators show a good performance of the MARINA 1.0 model for the Chinese rivers. For example, MARINA 1.0 modelled that the Huang River exported 46 kton of DIN in 2000. This is reasonably in line with experimental studies: 46 to 178 kton year⁻¹ of DIN during the period of 1990s-2012 (Strokal et al. 2016a). Tong et al. (2016) reported measured data for our studies rivers. However, those measurements are not easily comparable with our modelled results because of different drainage areas and period.

Strokal et al. (2016a) compared the results of MARINA 1.0 with other modelling studies. Results of MARINA 1.0 compare well to the spatial and temporal variation in nutrient loads of other studies. For example, Tao et al. (2010) reported an increasing trend of N inputs to the down-stream sub-basin of the Huang River from 1970s. Xu et al. (2014)

found that the nutrient concentration in the Huang and Huai rivers were higher than in the Zhujiang river. Tong et al. (2016) showed that nutrient inputs to the up-stream of the Huang River are lower than nutrient inputs to the down-stream sub-basins. These results are consistent with the results of MARINA 1.0. Furthermore, a sensitivity analysis was performed to show how sensitive the model outputs of MARINA 1.0 are to changes in model inputs and parameters (see details in Strokal et al., 2016a). Results indicated that river export of nutrients is sensitive to the changes in animal manure production, direct discharge of animal manure to rivers, synthetic fertilizer inputs to land, and river discharge.

In summary, the performance of the MARINA 1.0 model was evaluated from different aspects such as validation, comparison of the trends with existing studies and sensitivity analysis (see Strokal et al., 2016a). In our study, we further compared the results of MARINA 1.0 with existing studies and discussed model uncertainties in Section 3.3. All this builds trust in using the MARINA 1.0 model for our future analysis of the possibilities to avoid coastal eutrophication in the NCP.

3.2.5 Environmental targets for nutrient inputs to seas

We used the Indicator for Coastal Eutrophication Potential (ICEP) (Billen and Garnier 2007) to represent the risks on coastal eutrophication. The ICEP value was calculated based on the C:N:P:Si ratio = 106:16:1:20 (in kg C km⁻² day⁻¹). ICEP represents development of the non-siliceous algae in coastal water that can be harmful. The risk of coastal eutrophication is low if ICEP is below zero (Billen and Garnier 2007). The ICEP was calculated either for N (ICEP_N) and for P (ICEP_P) depending on limiting nutrient following Billen and Garnier (2007).

$$ICEP_N = \left[\frac{TN_{flx}}{(14\times16)} - \frac{Si_{flx}}{(28\times20)}\right] \times 106 \times 12$$
 IF TN_{flx}:TP_{flx}<16 (N limiting) **eq. 1**

$$ICEP_P = \left[\frac{TP_{flx}}{31} - \frac{Si_{flx}}{(28 \times 20)}\right] \times 106 \times 12$$
 IF TN_{flx}:TP_{flx}>16 (P limiting)**eq. 2**

where:

ICEP_N: Indicator for Coastal Eutrophication Potential for nitrogen (kg km⁻² day⁻¹);

ICEP_P: Indicator for Coastal Eutrophication Potential for phosphorus (kg km⁻² day⁻¹);

 TN_{flx} : total nitrogen (TN) fluxes by rivers to seas (kg km⁻² day⁻¹). TN fluxes were derived from point sources including direct discharge of animal manure and human excretion, and discharge from sewage systems to surface waters; diffuse sources including application of synthetic fertilizers, animal manure and human excretion on land, biological N fixation by crops, atmospheric N deposition, and leaching of organic matter.

TP_{flx}: total phosphorus (TP) fluxes by rivers to seas (kg km⁻² day⁻¹). TP fluxes were derived from point sources including direct discharges of animal manure and human excretion, use of P based detergents, and discharge from sewage systems to surface waters; and diffuse sources including application of synthetic fertilizers, animal manure and human excretion on land, weathering of P-contained minerals, leaching of organic matter.

Si_{flx:}: total dissolved silica (Si) fluxes by rivers to seas (kg km⁻² day⁻¹). Si fluxes were derived from the Global NEWS model (Beusen et al. 2009). Si fluxes are calculated by a multi-linear regression model including the soil bulk density, precipitation, slope, and areaa with the volcanic lithology (Beusen et al. 2009).

3.2.6 Maximum nutrient inputs to seas

We used ICEP to set the target for nutrient inputs to seas. We calculated these targets for individual basins. As it was described in the previous section, the risk for coastal eutrophication is low when ICEP value is below zero. We set the target as ICEP equals to zero with a uncertainty range from -1 to +1. The target for nutrient inputs to seas was, therefore, calculated from equation 3 and 4:

$$TN_{flx} = \left(\frac{ICEP_N}{106 \times 12} + \frac{Si_{flx}}{28 \times 20}\right) \times (14 \times 16)$$
 eq. 3

$$TP_{flx} = \left(\frac{ICEP_P}{106 \times 12} + \frac{Si_{flx}}{28 \times 20}\right) \times 31$$
 eq. 4

Where $ICEP_N = 0$ (-1 to +1), $ICEP_P = 0$ (-1 to +1) and the river export of Si (Si_{flx}) is known for Huai and Hai from the MARINA 1.0 model, the Si_{flx} for Huang was estimated based on measurements for the years from 2002 to 2004 (Gong et al. 2015).

3.2.7 Scenario description

3.2.7.1 Baseline scenario: Global Orchestration (GO)

In the following, we described our scenarios. We first presented the baseline scenario. Then, we described our alternative scenarios relative to the baseline.

Global Orchestration (GO) is a scenario of the Millennium Ecosystem Assessment (MA) and selected as a baseline here. GO assumes a globalized world with a reactive attitude towards environmental management (Alcamo et al. 2005c). In GO, the Chinese population will grow but the growth rate will be moderate. Under the rapid urbanization and increasing income, the inequality of income in the society will be reduced and the purchasing power is assumed to be high. People's diets will shift to protein-rich food consumption, and thus lead to an increasing animal production and a higher humanwaste production (Bouwman et al. 2009). The agricultural land is assumed to expand, and synthetic fertilizer inputs increase. In line with the rapid economic development and urbanization, GO assumes an increasing water withdrawal. The N removal rates in wastewater treatment are assumed to be increasing in the future as a result of new technologies, but the wastewater return flow from human consumption will also increase (Seitzinger et al. 2010; Van Drecht et al. 2009).

3.2.7.2 Alternative scenarios

We developed 54 alternative scenarios relative to the baseline (GO) scenario (Figure 3.3). These scenarios focused on three main improvements to reduce nutrient losses to waters: (1) reducing total N content in animal manure by improving the quality of animal feed in combination with the precision feeding (indicated by A); (2) improving N and P removal rate in wastewater treatment (indicated by W); and (3) recycling animal manure and balanced fertilization (indicated by P). This last option implies the reuse of animal manure on land, thus lowering the need for synthetic fertilizers. 54 alternative scenarios combine three improvements. For example, a scenario consists of a combination of A, W and P (see Figure 3.3).

For all 54 scenarios, we assumed reductions in nutrient inputs to cropland, to avoid overfertilization. In current farming practice, nutrient inputs to cropland from synthetic fertilizers and animal manure are generally 15-30% higher than the crop demand in the NCP (Chen et al. 2014; Chen et al. 2006; Cui et al. 2018; Ju et al. 2009; Li et al. 2011). This is because of poor nutrient management and subsidies by the government for the use of

synthetic fertilizers. However, recently introduced environmental policies may change this (e.g., "Zero Growth in Synthetic fertilizers after 2020"). Existing studies indicate that it is possible to reduce N inputs to cropland from synthetic fertilizers and animal manure by 18 to 32% without compromising crop yields (Chen et al. 2014; Cui et al. 2018; Ilebekk et al. 1978; Ju et al. 2009; Ma et al. 2010; Zhang et al. 2016b). These studies are partly based on results from 13,123 field trials (covering 37.7 million cumulative hectares) in 74 Science and Technology Backyards (STBs) within 21 Chinese provinces where scientists and farmers work together to avoid overfertilization of cropland while maintaining crop yields from 2005 to 2015 (Chen et al. 2014; Chen et al. 2006; Cui et al. 2018; Zhang et al. 2016b). The results indicate that currently many farmers tend to overfertilize their fields. Thus we assumed in our scenarios a 30% reduction in N inputs to cropland compared to GO without changing the crop yield. The same holds for P inputs, for most studied sub-basins (Li et al. 2015). An exception is the up-stream of the Huang River where we assumed a 15% reduction in P inputs from synthetic fertilizers and animal manure compared to GO. An important reason is that available P in the soil is currently limited for crop production, but will further accumulate in this regions till 2050. The up-stream of the Huang River is an example of such a region (Li et al. 2015).

1) Reducing total N in animal manure by improving the quality of animal feed Scenarios assuming reductions in the total N in manure are indicated by "A" in Figure 3.3. We distinguish A0, A1, and A2 scenarios in our matrix (Figure 3.3). A0 indicates scenarios without improved the quality of animal feed and thus N in manure stays as in G0. A1 and A2 scenarios assume improved the quality of animal feed. Thus, N in animal manure is assumed to be reduced relative to G0 (Lesschen et al. 2011). In China, animal farms has been shifting from traditional, small farms towards industrial farms (Bai et al. 2018). Industrial farms, generally, tend to provide the feed for animals with the higher quality (e.g. corn and soybean under precision feeding) compared to the traditional farms (e.g. kitchen waste). Modifying the feed composition combined with precision feeding can reduce N in animal excretion by up to 20% without damaging livestock productivity (Bai et al. 2014). Therefore, in this study, we assumed that by 2050 it is possible to reach 10% (A1 scenarios) or 20% of reduction (A2 scenarios) in the total N in animal manure relative to G0. We assumed that these reductions are technically

possible to achieve by 2050 (see existing studies on livestock production:Ma et al. 2013a; Wang et al. 2018a).

2) Increasing N and P removal of the wastewater treatment Scenarios assuming improvements in wastewater treatment are indicated by "W" in Figure 3.3. We distinguish W0, W5 and W8 scenarios. W0 indicates that N and P removal rate in wastewater treatment system stays as in GO (no improvements). W5 and W8 assume N and P removal rates will increase to 50% and 80%, respectively (Figure 3.3). The N and P removal in today's wastewater treatment in Chinese urban areas is generally low (Zhang et al. 2016a). The reasons for the low nutrient removal efficiency vary among regions. For instance, low temperatures and relatively old equipment play a role in northern China. Available technologies exist with higher N and P removal rates (Kartal et al. 2010; Shi et al. 2010; Winkler et al. 2012). For instance, the anammox process has been studied and applied in pilots in many regions (Ali et al. 2013; Cheng et al. 2017; Tang et al. 2017), resulting in high N removal rates (up to 90%) under cold temperatures (about 18 °C) (Winkler et al. 2012). P removal rates in the current conventional wastewater treatment plants can reach up to 90% reduction, and it can be combined with advanced treatment technologies for P recovery (Khiewwijit 2016). Here, we assumed that N and P removal rates could increase to 50% (W5 scenarios) or 80% (W8 scenarios) by 2050.

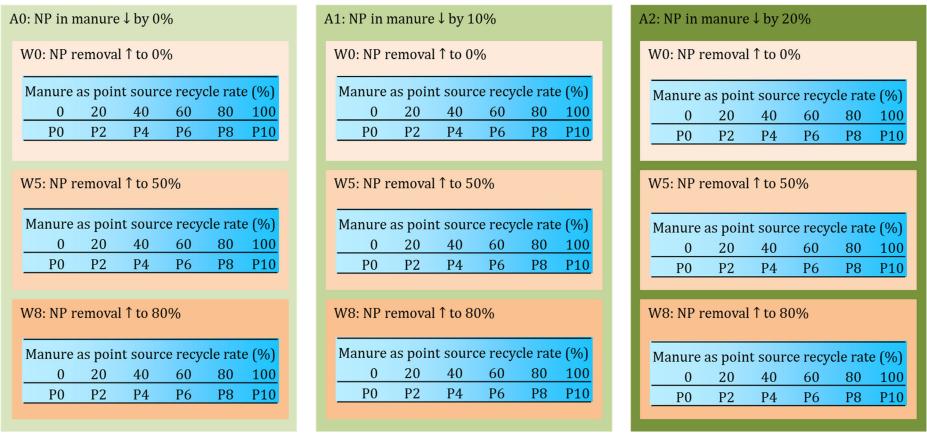


Figure 3.3 Overview of the 54 scenarios. The scenarios differ from the baseline (GO) as follows: lower N (nitrogen) and P (phosphorus) content of manure (indicated by A), higher N and P removal in wastewater treatment (indicated by W) and percentage of the point manure that is are applied on the land as fertilizer (indicated by P). A scenario consists of a combination of A, W and P. For instance: scenario A1W5P8 assumes a 10% reduction in the N and P content of manure, 50% of N and P removal rate in wastewater treatment, and 80% of point manure reused on land as fertilizer.

3) Replacing synthetic fertilizers by recycling manure

Animal manure can be used for replacing synthetic fertilizers. In our scenarios, we assumed different levels of recycling of manure on land as fertilizer; this would reduce point source inputs of manure to rivers (indicated by P). We assumed the following percentages of the manure recycling (Figure 3.3): no recycling (P0 scenarios), 20% (P2 scenarios), 40% (P4 scenarios), 60% (P6 scenarios), 80% (P8 scenarios) and 100% (P10 scenarios). These percentages refer to the amount of manure that is discharged directly into rivers as a point source in the GO scenario. We used the mineral fertilizers equivalency (MFE), which is the rate to replace synthetic fertilizers with animal manure. MFE is different for N and P. In general, one unit of N in animal manure can replace 0.6 unit of synthetic fertilizers, while one unit of P in animal manure can replace one unit of P in synthetic fertilizers (Jensen 2013). Thus, the share of animal manure in the total N inputs to land was assumed to be increasing; the scenarios assume 0 to 100% reuse of the animal manure that is in GO discharged as point sources to rivers (Figure A.2 in Appendix II). The total P inputs from animal manure and synthetic fertilizers are the same in all scenarios (Figure A.3 in Appendix II). Thus, the total nutrient inputs to land from animal manure are increasing from 0% to 100% of point manure recycling and the total nutrient inputs to land from synthetic fertilizers are decreasing, accordingly (Figures A.2 and A.3 in Appendix II).

3.3 Results

3.3.1 Targets for river export of N and P for 2050

Environment targets for TN and TP inputs to seas in 2050 differ among rivers, nutrient forms and ICEP ranges (Figure 3.4). The targets for TN and TP river exports to seas were calculated based on ICEP_N 0 ranging from -1 to +1. The GO scenario projected that the *Huang River* may export up to about 270 Gg of TN and up to 40 Gg of TP to the Bohai Gulf in 2050. Reaching ICEP 0 (-1 to +1) requires the river to export not \geq 19 (0 to 76) Gg of TN and 2 (0 to 10) Gg of TP in 2050. This implies almost a 90% reduction in river export of both N and P compared to the GO level (Figure 3.4). It may be easier to reduce dissolved inorganic and organic N and P because of their dominant share in TN and TP river export (Figure 3.4).

Under GO, we calculated that the *Huai River* may export up to 150 Gg of TN and around 35 Gg of TP to seas in 2050 (Figure 3.4). This river export of TN and TP considerably exceeds the ICEP 0 (-1 to +1) target for TN which is 71 (55 to 87) Gg and for TP which is 10 (8 to 12) Gg. To reach the target, TN inputs to seas need to be reduced by 50% and TP inputs to seas by 70% (Figure 3.4). The reduction in TN can be achieved by reducing the river export of DON that contributes 80% to TN. The target for TP can be achieved by reducing river export of DOP that contributes 60% to the TP.

The calculated TN and TP inputs from the *Hai River* to seas is 80 and 15 Gg year-1, respectively. These amounts exceed the target ICEP 0 (-1 to +1) by a factor of 4 (1.5 to 167) for TN and a factor of 6 (2 to 217) for TP. This implies a 80% to 90% reduction in TN and TP inputs to seas. The targets for TN and TP can be achieved by reducing the river export of dissolved organic forms, which contribute over 80% to the TN and TP exports (Figure 3.4).

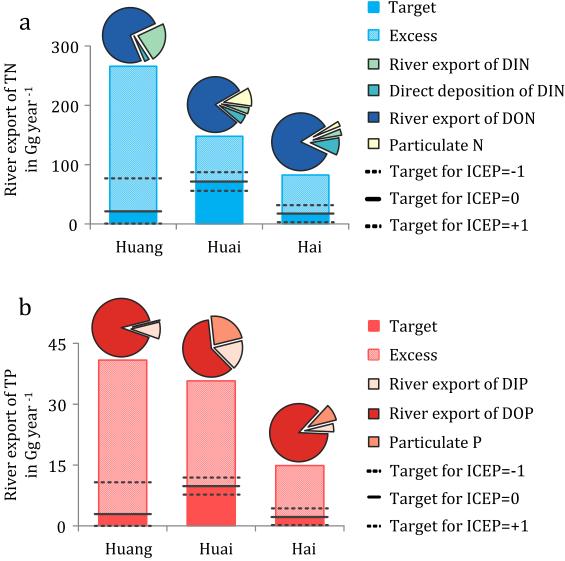


Figure 3.4 a: Total nitrogen (TN) and b: total phosphorus (TP) inputs to the sea from the rivers in 2050 (Gg year-1). The graph shows the targets for TN and TP inputs to seas when ICEP = 0 (with uncertainty range from -1 to +1) under Global Orchestration (GO) scenario. ICEP is the Indicator for Coastal Eutrophication Potential (see Section 3.2.4). GO assumes a globalized trends in socio-economic development with a reactive approach to manage environmental pollution (Seitzinger et al. 2010; Strokal et al. 2016a). DIN and DIP are dissolved inorganic N and P, respectively. DON and DOP are dissolved organic N and P, respectively. DON and DOP are dissolved organic N and P, respectively. DIN, DIP, DON and DOP are from the MARINA 1.0 model (Strokal et al. 2016a). Particulate N and P are from the Global NEWS model (Mayorga et al. 2010). Direct deposition of DIN is from Luo et al. (2014). ICEP values and the targets are calculated using equations in Section 3.2.4.

3.3.2 River export of N and P in the 54 alternative scenarios

Our results indicate that avoiding coastal eutrophication is technically possible in multiple ways for each basin. We developed 54 scenarios that differ in nutrient management and wastewater treatment (see Section 3.2.6 for scenario description).

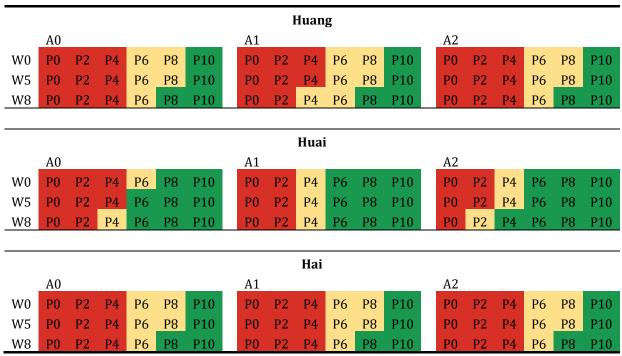
The ICEP_N +1 target for the *Huang River* can be achieved in 12 scenarios (out of 54) (Table 3.1 - Huang) and the ICEPP target in 10 scenarios (Table 3.2 - Huang). One of the ways to reach the ICEP +1 targets is to reuse all available animal manure on land to replace synthetic fertilizers (the A0W0P10 scenario in Figure 3.5 and the A0W0P10 scenario in Figure 3.6). Another way is to combine reuse of manure on land with improved wastewater treatment or/and improved animal feed (e.g., 80% of manure recycling with other options in A0W8P8, A1W8P8, A2W8P8 in Figure 3.5). However, reaching the target ICEP 0 may be difficult. Even under the A2W8P10 scenario, which is expected to have the highest potential reduction in nutrient inputs to seas, the TN (37 Gg year⁻¹) and TP (4 Gg year ⁻¹) are higher than the targets for nutrients inputs to seas (ICEP_N 0 = 19 Gg year⁻¹, ICEP_P 0 = 2 Gg year⁻¹). This implies that achieving the target for ICEP 0 requires other mitigation strategies for the Huang basin than for the other basins. This has two reasons. First, the ICEP 0 target for the Huang River is lower than for the Huai and Hai rivers, while the river export of TN and TP by the Huang River is higher (Figure 3.4). This indicates that the Huang River needs to reduce large amounts of nutrients to seas in 2050. Another important reason can be that the total dissolved silica (TDSi) exported by the Huang River is low because the river is generally dry. The Si is an important nutrient in establishing the ICEP target. Less Si may lead to more eutrophication and thus stricter ICEP target.

For the *Huai river*, there are more scenarios that can reach the target of ICEP_N than the target of ICEP_P. There are 28 out of the 54 scenarios in which the ICEP_N +1 target can be achieved (Table 3.1 - Huai). For example, it is possible to reach the ICEP_N +1 target by reusing 80% of point manure on land (Table 3.1 & Figure 3.5 - Huai - A0W0P8). However, reaching the ICEP_N target is difficult by only improving animal feed or/and improving sewage systems. Comparing with TN, we found fewer scenarios reached the target for ICEP_P. There are only seven scenarios reaching the ICEP_P +1 target (Table 3.2 - Huai & Figure 3.6). These seven scenarios incorporate options to improve the quality of

animal feed, to improve wastewater treatment systems and to reuse animal manure. This indicates a need for integrated pollution control options to reach the target of ICEPP +1 for Huai (Table 3.2 - Huai, A0W5P10, A0W8P10, A1W5P10, A1W8P10, A2W0P10, A2W5P10, A2W8P10). Reaching the ICEP_P 0 target is, however, difficult. Our results show that in none of the scenarios this is possible. Extra pollution control for TP is needed in addition to improving the quality of animal feed, improving removal efficiencies of sewage systems and recycling of animal manure. Yet, it is possible to reach the ICEP_N -1 target. Most of these scenarios that achieved the ICEP_N -1 are with reusing 40-100% of point manure on land (Table 3.1 - Huai, A0W0P8, A0W5P6, A2W8P4). According to the results, the potential reduction in nutrient inputs to seas is achieved by recycling animal manure. This reduction is larger than in the option for improving sewage systems or improving animal feed. Additionally, the effect of improving wastewater treatment may reduce more TN than the TP inputs to seas. Generally, improving wastewater treatment may not result in large reductions in TN and TP inputs to seas. This is because the N and P removal rates are already above 50% under the baseline scenario (GO), and the technical potential of removal rates can only be improved to 50% or 80%. This means that the improvement on N and P removal rates is very limited. Furthermore, only about 2-10% of TN and TP are from sewage. Thus the effect of improving wastewater treatment is not very obvious (Figure A.1 in Appendix II). This finding is also observed in the Huang and Hai rivers.

For the *Hai river*, the target for TN and TP inputs to seas can be reached in more scenarios than for the Huang River (Figures 3.5 and 3.6, Table 3.1 and 3.2). There are 12 scenarios with TN lower than the ICEP_N +1 target and 9 scenarios with TP lower than the ICEP_P +1 target. It is similar to the other two basins that more scenarios achieved the target for N than for P (Table 3.1 and 3.2). It can be explained by the difference in the source attribution between TN and TP (Figure A.1 in Appendix II). Point source of manure is the dominant source of both TN and TP in the rivers, and contributes $\geq 50\%$ to both TN and TP in the Huang, Huai and Hai basins. But sewage effluents contribute more to river export of TN than of TP. As a result, larger reduction is achieved for TN then TP by improving wastewater treatment. This is also reflected by the achieved reduction in nutrient inputs to seas from the different sources (Figures 3.7 and 3.8).

Table 3.1 Overview of scenarios for which river export of TN (total nitrogen) does not^a reach the ICEP_N = +1 target (red colour), almost reach^b (orange colour), or reach^c (green colour) for Huang, Huai and Hai river basins. The targets for TN when ICEP_N = +1 is calculated using equations in Section 3.2.4. ICEP is the Indicator for Coastal Eutrophication Potential (see details in Section 3.2.4). The 54 alternative scenarios differ from the baseline (Global Orchestration (GO) scenario) as follows: lower N content of manure (indicated by A), higher N removal in wastewater treatment (indicated by W) and percentage of the point manure that is applied on the land as fertilizer (indicated by P). A scenario consists of a combination of A, W and P. For instance: scenario A1W5P8 assumes a 10% reduction in the N content of manure, 50% of N removal rate in wastewater treatment, and 80% of point manure reused on land as fertilizer. The baseline scenario (GO) assumes a globalized trends in socio-economic development with a reactive approach to manage environmental pollution (Seitzinger et al. 2010; Strokal et al. 2016a).



a Do not reach: $TN > TN_{ICEP+1} + (TN_{ICEP+1} - TN_{ICEP0})$

b Almost reach: TN ≤ TN_{ICEP+1} + $(TN_{ICEP+1} - TN_{ICEP0})$

c Reached: TN ≤ TN_{ICEP+1}

Table 3.2 Overview of scenarios for which river export of TP (total phosphorus) does not reach the ICEP_P = +1 target (red colour), almost reach (orange colour), or reach (green colour) for Huang, Huai and Hai river basins. Almost reach: TP - TP_{ICEP+1} \leq TP_{ICEP+1} - TP_{ICEP+1} - TP_{ICEP+1} or TP_{ICEP+1} - TP_{ICEP+1} - TP_{ICEP+1} or TP_{ICEP+1} is the Indicator for TP when ICEP_P = +1 is calculated using equations in Section 3.2.4. ICEP is the Indicator for Coastal Eutrophication Potential (see details in Section 3.2.4). The 54 alternative scenarios differ from the baseline (Global Orchestration (GO) scenario) as follows: lower P content of manure (indicated by A), higher P removal in wastewater treatment (indicated by W) and percentage of the point manure that is applied on the land as fertilizer (indicated by P). A scenario consists of a combination of A, W and P. For instance: scenario A1W5P8 assumes a 10% reduction in the P content of manure, 50% of P removal rate in wastewater treatment, and 80% of point manure reused on land as fertilizer. The baseline scenario (GO) assumes a globalized trends in socioeconomic development with a reactive approach to manage environmental pollution (Seitzinger et al. 2010; Strokal et al. 2016a).

Huang																				
	A0							A1							A2					
W0	P0	P2	P4	Р6	P8	P10		P0	P2	P4	Р6	P8	P10		P0	P2	P4	Р6	P8	P10
W5	P0	P2	P4	Р6	P8	P10		P0	P2	P4	Р6	P8	P10		P0	P2	P4	Р6	P8	P10
W8	P0	P2	P4	P6	P8	P10		P0	P2	P4	Р6	P8	P10		P0	P2	P4	P6	P8	P10
Huai																				
	A0							A1							A2					
W0	P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10
W5	P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10
W8	P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10
Hai																				
A0							A1							A2						
W0	P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10
W5	P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10
W8	P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10		P0	P2	P4	P6	P8	P10

a Do not reach: $TP > TP_{ICEP+1} + (TP_{ICEP+1} - TP_{ICEP0})$

b Almost reach: $TP \le TN_{ICEP+1} + (TN_{ICEP+1} - TN_{ICEP0})$

 c^* Reached: TN \leq TN_{ICEP+1}

GO2050

A2W0P4

A2W0P6

A2W0P0

A2W0P2

A2W0P8

■ GO2050

A2W5P0

A2W5P2

A2W5P4

A2W5P6

A2W5P10

Target range

A2W5P8

■ GO2050

A2W8P0

A2W8P2

A2W8P4

A2W8P6

A2W8P8

A2W8P10 **=**

Target range

==

Hai

==

A2W0P10

Target range

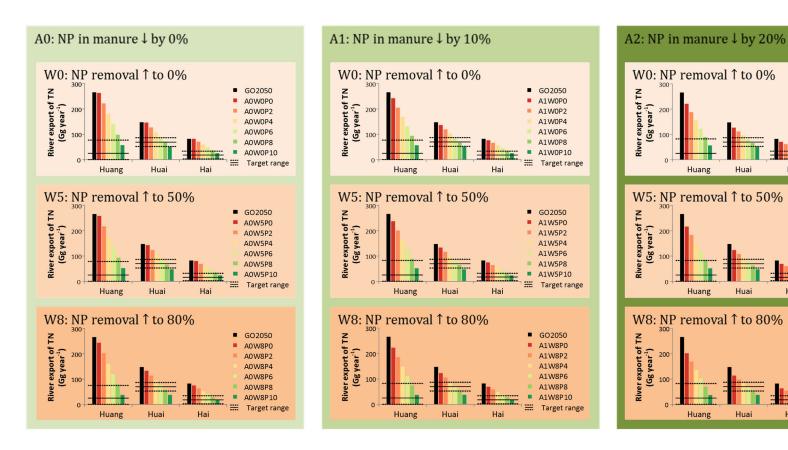


Figure 3.5 Total nitrogen (TN) from Huang, Huai and Hai basins to seas under 54 alternative scenarios. The graph shows the targets lines for TN inputs to seas when ICEP= 0 (with uncertainty range from -1 to +1) under Global Orchestration (GO) scenario. ICEP is the Indicator for Coastal Eutrophication Potential (see Section 3.2.4). GO assumes a globalized trends in socio-economic development with a reactive approach to manage environmental pollution (Seitzinger et al. 2010; Strokal et al. 2016a). The 54 alternative scenarios differ from the baseline (GO) as follows: lower N (nitrogen) and P (phosphorus) content of manure (indicated by A), higher N and P removal in wastewater treatment (indicated by W) and percentage of the point manure that is are applied on the land as fertilizer (indicated by P). A scenario consists of a combination of A, W and P. For instance: scenario A1W5P8 assumes a 10% reduction in the N and P content of manure, 50% of N and P removal rate in wastewater treatment, and 80% of point manure reused on land as fertilizer.

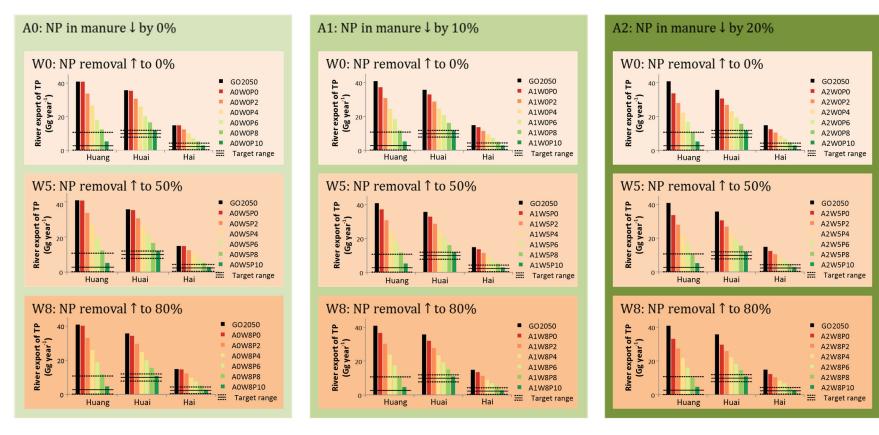


Figure 3.6 same as Figure 3.5 but for total phosphorus (TP).

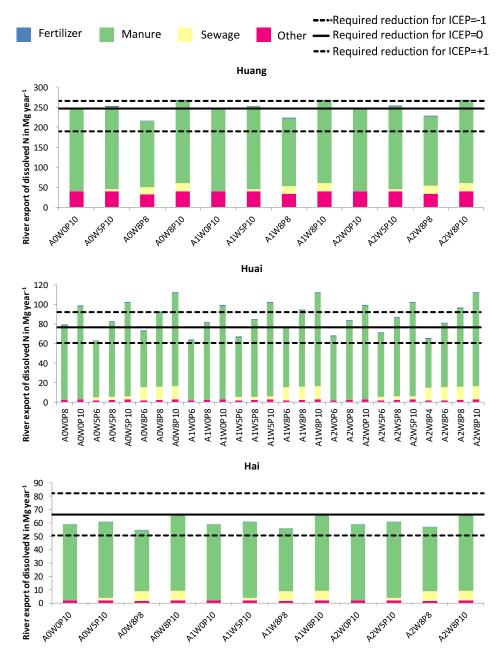


Figure 3.7 Achieved reduction in total nitrogen (TN) by sources under the selected alternative scenarios. These alternative scenarios reached the target of TN input to seas when ICEP $_N = +1$. Sources are classified into fertilizer, manure, sewage, and other. Fertilizer includes application of synthetic fertilizer. Manure includes animal manure on land and point manure. Sewage include N from wastewater treatment systems. Other includes fixation via crops, atmospheric deposition, weathering, and leaching of organic matters, application of human excretion on land, and direct discharge of human excretion to waters. The graph shows the targets for required reduction in TN inputs to seas when $ICEP_N = -1$, 0. +1 under Global Orchestration scenario (GO). ICEP is the Indicator for Coastal Eutrophication Potential (see details in Section 3.2.4). GO assumes a globalized trends in socio-economic development with a reactive approach to manage environmental pollution (Seitzinger et al. 2010; Strokal et al. 2016a). The alternative scenarios differ from the baseline (GO scenario) as follows: lower N content of manure (indicated by A), higher N removal in wastewater treatment (indicated by W) and percentage of the point manure that is are applied on the land as fertilizer (indicated by P). A scenario consists of a combination of A, W and P. For instance: scenario A1W5P8 assumes a 10% reduction in the N content of manure, 50% of N removal rate in wastewater treatment, and 80% of point manure reused on land as fertilizer. (see scenarios in green from Table 3.1).

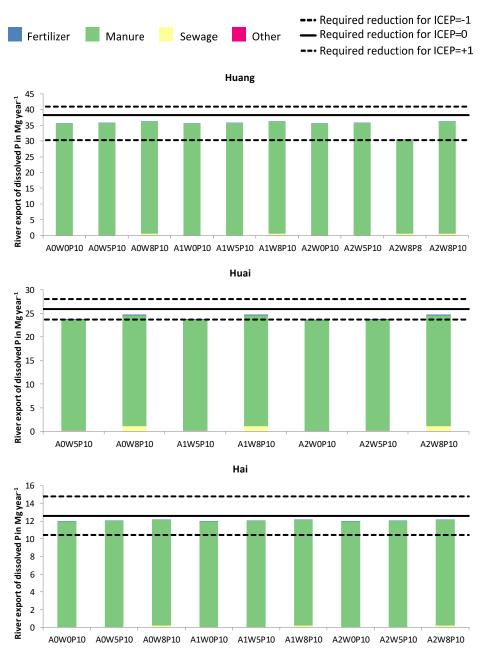


Figure 3.8 Achieved reduction in total phosphorus (TP) by sources under the selected alternative scenarios. These alternative scenarios reached the target of TP input to seas when ICEP_P = +1. Sources are classified into fertilizer, manure, sewage, and other. Fertilizer includes application of synthetic fertilizer. Manure includes animal manure on land and point manure. Sewage include N from wastewater treatment systems. Other includes weathering of P contained minerals, leaching of organic matters, P-based detergent use, application of human excretion on land, direct discharge of human excretion to waters. The graph shows the targets for required reduction in TP inputs to seas when ICEP_P = -1, 0. +1 under Global Orchestration scenario (GO). ICEP is the Indicator for Coastal Eutrophication Potential (see details in Section 3.2.4). GO assumes a globalized trends in socio-economic development with a reactive approach to manage environmental pollution (Seitzinger et al. 2010; Strokal et al. 2016a). The alternative scenarios differ from the baseline (GO scenario) as follows: lower P content of manure (indicated by A), higher P removal in wastewater treatment (indicated by W) and percentage of the point manure that is are applied on the land as fertilizer (indicated by P). A scenario consists of a combination of A, W and P. For instance: scenario A1W5P8 assumes a 10% reduction in the P content of manure, 50% of P removal rate in wastewater treatment, and 80% of point manure reused on land as fertilizer. (see scenarios in green from Table 3.1).

3.4 Discussion

3.4.1 Comparing with other studies

We quantified TN and TP inputs to seas by source using the MARINA 1.0 model. We found that over 60% of TN and TP in the studied rivers is in dissolved organic form. The dissolved organic forms of nutrients are mainly from agricultural activities, especially from the direct discharge of animal manure to rivers (point sources of manure) (Figure A.1 in Appendix II). This finding is in line with Strokal et al. (2016b); they indicate that the main source of nutrient losses to seas is animal manure (for most dissolved N and P forms) and synthetic fertilizers (for DIN) in China. Ma et al. (2010) also indicate that large amounts of nutrients are lost to the waters from agriculture in China.

Other models quantify nutrient inputs to waters at the global or continental scales, including our study area. For instance, Strokal et al. (2014), and Qu and Kroeze (2010, 2012) quantify nutrient inputs to seas from sixteen Chinese rivers using the Global NEWS model. The IMAGE-GNM (Integrated Model to Assess the Global Environment-Global Nutrient Model) also quantifies global annual river export of nutrients by sources (Beusen et al. 2015). These models calculated different (higher or lower) TN, TP, DIN, DON, DIP and DOP exports by the Huang, Huai and Hai rivers for 2000 than in our study. This can be explained by the fact that Global NEWS and IMAGE-GNM do not account for some important sources of nutrients in the Chinese rivers. The NUFER model quantifies nutrient use efficiencies and nutrient losses to waters at provincial and county levels (Ma et al. 2010; Wang et al. 2018b), but it does not specify nutrient losses to surface waters or ground water. The SWAT model analyzes the impact of agriculture on water quality considering physical processes in water and nutrient flows (Arnold et al. 2012). However, the model cannot quantify explicitly attribute sources of nutrients.

Strokal et al. (2016a) adopted the Global NEWS approach to calculate river export of nutrients to the MARINA 1.0 model. The model includes more pollution sources according to Chinese situation with updated information for reservoirs. For example, the model includes nutrients from direct discharge of animal manure and human excretion to rivers and uncollected human waste. Therefore, the modelled river export of nutrients to seas from the MARINA 1.0 model are higher or lower than the results from

the Global NEWS model. The MARINA 1.0 model runs at asub-basin level, while other models do not.

3.4.2 Uncertainties

We explored scenarios that may avoid coastal eutrophication in 2050. We are aware of the limitation of the MARINA 1.0 model. The model is a steady state model and it does not explicitly consider the dynamics in accumulation and release processes of N and P between soil and sediments, and waters (Strokal et al. 2016a). These dynamic processes in the soil can delay the effect of recycling of animal manure, especially for P in the soil because of the strong ability of P to accumulate. The process of converting P to the available inorganic P for crop production depends on many factors and one of these is including the amount of metal cation in the soil (Schachtman et al. 1998). We assumed that the river export of P will not be largely influenced by including the legacy of P (Strokal and de Vries 2012). This is because P in the rivers originated mainly from point sources compared to other sources (see Section 3.3.2). Furthermore, the performance of the MARINA 1.0 model has been evaluated by Strokal et al (2016a) and here we only briefly introduced the model performance in Section 3.2.6. The model was also applied in other studies and that gives trust in using the model for studying water pollution problems in the Chinese rivers and seas (Strokal et al. 2016b). We believe that the uncertainties of the MARINA 1.0 model will not largely change the main conclusion of our study.

Another source of uncertainties is related to ICEP. ICEP is an annual indicator for coastal eutrophication. It is based on the Redfield ratio of N:P:C:Si that is generic for the marine ecosystem. However, such ratio might also differ among the ecosystems in time and space. Other factors, such as water depth, light and water temperature may also be important. Nevertheless, we believe that ICEP provides a good basis for our study to explore options for water pollution reduction. It provides valuable insights in the impact of excess nutrients on marine ecosystems. More importantly, it is relatively easy to interpret, and can be combined easily with MARINA 1.0 to further explore solutions for coastal eutrophication. Our study is the first to use ICEP to quantify the annual targets for N and P inputs to seas. This allowed us to explore solutions to reach those targets. This is the relevant information for policy making to reduce coastal eutrophication.

Uncertainties in our results may also associate with the assumptions that we made for alternative scenarios. We found that in scenarios with relatively low percentages for recycling of animal manure (≤ 40% recycling rates, as in scenarios P0 and P20), the environmental targets are not reached, nevertheless, nutrient fluxes are considerably reduced. For example, N fluxes in Huai under the scenario A2W8P2 are considerably reduced, but just not enough to reach the environmental target (Table 3.1). We found that 40-80% of the manure (under the baseline scenario) that is directly discharged to rivers needs to be recycled on land to avoid coastal eutrophication. This implies that the nutrient cycles in food production in the NCP need to be closed. We showed that more circularity is technically possible, but we did not assess economic, social and institutional feasibilities. Closing nutrient cycles in food production requires adjustment in farming practices. It may also require promoting organic fertilizers, and training in application of organic fertilizers. Since 2015, the Ministry of Agriculture introduced a series of instruments to promote application of animal manure, such as subsidising the use of organic fertilizers and preferential policies for organic fertilizers production (MOA 2016). Meanwhile, the Ministry of Environmental Protection strengthens the monitoring and regulating the waste disposal of the industrialized animal farms. In addition, the Chinese government, universities and research institutes lunched the Scientific Backyards and Technology (STBs) platform that aims to educate farmers with farm- or region-specific knowledge for agricultural practices. By 2015, there are 72 STBs that cover 21 provinces in China functioning (Chen et al. 2014; Chen et al. 2006; Li et al. 2017a; Zhang et al. 2016b). The Chinese government recognized the urgency of reducing water pollution from agricultural activities. Therefore, we assumed that the recently introduced environmental policies will strongly facilitate the recycling of manure on land. Thus, achieving the 40-100% recycling of animal manure on land in 2050 might be technically possible.

We assumed improvements in N and P removal during treatment in sewage systems in our alternative scenarios. Our assumptions are based on conventional technologies that have been applied in, for instance, Europe and the US. We realise that reaching the 80% of N and P removal might be difficult, but possible by 2050 taking into account the fast technological development (Kartal et al. 2010; Khiewwijit 2016; Shi et al. 2010; Winkler et al. 2012).

We also made assumptions on reducing N in animal excretion by improving animal feed. Currently, mixed farming systems and industrialized animal farms are responsible for 90% of animal production in China (Li et al. 2008). Mixed farming systems are mostly small holders, and feed sources of the mixed farming systems mainly rely on crop residues and grazing. Promoting better quality of animal feed may require educating small holder farmers. But, in the recent decade, mixed farming systems are transferring to more industrialized farms to increase production and to save cost on resource use (Li et al. 2008). Thus, promoting the use of animal feed with better quality is a promising trend. We believe it is possible to reach the 20% reduction in N and P in animal manure in 2050 based on experimental studies (Bai et al. 2014; Lesschen et al. 2011).

3.5 Conclusion

We quantified total nitrogen (TN) and total phosphorus (TP) export from the Huang, Huai and Hai to the Bohai Gulf and Yellow Sea in the North China Plain (NCP) for the year 2050. We aimed to explore possibilities to avoid coastal eutrophication in the NCP. For this, we set environmental targets for river export of nutrients, such that risks for coastal eutrophication are low, based on the Indicator for Coastal Eutrophication Potential (ICEP). Then we calculated the maximum TN and TP fluxes from the Huang, Huai and Hai rivers. We compared the maximum levels to the baseline scenario (Global Orchestration (GO)). Finally, we developed 54 alternative scenarios to explore which combination(s) of options can lead to low eutrophication risks ICEP=0, (-1 to +1). These options focus on recycling manure to replace synthetic fertilizers, improving animal feed and increasing nutrient removal rate in wastewater treatment systems.

In our baseline scenario, rivers of the NCP are projected to export much more nutrients to the seas in 2050 than in the past, causing more eutrophication. The Huang River is projected to export around 270 Gg of TN and 40 Gg of TP, which is more than the Huai and Hai river in 2050. The Huai River is projected to export around 150 Gg of TN and 35 Gg of TP. For the Hai River, these values are 80 Gg for TN and 15 Gg for TP.

Avoiding coastal eutrophication may be possible by 2050, if export of N and P by the Huang, Huai and Hai rivers is reduced by 50-90% relative to the baseline scenario. These reductions are especially needed for dissolved inorganic and organic forms of N and P because of their dominant share in the total N and P in the rivers. Avoiding future

coastal eutrophication is technically possible with different combinations of management options. We identified 12 scenarios for N and 10 scenarios for P in which the risk on coastal eutrophication is low for the Huang River. For the Huai River, the target can be reached in 25 scenarios for TN and 7 scenarios for TP. For the Hai River, we identified 9 and 14 scenarios reaching targets for N and P, respectively.

Recycling manure on land to replace synthetic fertilizers is essential to avoid coastal eutrophication. In the scenarios that achieved the targets, 40 to 100% of this manure is assumed to be recycled on land. This consequently reduces the use of synthetic fertilizers in the scenarios. In the Huang basin, at least 80% of point source manure (TN and TP) is recycled in these scenarios; in the Huai basin at least 40% of TN and 100% of TP, and in the Hai basin at least 80% for TN and 100% for TP. Our results also show that reducing TP fluxes is more difficult than reducing TN. This may be associated with the sources of TP fluxes. Not only animal manure, but also sewage is an important source of TP in the rivers. Hence, the P removal rate in sewage systems is already at a relative high level under the baseline scenario.

Our study explores scenarios to avoid future eutrophication in Chinese seas. We show how important it is to recycle animal manure in the North China Plain. Replacing synthetic fertilizers by recycled manure is essential in scenarios to avoid future coastal eutrophication. Our study also shows how back-casting exercises can be used to explore desired futures with low risks for coastal eutrophication, and possibilities to reach such futures. This may help policy-makers in developing effective environmental policies.

Acknowledgements

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Chapter 4

Equality in River Pollution Control in China

Abstract

Water pollution is a serious problem in China. This study focuses on equality in pollution control in the Yangtze, Yellow and Pearl. We first quantified environmental targets for nitrogen (N) and phosphorus (P) at the river mouth. We used the Indicator for Coastal Eutrophication Potential and the Model to Assess River Inputs of Nutrients to seAs (MARINA) to project river export of nutrients. Next, we allocated the environmental targets to sub-basins as allowable levels, based on a Gini optimization approach. We searched for minimum inequality in pollution per unit of GDP, population, basin area, and agricultural area. Our results indicate that without pollution control, the river export of nutrients in 2050 exceed allowable levels. To meet the allowable levels while striving for equality, total dissolved N and P exports from sub-basins need to be reduced by 60 to 97%. The required reductions are largest for sub-basins of the Yellow River. For P, reducing point source inputs to rivers (manure and sewage) may be enough to avoid that allowable levels are exceeded in many sub-basins. For N, more needs to be done. Some sub-basins need to reduce their pollution more than others. Equality considerations call for reducing both point (e.g. recycling manure resources on the land) and diffuse (improve nutrient use efficiencies in agriculture) sources of N in the rivers. Our study is the first to link a Gini based optimization approach with the MARINA model. It may support decision making aimed at cleaner production and at equality in pollution control.

Under revision as:

Ang Li, Qiang Yuan, Maryna Strokal, Carolien Kroeze, Lin Ma, Yi Liu. Equality in River Pollution Control in China. This manuscript is under revision after positive review comments. To be resubmitted soon to the Science of the Total Environment

4.1 Introduction

Water pollution and eutrophication along Chinese coasts have been threatening marine ecosystems and human health for decades (Strokal et al. 2016a; Tang et al. 2006; Tong et al. 2016). In the 2000s, harmful algae blooms developed about 60 to 120 times per year along the Chinese coasts (SOA 2010). An important reason for coastal eutrophication are increasing nutrient losses from food production and other human activities (Strokal et al. 2016a; 2017). To avoid this in the future, nutrient export by Chinese rivers needs to be reduced from anthropogenic activities.

Several assessments of technologies or management strategies to reduce future nutrient losses to rivers and thus coastal eutrophication exist. Strokal et al. (2017) explored optimistic futures to reduce river export of nutrients from human activities by 2050 with the MARINA model (Model to Assess River Inputs of Nutrients to seAs). These optimistic futures assumed implementation of best available technologies in sewage and agriculture to avoid future river pollution in 2050. Wang et al. (2018a) explored nutrient management options to increase nutrient use efficiencies in agriculture and thus to reduce nutrient losses to the Chinese rivers. Li et al. (2017a) analyzed the effect of "Double High Agriculture" on reducing river export of nutrients to seas for China. Li et al. (2019a) explored management options in agriculture and sewage to reach allowed nutrient levels at the river mouths in China by 2050. Maximum nutrient levels were calculated based on ICEP (Indicator for Coastal Eutrophication Potential); ICEP can be used to identify pollution levels consistent with low risks for coastal eutrophication (Garnier et al. 2010; Li et al. 2019a).

However, these existing studies hardly consider equality when designing nutrient pollution reduction strategies for Chinese river basins. Clearly, some sub-basins contribute more to pollution at the river mouth than others. But is it fair to let these regions invest most in pollution reduction? Or should we aim for equality in per capita pollution levels? Or should we strive for equal pollution levels per unit of GDP? In air pollution control some of these questions are answered: European air pollution control, for instance, aims at least-cost solutions (Amann et al. 2011; Kiesewetter et al. 2014). Other studies allocated nutrient pollution permits using, so called, the grandfathering approach (Wang et al. 2016). Equality in pollution reduction has been considered in

economic studies (Biancotti 2006; Lambert 1985; Rahman et al. 2009), but hardly for large Chinese rivers.

Equality in pollution control could be based on region-specific pollution reduction targets, since sub-basins of large rivers differ in socio-economic characteristics (e.g., GDP, population, agriculture). Equality considerations may thus imply that allowable pollution levels differ among sub-basins. For example, if we aim for equal pollution per GDP and person, rich and populated sub-basins may be allowed to discharge more nutrients than sub-basins with poor economies and a smaller population. Such considerations, however, hardly play a role in recent studies on reducing nutrient pollution in Chinese rivers.

One way to account for equality in allocating pollution permits is based on Gini optimization. The Gini coefficient is a widely used measure of social inequality in a region (see Section 4.2). Gini optimization was done for some individual, small watersheds or cities (Sun et al. 2010; Wang et al. 2016; Yuan et al. 2017). For example, Yuan et al. (2017) applied Gini optimization to equally allocate discharge permits of waste water among cities in Jiangsu province in China. Such approaches can also be applied in studies on reducing coastal eutrophication. This could be done by defining allowable N and P loads at the river mouth consistent with low risks for coastal eutrophication (Li et al. 2019a). Large rivers such as the Yangtze, Yellow and Pearl export are dominant contributors to N and P in the coastal waters in China (Strokal et al. 2016a; 2017). So far, however, no studies exist that allocate allowable nutrient pollution levels to sub-basins of these large rivers while considering equality.

This study focuses on equality in pollution control in the Yangtze, Yellow and Pearl. We quantify desired nutrient pollution levels for sub-basins of these three rivers from both an environmental and equality point of view. Thus the targets ensure low risks of coastal eutrophication, while considering socio-economic equality. We allocate nutrient pollution levels to sub-basins, while aiming for equality in pollution per unit of land, agricultural land, income, or population. To this end, a Gini optimization approach will be applied. Our study is the first to link a Gini based optimization approach with the MARINA model. In the following, we first, we define allowable nutrient pollution levels, based on the ICEP indicator. Next, we used the Gini optimization approach to allocate allowable nutrient pollutions levels to sub-basins (more details in Section 4.2).

4.2 Material and Methods

4.2.1 Allocating allowable nutrient pollution levels to sub-basins

We quantify the allowable nutrient pollution levels for sub-basins in three steps (Figure 4.1).

The first step is to quantify allowable nutrient pollution levels for the Yellow (Huang He), Yangtze (Chang Jiang), and Pearl (Zhu Jiang) rivers at the river mouth. Allowable nutrient pollution levels are calculated from river exports of N and P to seas that ensure low risks for coastal eutrophication. These allowable levels are calculated based on the Indicator for Coastal Eutrophication Potential (ICEP) approach (Garnier et al. 2010). A detailed explanation on this approach is explained in Section 4.2.3.

The second step is to allocate the allowable nutrient pollution levels for these three large rivers to their sub-basins, based on a Gini optimization approach searching for solutions with lowest inequality. We used Gini to minimize inequality in pollution per unit of GDP, population, basin area, and agricultural area. These four indexes are chosen to represent the socio-economic development of the sub-basins. Details on the indexes can be found in Section 4.2.4. The Gini coefficient approach is often used to represent equalities according to the Lorenz curve (Bosi and Seegmuller 2006; Lambert 1985; Rahman et al. 2009). We first calculate the Gini coefficients per unit of GDP, population, basin area, and agricultural area for sub-basins in large rivers and for the year 2050. Then we allocate allowable pollution levels while minimizing the Gini coefficients (to smaller or equal to 0.3) for each index. Finally, we set the constraint on minimizing the sum of the Gini coefficients for four indexes. The calculation is further explained in Section 4.2.4.

The third step is to quantify the required reduction in nutrient pollution per sub-basin. This is done by quantifying the gap between the actual river export of nutrients and the allowable nutrient pollution levels for each sub-basin. To quantify the gap, we ran MARINA 1.0 to quantify the actual river export of nutrients and then compared this with the allowable nutrient pollution level. This indicates the required reduction for each sub-basin. The description of MARINA 1.0 can be found in Section 4.2.3.

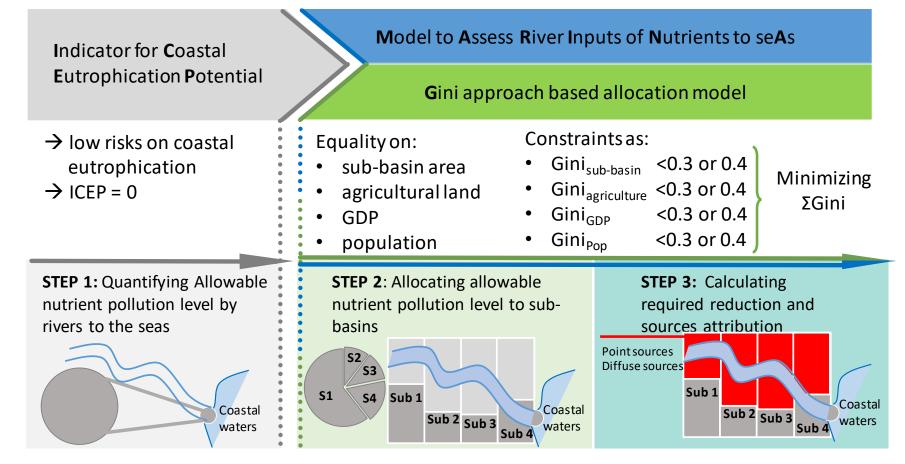


Figure 4.1 Schematic overview of allocation of allowable nutrient pollution levels.

4.2.2 Study Area

We focus on three large rivers in China: the Yellow, Yangtze and Pearl. These rivers cover a large part of China (Strokal et al. 2016a). In the MARINA model, the drainage area of the Yellow River is divided into six sub-basins draining into the Bohai Gulf. The Yellow is a relatively dry river, compared to other rivers in China. The drainage areas of the Yangtze and Pearl are relatively wet and are divided into ten and six sub-basins, respectively. Yangtze flows into the Yellow Sea and Pearl flows into the South China Sea (Figures A.1 and A.2 in Appendix III).

4.2.3 MARINA model and ICEP

We applied the Model to Assess River Input of Nutrients to seAs (MARINA 1.0) model to quantify annual N and P export by Yellow, Yangtze and Pearl rivers at a sub-basin scale. The model is developed based on the Global Nutrient Export by WaterShed (NEWS-2) model (Mayorga et al. 2010). MARINA 1.0 quantifies river export of dissolved inorganic (DIN, DIP) and dissolved organic (DON, DOP) N and P for the years 1970, 2000 and 2050 for six large Chinese river basins including Liao, Yellow, Hai, Yangtze, Huai, and Pearl. These six large basins are divided into 25 sub-basins. The sub-basins are classified into the up-, middle- and downstream. Nutrients travel generally longer from upstream than from middle- and downstream to seas. The river export of nutrients is quantified as a function of human activities on land (e.g., crop and animal production, urbanization), sub-basin characteristics (e.g., land use, hydrology) taking into account retentions of nutrients on land and in rivers (Strokal et al. 2016a).

River export of N and P is calculated by point sources and diffuse sources. Point sources include sewage effluents (they contain also detergents for P), and direct discharges of animal manure and human waste (untreated) to rivers. The model accounts for N and P removal in waste water treatment systems. Animal manure that is directly discharged to rivers is quantified from the total animal manure excretion, multiplied by a modelled fraction that directly discharged to rivers. A similar approach is used to quantify N and P losses from uncollected human waste (untreated) (Strokal et al. 2016a). For diffuse sources, the model considers atmospheric N deposition, biological N fixation, and weathering of P-contained minerals, application of synthetic fertilizers, animal manure

and human waste on land. In agricultural land, some N and P can be exported from the system through animal grazing and crop harvesting. Retentions of N and P in soils are taken into account when quantifying N and P inputs to rivers. Retention on land is modelled as a function of annual runoff from land to streams. Leaching of organic matter is another diffuse source for DON and DOP in rivers (Strokal et al. 2016a).

Some nutrients may retain or be lost from the river network. For example, water consumption can remove nutrient from the rivers (generic for nutrient forms. Denitrification (for DIN) and sedimentation (for DIP) processes can contribute to DIN and DIP losses and retentions in rivers. Dammed reservoirs along the river network can serve as sinks and retain nutrients (for DIN and DIP) in rivers. These retentions and losses influence river export of the nutrients and are considered in the MARINA 1.0 model.(Strokal et al. 2016a) MARINA 1.0 uses information on dams from the Global Reservoir and Dam Database (Lehner and Döll 2004; Strokal et al. 2016a).

The performance of MARINA 1.0 was assessed in earlier studies (Strokal et al. 2016a; Strokal et al. 2017). MARINA 1.0 was evaluated in different ways (see details in Strokal et al., 2016). Model validation indicates that the modelled annual river export of N and P for Chinese rivers is generally in line with the available measurements (e.g., R2 is 0.84, see Strokal et al. (2016a)). In addition, sensitivity analyses reveal that the calculated river export of nutrients is relatively sensitive to the following parameters: animal manure production, the fraction of animal manure that is directly discharged to rivers, application of synthetic fertilizers on land, and river discharge. An important parameter is the fraction of animal manure that is directly discharged to rivers. These fractions, and other data on animal production, were derived from the Nutrient Flows in Food chains, Environment and Resources use (NUFER) model (Ma et al. 2010). NUFER was developed for China based on field survey of 50 thousand farms from 1999 to 2008. Several other model inputs were from the gridded database of the Global NEWS-2 model. The model inputs and results of MARINA 1.0 were evaluated by comparing them with an independent dataset (the Chinese Statistical Year Book), and by comparing with other modelling studies. Results of all these options gain trust in the model performance (see Strokal et al. (2016a)). In this study, we use MARINA 1.0 in combination with the Gini optimization (Section 4.2.4) and ICEP (see below) approaches.

In this study, we quantify the river exports of nutrient for the year 2050 under the Global Orchestration (GO) scenario from the Millennium Ecosystem Assessment (MA). The GO scenario reflects a world that is globalized, and in which society has a reactive attitude towards environmental management (Alcamo et al. 2005b). This scenario has been used as a baseline in recent analyses (Strokal et al. 2017). In GO, the Chinese population will increase but the growth rate will be moderate. GO assumes a rapid urbanization, increasing income and a high purchasing power. The inequality of income will be reduced. People will consume more protein-rich food and thus result in an increasing animal production, and generating more human waste (Bouwman et al. 2009). The agricultural land area will grow and synthetic fertilizer use will increase. Water consumption will increase because of the rapid urbanization and economic development. New technologies will be applied to increase the N removal rate in the wastewater treatment plant. But people will generate more wastewater because of the increasing human consumption (Seitzinger et al. 2010; Van Drecht et al. 2009).

In this paper, we use the Indicator for Coastal Eutrophication Potential (ICEP) to represent the risks for coastal eutrophication (Billen and Garnier 2007; Garnier et al. 2010). ICEP is quantified considering the N and P fluxes and the requirements of the diatoms based on the Redfield ratio (C:N:P:Si =106:16:1:20; eq.1-2). ICEP values are expressed in kg C km⁻² day⁻¹. The risks is high when ICEP values are positive (>0). This indicates that N and P fluxes are in excess over Si (silica) fluxes. As a results, the ratio of N:P:Si is disturbed and non-siliceous diatoms (e.g. harmful algae like cyanobacteria) may develop instead. ICEP is quantified following equations1 and 2 (Garnier et al. 2010):

$$ICEP_N = [N/(14 \times 16) - Si/(28 \times 20)] \times 106 \times 12$$
, IF N:P<16 (N limiting) eq. 1
 $ICEP_P = [P/31 - Si/(28 \times 20)] \times 106 \times 12$, IF N:P>16 (P limiting) eq. 2

Where:

ICEP_N: Indicator for Coastal Eutrophication Potential when nitrogen is limited (kg km⁻² day⁻¹);

ICEP_P: Indicator for Coastal Eutrophication Potential when phosphorus is limited (kg km⁻² day⁻¹);

N: river export of nitrogen to seas (kg km⁻² day⁻¹).

P: river export of phosphorus to seas (kg km⁻² day⁻¹).

Si: river export of dissolved silica (Si) to seas (kg km⁻² day⁻¹).

Equations 1 and 2 can be used to derive the maximum river export of N (N_{max}) and P (P_{max}) to seas to ensure low risks for coastal eutrophication (equations 3 and 4). This is what we consider the allowable nutrient export at the river mouth. We calculate the allowable nutrient pollution levels at river mouth as:

$$N_{max} = {ICEP_N / (106 \times 12)} + {Si / (28 \times 20)} \times (14 \times 16)$$
 eq. 3

$$P_{max} = {ICEP_P / (106 \times 12)} + {Si / (28 \times 20)} \times 31$$
 eq. 4

4.2.4 Multi-index Gini coefficient method

4.2.4.1 Multi-index Gini coefficient

The Gini coefficient is an indicator to measure social inequality in a region according to the Lorenz curve (Bosi and Seegmuller 2006; Lambert 1985; Rahman et al. 2009). Figure A.3 (in Appendix III) shows a graphical representation of the Lorenz curve. In the Lorenz curve, A is the area between the Lorenz curve and the line of equality (45 degree), and B is the area below the Lorenz curve. The Gini coefficient is the ratio of area A to the total area of A and B.(Biancotti 2006; Cullis and Koppen 2007; Kleiber and Kotz 2002; Moyes 2007; Xu 2004) The value of the Gini coefficient is between zero and one. When the radian of the Lorenz curve is higher, the equality is lower. A Gini coefficient close to one implies absolute inequality. A Gini coefficient close to zero indicates absolute equality. Internationally, 0.4 is generally considered as a threshold above which an inequality risk is observed (Xiao et al. 2009).

We applying the Gini coefficient to allocate the allowable nutrient export, using four indexes on the x-axis and the allocated nutrient export by rivers on the y-axis in the Gini diagram. Based on this, the Gini coefficient for each index can be calculated.

Our multi-index Gini optimization approach for river export of nutrients in 2050 thus applies the following equations:

Optimization object: $\min \sum_{j=1}^{n} G_j$ eq. 5

$$G_i = 1 - \sum_{i=1}^{n} (X_{i(i)} - X_{j(i-1)})(Y_i + Y_{i-1})$$

eq. 6

Constraints:

Total amount of allowable nutrient export by rivers to the Chinese Seas:

$$W = \sum_{i=1}^{n} S_i$$
 eq. 7

Constrain for each Gini coefficient:

$$G_i \leq G_w$$
 eq. 8

Order:

$$K_{j(i)} = \frac{S_i}{M_{j(i)}}$$
 eq. 9

$$K_{j(i-1)} \le K_{ji} \le K_{j(i+1)}$$
 eq. 10

Where:

G: the Gini coefficient;

j: the indexes of the Gini coefficient, indexes include sub-basin area, agricultural land area, GDP and population;

i: the sub-basins in the river;

G_i: Gini coefficient for index j after the optimization;

G_w: the warning line (threshold) for the optimized Gini;

W: the allowable nutrient pollution level for river exports of total dissolved N and P (TDN and TDP) at the river mouth;

 $X_{j(i)}$: the accumulative percentage of index j of the ith sub-basin;

Y_i: the accumulative percentage of the allowable nutrient export from the ith sub-basin;

S_i: the allowable nutrient pollution level for river exports of TDN and TDP by the ith subbasin to seas;

 $M_{j(i)}$: the value of index j in the ith sub-basin;

 $K_{j(i)}$: the allowable nutrient export per unit of index j in i^{th} sub-basin after the optimization.

4.2.4.1 Index description

Four indexes are selected for the Gini optimization to reflect socio-economic equity in the sub-basins of Yangtze, Yellow and Pearl rivers. Two of these indexes are the subbasin area and agricultural land area. These two indexes represent the agriculture development of the sub-basins. They are related to the potential for future urbanization and diffuse pollution, respectively. Thus, minimizing the Gini coefficient with respect to sub-basin area and agricultural land area index would guarantee that the allocation of allowable pollution level for river exports of total dissolved N (TDN) and P (TDP) is proportional to the magnitude of the development potential and diffuse source scale from agricultural activities (Yuan et al. 2017). Population and income (GDP: gross domestic products) are selected as two other indexes to reflect socio-economic equity. These consider that each person has an equal right to pollute. To guarantee equity among people in affecting coastal water quality, the Gini coefficient is minimized by population. GDP is frequently used in existing studies to reflect the economic development. We consider that the strict nutrient pollution control may negatively affect the economic development. The local government would like to get more allowable pollution rights to maintain the economic development. The Gini coefficient with respect to GDP can ensure that the region with higher GDP can get more allowable pollution level, thus resulting in an equal fact that the impact level on the water quality of the sea per unit GDP among the sub-basins is rather similar. In this study, the data for the GDP, population, basin area, and agricultural area are from the MARINA 1.0. We used the gridded GDP data for China from Huang et al. (2014) to generate the sub-basin specific GDP.

4.3 Results

4.3.1 Allowable nutrient pollution levels

We describe the results for TDN and TDP river exports. Total dissolved N (TDN) includes river export of dissolved inorganic and organic N (DIN and DON). Total dissolved P (TDP) consists of river export of dissolved inorganic and organic P (DIP and DOP). We first present the allowable nutrient export by rivers that is quantified based on the ICEP

approach (Section 4.3.1.1). Next, we present the allocated allowable nutrient export by rivers to the sub-basins based on the Gini optimization approach. We compare the results with the actual river export of nutrients by sub-basin from the MARINA 1.0 model (Section 4.3.1.2). In this way, we identify the required reduction to meet the allowable river export of nutrients by sub-basin. Our analysis is for the year 2050.

4.3.1.1 Allowable nutrient export by rivers to seas

Results show that river export of TDN and TDP at the river mouth largely exceed the allowable levels (Table A.1 in Appendix III). For Yangtze, the allowable river export of TDN is 395 Gg year-1 and of TDP is 55 Gg year-1 (Table A.1). For the Yellow River, the allowable river export is 20 Gg year-1 for TDN and 3 Gg year-1 for TDP. And for the Pearl River these levels are 88 Gg of TDN and 12 Gg of TDP (Table A.1 in Appendix III). These levels are all considerably lower than the nutrient export by the rivers as calculated for 2050 in the GO scenario. More than 90% of TDN and TDP river export needs to be reduced to meet the allowable pollution level for the Yellow River. For Yangtze, about 75-80% of river export of TDN and TDP needs to be reduced, and for the Pearl River more than 85%.

4.3.1.2 Equality in river pollution control: required reductions by sub-basin

Using the allowable nutrient export at the river mouth as a starting point, we quantify required pollution reductions at the level of sub-basins while accounting for equality. The required reductions for sub-basins follow from the Gini optimization approach (Gini constraints at below 0.4, see Figure A.5 in Appendix III) and are presented in Figures 4.2, 4.3, A.4 and Table A.1 in Appendix III.

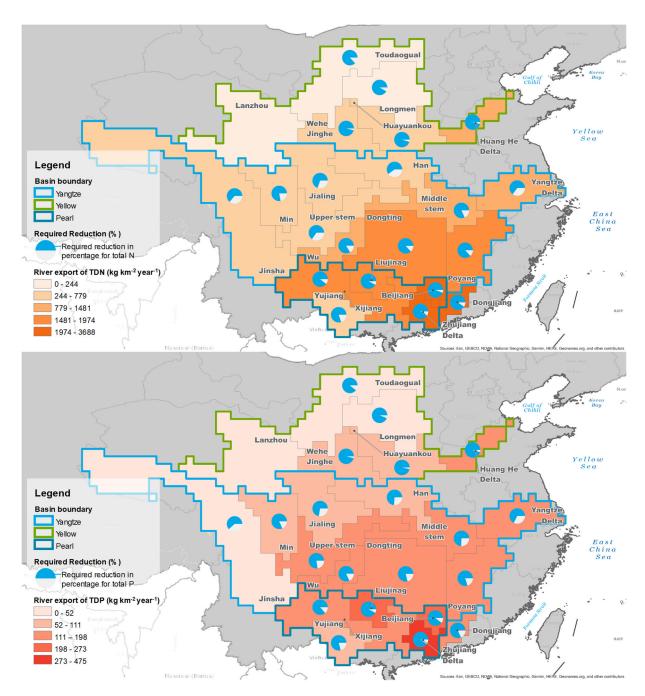


Figure 4.2 River export of total dissolved nitrogen (TDN) and phosphorus (TDP) from sub-basins of Yellow, Yangtze and Pearl rivers (kg km-2 year-1) and the required reductions to meet the nutrient allowable levels to avoid coastal eutrophication in 2050 (%, blue parts in the pies). See Section 4.2 on details for the methodology.

In the GO scenario for 2050, the calculated 2050 river export of TDN and TDP increases from up- to downstream areas. For instance, the downstream sub-basins of the Yellow River export 1,974-3,688 kg km⁻² year⁻¹ of TDN and 273-475 kg km⁻² year⁻¹ of TDP to the coastal waters in 2050. This is high compared to the other sub-basins of the Yellow River. The required reduction rates also increase from up- to downstream sub-basins of the Yellow River (Figure 4.2). Each sub-basin needs to reduce 93% of TDN and TDP to meet the allowable nutrient levels (Figures 4.2 and A.4 in Appendix III). The largest reduction for TDN is calculated for middlestream Wehe Jinghe and the downstream Yellow River Delta (96%) and for TDP for the middle-stream Huayuankou (97%) (Figures 4.2 and A.4 in Appendix III).

For the Yangtze River we calculate that middle- and downstream sub-basins generally export more TDN and TDP than upstream sub-basins to the coastal waters in 2050 in GO (Figure 4.2). The river export of TDN ranges from 244 to 1,481 kg km⁻² year⁻¹ among the upstream sub-basins Jinsha, Ming, Jialing, Wu and Upper-stem (Figure 4.2). For middle- and downstream sub-basins this range is 244- 1,974 kg km⁻² year⁻¹. Upstream sub-basins are projected to export 0-198 kg km⁻² year⁻¹ of TDP, and middle- and downstream sub-basins 52-198 kg km⁻² year⁻¹ (Figure 4.2). To meet the allowable nutrient pollution levels, 64-84% of river export of TDN and 62-82% of river export of TDP need to be reduced from upstream sub-basins (Figures 4.2 and A.4 in Appendix III). For the middle- and downstream sub-basins, these reductions are 60-87% for TDN and 68-80% for TDP (Figures 4.2 and A.4 in Appendix III). The largest required reduction for TDN is calculated for Dongting and Poyang (87%) (middle-stream sub-basin). For TDP, this is for Wu and Min (82%) (upstream sub-basin, Figures 4.2 and A.4 in Appendix III).

Results indicate that the TDN and TDP export by the Pearl River is generally higher than that of the Yellow River and Yangtze River. This can be explained by the fact that the two downstream sub-basins (Dongjiang and Zhujiang Delta) are projected to export the highest amount of TDN and TDP in 2050 in GO. Reasons for the high river export of TDN and TDP by Dongjiang and Zhujiang Delta are associated with human activities and hydrology. The drainage area of the Pearl River is generally much wetter than the drainage areas of the other rivers in China. This may lead to more nutrients in the coastal waters. Zhujiang Delta is an urbanized, downstream sub-basin and thus may export more nutrients. To meet the allowable nutrient pollution levels, river export of

TDN (80 to 94% for sub-basins) and TDP (79 to 92% for sub-basins) by Pearl River from sub-basins is required to be reduced. These required reductions for sub-basins of the Pearl River are generally lower than for the sub-basins of the Yellow River.

The percentage reduction in river export of TDN and TDP by sub-basins when we consider both equality and environment in pollution control (optimized target with minimum inequality) is different from it when only environment is considered (non-optimized target). And the differences between the non-optimized target and optimized target can be found in all three rivers for both TDN and TDP (Figure 4.3). For example, to reach the non-optimized target of river exported TDN, all sub-basins in the Yangtze River requires to abate at least 78% of TDN. However, considering the equality in pollution control, the percentage reduction for TDN can be ranged from 60% (in Han sub-basins) to 87% (Dongting and Poyang) (Figure 4.3). This implies that some sub-basins, such as Poyang, need to take more responsibility than others, such as Han, to control the TDN pollution in the big rivers when socio-economic equality is considered.

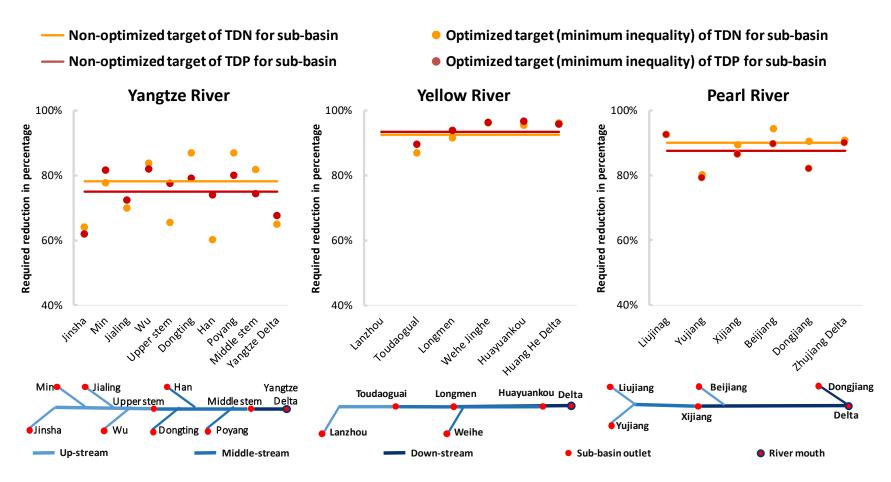


Figure 4.3 Percentage reduction in river export of total dissolved nitrogen (TDN) and phosphorus (TDP) to the river mouth from sub-basins of the Yangtze River, Yellow River and Pearl River. Lines refer to the non-optimized target for TDN and TDP. Dots refer to the optimized target (minimum inequality) for TDN and TDP.

4.3.2 Required reduction to meet the allowable nutrient pollution levels

To identify the opportunities to meet the allowable nutrient pollution levels, we compared the required reduction rates with the source attribution on river export of nutrients to seas (Figure 4.4). The source attribution differs largely among the subbasins and nutrient forms. Therefore, the opportunities to reach the allowable nutrient pollution level also differs among sub-basins and nutrient forms.

To reach the allowable pollution levels of TDN for sub-basins in the Yangtze River, controlling both point and diffuse sources is necessary. The required reduction for TDN river export from five out of ten sub-basins of the Yangtze River can be achieved by reducing pollution from the point source (Figure 4.4). These five sub-basins are Jinsha (upstream), Jialing (upstream), Upper stem (upstream), Han (middle-stream), and Delta (downstream). For the other five sub-basins of Yangtze, required reductions for TDN can be achieved by reducing from both point and diffuse sources. For the Yellow River, reducing TDN from point sources may reach the allowable pollution level for most sub-basins. Exceptions are the Wehe Jinhe and Huang He Delta sub-basins where reducing TDN from point source may not be enough. Thus reducing TDN from diffuse sources in those sub-basins may also be needed to reach the allowable pollution level for TDN (Figure 4.4). For the Pearl River, results indicate that both point and diffuse sources are important to reach the allowable pollution level for TDN river export (Figure 4.4).

For TDP reducing point source pollution may be essential to meet the allowable pollution level. In the Yangtze River, the required reduction for TDP can be reached by only reducing point sources (Figure 4.4). This holds for most sub-basins in the Yellow River and Pearl River. An important reason is that TDP is projected to export dominantly from point sources in 2050. For example, 80 -100% of TDP river export is projected from point sources (sewage and animal manure discharge to rivers as point source) for all sub-basins of three rivers in 2050. The contribution of point sources is generally larger than the required reduction rate of TDP (from 62 to 97% for most sub-basins). There are exceptions for several sub-basins: Poyang (middle-stream) in Yangtze, Liujiang (upstream), Beijiang (middle-stream) and Zhujiang Detal (downstream) in

Pearl River. Reducing TDP export from those sub-basins may require efforts in reducing from both point and diffuse sources (Figure 4.4).

We also calculated the nutrient input to rivers and then compared with the allowable nutrient input to rivers (Figure A.4 and Table A.2 in Appendix III). We found the share of point source is larger than the required reduction rate in two out of ten sub-basin in Yangtze, three out of six sub-basin in Yellow. For the N input to seas, the share of point source is larger than the required reduction rate in five out of ten sub-basins in Yangtze, four out of six sub-basins in Yellows. This is because that nutrient can be retained in the sediments of the river or lost (through denitrification or water withdraw) when it travels to the coast.

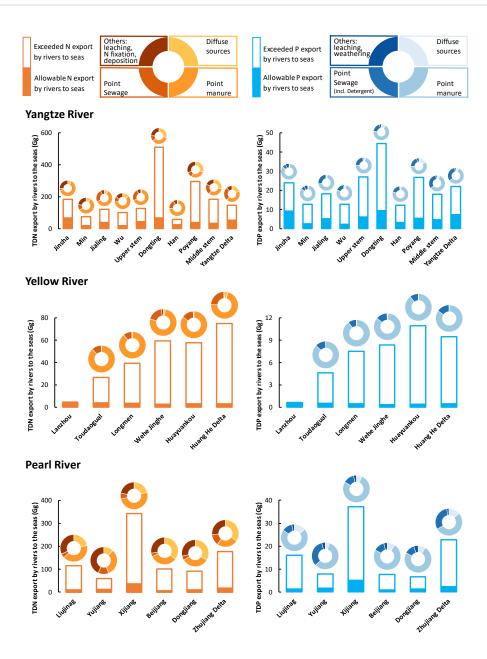


Figure 4.4 Total dissolved nitrogen (TDN) and total dissolved phosphorus (TDP) export by Yangtze, Yellow and Pearl rivers by sub-basins. The bars indicate the total TDN and TDP river export and the allowable levels (Gg year⁻¹). The pies show the relative shares of sources in the river export of nutrients. Diffuse sources include applied synthetic fertilizers, animal manure on land, uncollected human waste. Point sources include manure discharges to rivers as point sources (point manure in the legend), human waste discharges to rivers (untreated) and sewage effluents (treated) (indicated as point sewage in the legend). Other sources include leaching, biological N fixation, atmospheric deposition (for N), weathering of P-contained minerals (for P). Source: MARINA model output. See Section 4.2 on details for the methodology.

4.4 Discussion

Comparison with other modelling studies. In this study, we combined the ICEP approach and Gini optimization with the MARINA model to allocate allowable nutrient pollution levels to sub-basins of Yangtze, Yellow and Pearl rivers considering socioeconomic equality. This is the first study using this combined approach. Previous modelling studies on nutrient pollution in China mostly focused on quantifying the nutrient export by rivers, nutrient sources and/or exploring the effect of nutrient management strategies on nutrient pollutions. For example, the Soil and Water Assessment Tool (SWAT) model has been widely applied to Chinese rivers, lakes or reservoirs to quantify nutrient losses from diffuse sources (Li et al. 2009; Yang et al. 2008). The Integrated Model to Assess the Global Environment - Global Nutrient Model (IMAGE-GNM) is another example. IMAGE-GNM has been used to quantify the nutrient pollution in the Yangtze basin from 1900 to 2010 (Liu et al. 2018). Wang et al. (2018b) calculated nutrient losses from food production to water systems using the NUFER model at the country level for China. Strokal et al. (2017) with the MARINA 1.0 analysed optimistic futures to reduce nutrient pollution from six large Chinese rivers in the future (2050). However, these existing modelling studies did not look at the maximum nutrient pollution level for sub-basins. Our study adds new insight on allowable pollution levels for sub-basins considering socio-economic equality.

Our study is unique in the way of applying the Gini approach. We applied the Gini approach to account for equality in allocating the allowable pollution levels (from the river moth) for sub-basins (up-, middle-, and downstream). We did it for three largest rivers in China. Existing studies used mainly the Gini optimization approach for local analysis. Gini was used to allocate pollution permits on pollutants for provincial or central government (Wang et al. 2016; Yuan et al. 2017). This was done for different pollutants. For example, the Gini approach was applied to allocate rights for the carbon and SO₂ emissions from industries, discharge permits for chemical oxygen demand (COD) and Ammonia Nitrogen (NH₃-N) in wastewater among cities (Li et al. 2016; Shen et al. 2018; Sun et al. 2010; Wang et al. 2016; Zhou et al. 2015). These studies allocated the pollution permit at an administrative scale (e.g., cities). In this study, we explored the allocation methods of the allowable nutrient pollution at the bio-physical scale (sub-

basin). We considered both bio-physical processes of nutrient flow in rivers and socioeconomic development of sub-basins.

Uncertainties and limitations. In this study, we set the constraints for minimizing the sum of Gini coefficients for four indexes (GDP, population, basin area, and agricultural area). Our constraint that the Gini coefficients for indexes should be below 0.4 was used in similar studies (Xiao et al. 2009; Yuan et al. 2017). We performed a sensitivity analysis to explore how sensitive our results are to the value of the constraint. Results indicate that the optimization problem is infeasible (i.e. we cannot find optimal allowable nutrient pollution levels) when the constraint is set to below 0.2. When using a constraint of 0.3, the allowable nutrient pollution level for each sub-basin is slightly changed compared with the results when using the constraint of 0.4 (Table 4.1 and A1 in Appendix III). An exception is the Jinsha sub-basin in Yangtze River, for which the difference is 30%. For other sub-basins, the allowable nutrient pollution levels did not change largely (5% higher or lower compared to the results with the Gini constraint of below 0.3) (Tables A.1 and A.2 in Appendix III). Moreover, we found the sum of the Gini coefficient for four indexes to be lower for 0.4 constraint than for 0.3 (Table 4.1). This implies that allowable nutrient pollution levels for sub-basins for the 0.4 constraint are comparable with those for the 0.3 constraint.

Our study selected GDP, population, agricultural land area and sub-basin areas as four indexes to represent the socio-economic development of GDP, population, basin area, and agricultural area are often selected as the indexes to reflect the socio-economic development in similar studies (Wang et al. 2016; Yuan et al. 2017). Wang et al. (2016) selected population, GDP and river length to reflect the social, economic and environmental efficiency. In Yuan et al. (2017), GDP, population and environmental receiving capacity of pollutants are selected as indexes to reflect socio-economic and environmental aspects of the region.

We applied MARINA 1.0 to quantify the actual river exports of nutrient in 2050. Although this model has been widely applied and validated for rivers in China, we are aware of the limitations of the MARINA 1.0. The model does not explicitly account for the dynamics in release and accumulate process of nutrient in the environment (air, soil and water) (Strokal et al. 2016a). These processes may over or under estimate the river exports of nutrient. For example, animal manure is a main pollution sources of river

exports of nutrient in China. The animal manure contains organic forms of P that can first accumulate and convert to available inorganic P that can be used by crops. This process is depending on various factors, such as the level of metal cation in the soil (Schachtman et al. 1998). We assumed that including this legacy of P will not significantly influence the calculated river export of P.

We used the ICEP approach to quantify the allowable nutrient pollution level for Yangtze, Yellow and Pearl rivers. ICEP is based on the Redfield ratio of N:P:C:Si, which is generic for the marine ecosystem. This ratio is affected by many factors, such as water temperature. So the ratio might be different among the ecosystem in time and space. In addition, we realized ICEP reflect the risk for coastal eutrophication for certain year. Nevertheless, ICEP is a good basis for this study to quantify the allowable nutrient pollution level for rivers. As we described in Section 4.3.1, the calculated allowable nutrient pollution level for the Yellow River, is higher than for the other rivers. Yellow River is the second largest basin that is characterized by low precipitation and runoff. There are two reasons for higher allowable nutrient pollution level for Yellow. The first reason is associated with the hydrological condition of the basin. Water consumption along the river is high and often exceeds the water recharge. Thus, water discharge at the river moth is very low. As a results, less nutrient and Si reach the coastal water. Si is one of the limiting factor for harmful algae bloom. Relatively low Si export lead to a low allowable nutrient level for Yellow River. Therefore, large reductions for TDN and TDP export are needed to avoid coastal eutrophication.

Our study identified opportunities to reduce river export of TDN and TDP to the allowable nutrient pollution levels. We show which sub-basins require higher reductions to meet the allowable pollution levels and from which sources. This information, especially the nutrient pollution reduction by sources, can be used by policy makers to further establish region-specific strategies to reduce nutrient pollution. According to the Fiver-year Plan for the National Economic and Social Development, water pollutant discharge permits are allocated based on pollutant loads at the administrative scale (provinces or cities) for every five years (Wang et al. 2016). This policy only includes discharge permits on NH₃-N from industrial and domestic sources. However, this is not enough to reduce the coastal eutrophication. Our study provides allowable nutrient pollution levels for both TDN and TDP. Moreover, our study indicates

either point or diffuse sources should be the focus on reducing the nutrition pollution for each sub-basin. This study strengths the current policy on water pollution and provides new insight to the formulation of sub-basins specific nutrient reduction strategies.

Table 4.1 Gini coefficient for four indexes (including gross domestic products (GDP), population, basin area, and agricultural area) for the Yellow, Yangtze, and Pearl rivers for TDN and TDP in 2050 under Global Orchestration scenario, when the Gini coefficient is constrained below 0.3 or 0.4.

-		Gini coefficient value			
Basin		Sub-basin area	Agricultural	Population	GDP
			land area	Density	
Yellow River	GO(TDP)	0.559	0.528	0.107	0.177
	GO(TDN)	0.526	0.494	0.172	0.174
	Gini < 0.3	0.174	0.142	0.300	0.293
	Gini < 0.4	0.136	0.104	0.334	0.333
Yangtze River	GO(TDP)	0.326	0.226	0.322	0.442
	GO(TDN)	0.231	0.132	0.222	0.364
	Gini < 0.3	0.250	0.156	0.138	0.300
	Gini < 0.4	0.180	0.075	0.196	0.365
Pearl River	GO(TDP)	0.226	0.222	0.157	0.369
	GO(TDN)	0.222	0.228	0.136	0.395
	Gini < 0.3	0.198	0.254	0.023	0.300
	Gini < 0.4	0.134	0.198	0.065	0.357

4.5 Conclusion

This study focuses on equality in pollution control in the Yellow, Yangtze and Pearl. Without pollution control, river export of nutrients for the year 2050 is higher than desirable from an environmental point of view. We calculate that river export of TDN and TDP in 2050 needs to be reduced by 78-93% to avoid coastal eutrophication. We focused on the question how to allocate these large reductions among sub-basins in an equal way. Our results indicate that equality in pollution control at the sub-basin level, requires reductions ranging from 60 to 96% for TDN and from 62 to 97% for TDP. For TDP, middle- and down-stream sub-basins of the Yellow and Yangtze rivers, in general, need to reduce more. This is different for the Pearl river where required reductions from upstream can be as high as for middle- and down-stream sub-basins. Clearly, some sub-basins need to reduce pollution levels more than others when equality is considered.

Equality in river pollution control requires sub-basin-specific, and pollutant-specific reductions in polluting activities. Opportunities for cleaner production are different for TDN and TDP. The direct discharges of animal manure to rivers (as point source) and sewage systems are important causes of nutrient pollution in 2050. This holds especially for TDP. Therefore, in most sub-basins, reducing TDP from sewage and point manure may be enough to meet the allowable TDP inputs to seas. This is different for TDN where diffuse sources (e.g., over-use of synthetic fertilizers) are also important contributors. Thus, meeting the allowable levels of TDN may require reduction of TDN from both diffuse and point sources.

Our study is the first to link a Gini based optimization approach with the MARINA model. It can support decision making on pollution control. We show how sub-basin-specific policies to reduce nutrient pollution can be formulated while considering equality.

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Chapter 5

Water pollution from food production: lessons for optimistic and optimal solutions

Abstract

Food production is a source of various pollutants in aquatic systems. For example, nutrients are lost from fertilized fields, and pathogens from livestock production. Water pollution may impact society and nature. Large-scale water pollution assessments, however, often focus on single pollutants and not on multiple pollutants simultaneously. This study draws lessons from air pollution control for large-scale water quality assessments, where multi-pollutant approaches are more common. To this end, we present a framework for future water pollution assessments searching for optimistic and optimal solutions. We argue that future studies could shift their focus to better account for societal and economic targets. Participatory approaches can help to ensure the feasibility of future solutions to reduce water pollution from food production.

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5.1 Introduction

Food production is expected to intensify in the coming years (Robertson and Vitousek 2009; Springmann et al. 2018). This is a result of the growing population that need more food (Springmann et al. 2018). Intensified food production is, however, a source of multiple pollutants in aquatic systems (Ippolito et al. 2015; Strokal et al. 2016a; Vermeulen et al. 2017). Overuse of chemicals and poor management strategies in the crop production sector result in losses of pesticides (Ippolito et al. 2015), heavy metals, pathogens (Vermeulen et al. 2017), and nutrients (Gu et al. 2013; Ma et al. 2013a; Strokal et al. 2017; Wang et al. 2018a) in rivers from fertilized fields. Intensifies livestock production is often a source of nutrients (Gu et al. 2013; Ma et al. 2013a; Strokal et al. 2017; Wang et al. 2018a), pathogens (Vermeulen et al. 2017) and antibiotics in rivers (Robertson and Vitousek 2009). In many world regions, aquatic systems experience multi-pollutant problems (Diaz and Rosenberg 2008). China is one of the examples, where aquatic systems are largely contaminated by pollutants from food production (Gu et al. 2013; Li et al. 2017a; Ma et al. 2013a; Strokal et al. 2017; Wang et al. 2018a). Multi-pollutant problems are also reported for many rivers of North America and Europe. This holds especially for densely populated areas. In the future, food production may add more pollutants to aquatic systems, impacting society (e.g., diarrhoea from pathogen contamination) and nature (e.g., harmful algae blooms from excess nutrients). The existing studies differ in their search for solutions to reduce water pollution from food production. Here we focus on two types of analyses: searches for *optimistic* and for *optimal* solutions.

Optimistic solutions show us to what extent environmental problems can be solved in scenarios reflecting maximum technical, economic and societal potentials to solve environmental problems. In scenarios searching for optimistic solutions, the full implementation of management strategies is often assumed to reduce pollution from human activities, for example, food production (Gu et al. 2013; Ma et al. 2013a; Strokal et al. 2017; Wang et al. 2018a).

Optimal solutions account for trade-offs, and show us how environmental targets can be met in the most cost-effective, equitable, or acceptable ways. Optimization analyses typically aim to achieve certain targets while looking for the optimal combination of environmental measures (Kahil et al. 2018; Wagner et al. 2013). Optimization analyses

can be combined with participatory approach to include stakeholders' interest. This is particularly relevant for sustainability targets, such as the Sustainable Development Goals (SDGs).

Multi-pollutant, large-scale optimization analysis are more commonly applied in air quality control (Amann et al. 2004; Hordijk and Kroeze 1997; Wagner et al. 2013) than in water pollution control. Water quality studies often analyse single pollutants and not multiple pollutants simultaneously (Gu et al. 2013; Ippolito et al. 2015; Li et al. 2017a; Ma et al. 2013a; Strokal et al. 2017; Strokal et al. 2019; Vermeulen et al. 2017; Wang et al. 2018a). This holds especially for large-scale water quality assessments.

In this study, we, therefore, draw lessons from air pollution control for large-scale water quality assessments, where multi-pollutant approaches are more common. We present a framework for future water quality assessments searching for optimistic and optimal solutions. Finally, we provide concluding remarks.

5.2 Lessons from air pollution control for water quality assessments

In the following, we draw three main lessons from existing models. In our discussion, we refer to the representative models that have been successfully applied for air pollution control at a continental or global scale and take a multi-pollutant perspective. We use these models as illustrative examples for water quality assessments. We identify opportunities for further development of existing water quality models.

Lesson 1: Integrated models for air pollution control have been more successful tools for international decision making than water pollution models.

Several integrated models exist for air pollution control taking a multi-pollutant perspective. RAINS (Regional Air Pollution Information and Simulation) model and its extended version for greenhouse gasses, GAINS (Greenhouse Gas and Air Pollution Interactions and Synergies) are illustrative examples of how integrated models can successfully be used in international negotiations related to environmental problems. RAINS and GAINS can be used to quantify emissions and air pollution impacts, and to identify least-cost strategies for air pollution control (cost-optimization). RAINS supported the formulation of "the European Commission's 1995 Acidification Strategy"

(http://www.iiasa.ac.at/). RAINS and GAINS played an essential role in international negotiations on the Convention on Long-Range Transboundary Air Pollution (LRTAP, http://www.unece.org/fileadmin//DAM/env/lrtap/welcome.html). This convention was an international agreement to deal with air pollution in Europe signed in 1979. The convention was extended to eight protocols on emission reduction targets for multiple pollutants in the air. Today, more than 50 countries in the world are taking part in this convention. The role of the models is in providing scientific information to support negotiations. This information includes quantified emissions of air pollutants (e.g., sulfur dioxide, nitrogen oxides, ammonia, and volatile organic compound) and greenhouse gasses (e.g., carbon dioxide, methane, and nitrous oxide) from European countries, environmental impacts of those emissions, effects of reduction strategies and costs of emission control (Amann et al. 2004; Hordijk and Kroeze 1997; Wagner et al. 2013).

The success of the RAINS and GAINS models in international negotiations can be explained by three main reasons. First, these models integrated multiple pollutants and their multiple effects. For example, emissions of sulfur dioxide, nitrogen oxides and ammonia cause acidification of forests and water. Nitrogen oxides and ammonia are also important contributors to eutrophication problems. Second, the models considered regional differences in socio-economic development and ecosystem sensitivities. The models contributed to an increased awareness among different stakeholders of the need to develop regional solutions, while accounting for transboundary emissions. Third, the models are able to provide a scientific basis to support a dialogue between different stakeholders. Models support the identification of optimal solutions (e.g. cost-effective) for reducing air pollution (Amann et al. 2004; Wagner et al. 2013). Today, these models are applied to many world regions (for China and India) with a 5-year time step up to 2050.

Water pollution models for multiple pollutants have not been as widely used as air pollution models in international negotiations. An important reason is that multipollutant models are successful in water quality assessment for the present day, but rather limited for future assessments of water quality at the continental or global scale. Several continental and global water quality models exist for individual groups of water quality parameters (e.g. nutrients). Examples of such models are Global NEWS-2 (Nutrient Export from WaterSheds) for nutrients (Mayorga et al. 2010; Seitzinger et al.

2010), IMAGE-GNM (Global Nutrient Model) for nutrients (Beusen et al. 2015), GloWPa (Global Waterborne Pathogen) for pathogens (Vermeulen et al. 2017; Vermeulen et al. 2019), VIC-RBM (Variable Infiltration Capacity - River Basin Model) for water temperature (Van Vliet et al. 2012), Global TCS (Triclosan) for triclosan (Van Wijnen et al. 2018), global plastic model (Van Wijnen et al. 2019) and the global pesticide model (Ippolito et al. 2015). Some water quality models exist for national assessments of water quality. Examples of such models are SPARROW (SPAtially Referenced Regressions On Watershed attributes) for the United States (Schwarz et al. 2006) and MARINA (Model to Assess River Inputs of Nutrients to seAs) for China (Strokal et al. 2016a), with both models designed for nutrient pollution assessment. The WorldQual model accounts for more than one group of pollutants in continental water quality assessments, but not for the future (Strokal et al. 2019). WorldQual quantifies biochemical oxygen demand, faecal coliform bacteria, total dissolved solids and total phosphorus (P) in river reaches for Africa, Latin America and Asia. A few more models account for multiple pollutants in aquatic systems at the national or continental scale (EPA 2019). Detailed review of existing, large-scale water quality models is presented in Strokal et al. (2019).

Lesson 1 highlights the opportunity for existing global and continental water quality models to further develop toward multi-pollutant assessments. This is needed to explore scenarios in which we search for optimistic and optimal solutions that could simultaneously reduce water pollution of multiple pollutants (see Lesson 2 below).

Lesson 2: Models can support the search for optimistic and optimal solutions for multipollutant problems in water, by assessing maximum technical feasibility and costeffectiveness.

Models are often used as scenario tools to analyse future water quality. Models are able to project the future water quality by Business as Usual (BAU) scenarios. In scenarios searching for solutions, BAU scenario often used as a baseline scenario, accounting for climate change and socio-economic developments. Climate change scenarios exist, for example, the IPCC Specific Report on Emissions Scenarios (SRES) (Nakicenovic et al. 2000) or the Representative Concentration Pathways (RCPs) (Van Vuuren et al. 2011). Scenarios exploring changes in socio-economic development in the future are, for example, the Millennium Ecosystem Assessment (MA) scenarios (Alcamo et al. 2005a), or the Shared Socioeconomic Pathways (SSPs) (O'Neill et al. 2014). Storylines of the

climate and socio-economic scenarios are incorporated into water quality models (e.g., Global NEWS-2, GloWPA). These storylines often form the basis of the alternative scenarios that aim at searching for optimistic and optimal solutions to reduce water pollution.

We can use water quality models to assess the maximum technical feasibility and cost-effectiveness of solutions for pollution abatement (Strokal et al. 2017; Wang et al. 2018a). Some of the existing water quality models (e.g., *MARINA*) are used to assess the maximum technical feasibility of solutions for reducing eutrophication problems (Robertson and Vitousek 2009). Differences in socio-economic development and climate change among sub-basins are considered. For example, focusing on the maximum technical potential to avoid coastal eutrophication in 2050, Strokal et al. (2017) showed the possibility to avoid coastal eutrophication by implementing advanced technologies (e.g. recycling animal manure to replace synthetic fertilizer) aiming at reducing losses of nutrients to aquatic systems. Similar study has also been conducted for the pathogens (Vermeulen et al. 2017). Scenarios reflecting the maximum economic and societal potential to solve multi-pollutant problems are less studies for large-scale water quality assessments.

Use of models for cost-effectiveness analyses are, however, less common for multipollutant water quality assessments. This is more common for models for air pollution assessments as we highlighted before. RAINS and GAINS are able to explore solutions with the maximum technical potential, and identify the least-cost strategies to reduce emissions of multi-pollutants to the atmosphere (Amann et al. 2004; Wagner et al. 2013) (see Lesson 1 above). Taking the cost-optimization approaches from air pollution model as example, such as RAINS and GAINS (Brooke et al. 1988), water quality models can develop further as tools for cost-effectiveness analyses from a multi-pollutant perspective. Another similar example is the Hydro-Economic Optimization model (ECHO) (Kahil et al. 2018). ECHO gives insights on cost-effective allocation of water across different sectors for Africa in a spatially explicit way. A few studies allocate wastewater discharge permits to cities in the most fair way, while considering socio-economic development (Chen et al. 2012; Sun et al. 2010; Wang et al. 2016; Yuan et al. 2017). These insights from existing optimization approaches can form a good basis to develop a cost-optimization model for water quality assessments at the large scale.

Lesson 3: To account for societal feasibility in water pollution assessment participatory approaches may be needed.

Accounting for the societal feasibility of implementing environmental solutions is important. This is because it gives us a better understanding of whether society is prone to accept certain measures or not. This will improve our water quality assessments, where technical, economic and societal aspects are accounted for. Such assessments will facilitate the formulation of effective environmental policies to reduce water pollution in the future.

Accounting for societal aspects is challenging, but not impossible. Several ways to do this exist. One is to invite stakeholders to co-design solutions based on existing scenarios (e.g., based on SSPs). Then, effects of such solutions can be tested by models. Another way is to involve stakeholders in the whole cycle of developing scenarios. Participatory approaches can help (Alcamo 2001; Kok et al. 2018; Pedde et al. 2019). An example is the 'Story-And-Simulation' approach (SAS). This approach has been used to develop scenarios for environmental problems (Alcamo 2001). Experts (e.g., modellers) together with stakeholders translate qualitative narratives into quantitative scenarios for models. This process is iterative and consists of several steps in which stakeholders are involved (see Alcamo 2001; Kok et al. 2018; Pedde et al. 2019 as example). Participatory approaches are part of the Water Future and Solutions Initiative, lunched by the International Institute for Applied Systems Analysis (IIASA, http://www.iiasa.ac.at/). This initiative is a good example how to bridge science to society and policy at different scales using various modelling tools (Satoh et al. 2017). There is a need to link the relevant sustainable development goals (SDGs) to the participatory approaches. For example, SDG 2 Zero Hunger (food production) and SDG 6 Clean water and sanitation (water quality) can be used as a scientific basis to support co-design of solutions with stakeholders during the participatory workshops.

5.3 Framework for future water quality assessments

We present a framework for future water quality assessments searching for optimistic and optimal solutions (Figure 5.1 and 5.2). We design this framework based on the lessons that we draw for large-scale water quality assessments (Section 5.2). Our framework provides an illustrative example of how different modelling approaches can be combined, to explore optimistic and optimal solutions for water pollution from food production or other pollution sources (e.g. human waste) taking a multi-pollutant perspective. The frame work covers drivers (food production and water pollution controls), pressure (pollutant losses), state (pollutant loads and concentrations), and impact (water pollution impact on nature and society) (Figure 5.1). For the water pollution impact, various indicators can be integrated into the framework. For example. Indicators for Coastal Eutrophication Potential can be used to reflect the impact of nutrient enrichment in the coastal water (Garnier et al. 2010).

The framework allows for two types of analyses: exploring optimistic and optimal solutions for water pollution (Figure 5.1). It focuses on water pollutants from food production, such as nutrients, pesticides, and pathogens (Beusen et al. 2015; Ippolito et al. 2015; Vermeulen et al. 2017). Exploring optimistic futures can be done through scenario analyses: starting from storylines, and optimistic assumptions about emission control. We can analyze future trends in water pollution, the costs of emission control, and the impacts of pollution on nature and society. Exploring optimal solutions typically starts from environmental targets, and aims at analyzing optimal (e.g. cost-effective) solutions to reach these targets. Our framework thus follows Lessons 1 and 2 as formulated above.

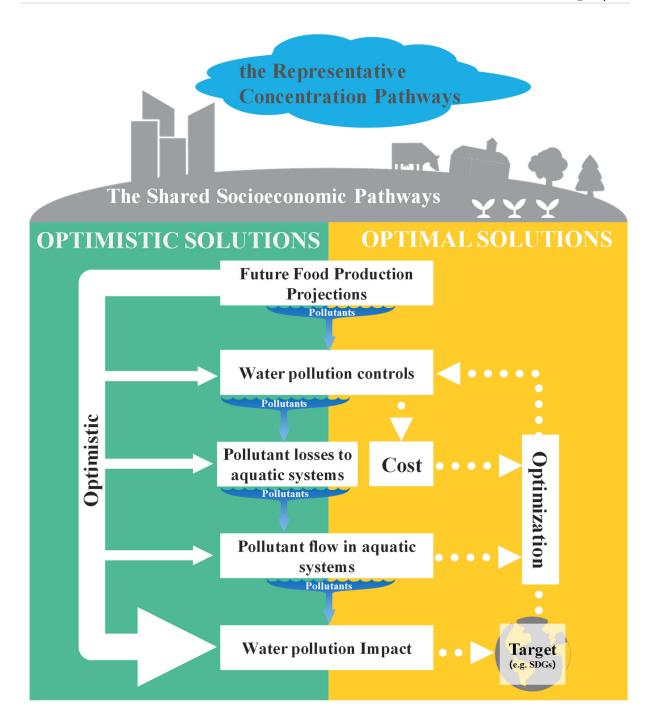
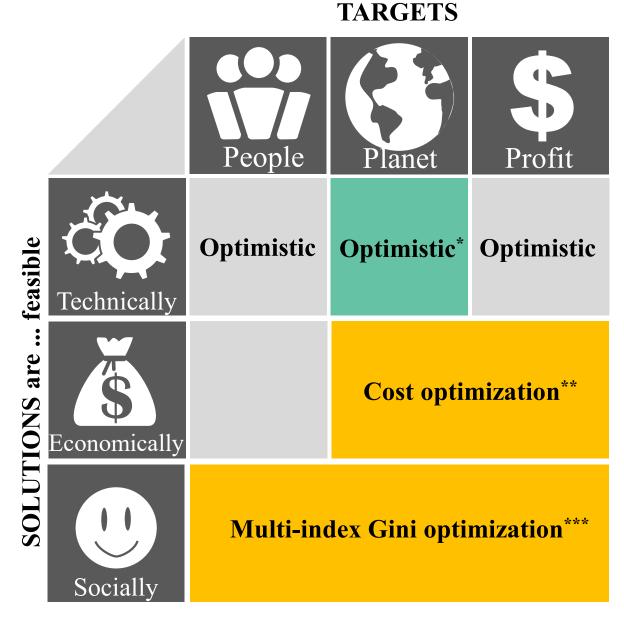


Figure 5.1 Framework for future water quality assessments searching for optimistic and optimal solutions. Examples of targets are shown in Figure 5.2.



^{*} Optimistic scenario analysis: (Gu et al. 2013; Ma et al. 2013a; Strokal et al. 2017; Wang et al. 2018a)

** Cost optimization: (Jiang and Hellegers 2016; Lescot et al. 2013; Perni and Martínez-Paz 2013; Udías et al. 2014)

Figure 5.2 Overview of how targets (for people, planet and profit) and solutions are linked. Colours in the cells follow the colours in Figure 5.1. So far, scenario analyses searching for optimistic solutions for water quality focused mostly on meeting *environmental* targets by *technical* solutions (the green box in the graph). Yellow boxes refers to optimization analyses that can be applied to large-scale water quality issues, of which some examples can be found in the literature. Grey cells indicate types of analyses that are not yet widely performed.

^{***} Multi-index Gini optimization: (Chen et al. 2012; Sun et al. 2010; Wang et al. 2016; Yuan et al. 2017)

Existing models could form the basis of the framework. To address the impact of food production on water quality, the framework should be able to quantify the pollutant losses to waters from the food production chain. It should also include control measures to reduce the pollutant losses from the food production chain. It also needs to account for the transport of pollutants through the environment, and retention processes. Pollutants may be transported by rivers from upstream to downstream and eventually entering the seas. During the transportation, pollutants can be lost or retained in the river systems. Examples are nitrogen losses due to denitrification, P retentions in sediments and retentions of various pollutants due to river damming. Finally, the framework should account for effects of pollutants in the environment, on nature and society. Several models exist to quantify pollutant flows from food production to the aquatic systems at large scales (Strokal et al. 2019; Tang et al. 2019; Van Vliet et al. 2019). These models can be used to identify 'hotspots' of water pollution, and to analyze past and future trends in water pollution (Strokal et al. 2019; Wang et al. 2018b). They could form the basis of the framework.

Exploring *optimistic* solutions could start from optimistic storylines about future trends in society, and about what is technically feasible in terms of pollution control (Figure 5.1). Models can then be used for scenario analysis, analysing future trends in water pollution while assuming full implementation of existing and future technologies to reduce water pollution. One could compare the results with, for instance, targets for pollution control deduced from people, planet or profit boundaries (Figure 5.2). For China, some examples exist of modelling studies exploring optimistic scenarios for reducing nutrient pollution by technically feasible options at basin or national scale (Li et al. 2017b; Strokal et al. 2017; Wang et al. 2018a). These examples indicate that it is technically possible to reduce pollution to low levels in the future. So far, scenario analyses searching for optimistic solutions focused mostly on meeting environmental targets by technical solutions (green box in Figure 5.2). In addition to optimism about technologies, one could also add optimistic assumptions about human behavior. For instance, storylines may assume sustainable development in society, reflected, for instance, by environment-friendly behavior. In such futures, farmers and consumers will be concerned about the environment and thus do not overuse agrochemicals in crop productions, and move to vegetarian diets. Optimistic futures may, furthermore, assume that industry and waste water treatment may aim for green development.

Exploring *optimal* solutions for water pollution, could start from environmental targets, to be reached in an optimal way (Figure 5.1). Optimal can be interpreted here as economic, technical or social optimum (Figure 5.2 and Section 5.1). In Figure 5.1, we give an example of searching for cost-effective solutions. Cost-optimization has been successfully applied in controlling the air pollution in European countries (see Section 5.2). To account for people, planet and profit simultaneously in optimization analyses, the Gini coefficient could be used (Figure 5.2). The Gini coefficient reflects equality of income or wealth within society according to the Lorenz curve (Lambert 1985; Rahman et al. 2009). The Gini coefficient can also be used to reflect the equality in use of environmental resources, such as allocating the waste discharge permit (Chen et al. 2012; Sun et al. 2010; Wang et al. 2016; Yuan et al. 2017). Absolute equality in a country is reached when all people have an equal share in resources, or in economy. The Gini coefficient can be used in optimization analysis to search for strategies to meet targets (for people, planet or profit) in such a way that social equality is maximized. The Gini coefficient for pollutant discharge can be quantified for various indexes, such as population density or gross domestic product. Multi-index optimization involves optimization of the equality in the discharge of water pollution for multiple indexes. One could apply this approach in water pollution assessment, for instance to allocate pollution rights (Yuan et al. 2017).

Optimistic scenarios and optimization approaches can assist decision makers in their search for solution to water pollution (see Lessons 1 and 2). To implement the framework proposed in Figure 5.1, some hurdles have to be taken if we want to apply it for multi-pollutant problems. First, existing large scale water quality models run at different spatial and temporal scales. The abovementioned global and regional water quality models (Sections 5.1 and 5.2) calculate pollutant flows at scales of 0.5° grid (e.g. IMAGE-GNM, GloWPa, VIC-RBM), basin scale (e.g., Global NEWS-2, Triclosan model), or sub-basin scale (e.g. MARINA, WorldQual) (Beusen et al. 2015; Ippolito et al. 2015; Strokal et al. 2019; Vermeulen et al. 2017). Some of them are process-based (e.g., IMAGE-GNM, VIC-RBM) while others take a lumped, parameter-based modelling approach (e.g., Global NEWS-2, MARINA). Most of them are steady-state models that quantify the annual pollutant flows (details on model reviews are in Strokal et al. 2019). A few models quantify seasonal nutrient flows from land to seas globally (McCrackin et al. 2014) or nationally (Schwarz et al. 2006). However, pollution control typically takes

place at international, national, or local scales (administrate scale), and in shorter timeframes. It is a challenge to integrate biophysical and administrative scales in water assessments.

A second challenge is how to account for societal feasibility. Lesson 3 above calls for participatory approaches. Stakeholders could be involved in formulating storylines, targets and in identifying optimistic and optimal solutions, while using the modelling framework presented in Figure 5.1. This will help to ensure that optimistic futures are realistic, and that optimal solutions account for trade-offs.

Our presented framework can 1) advance the field of water quality modelling; 2) help to integrate people, planet, and profit-related targets with technical, economic and social solutions; 3) help to link water and food security assessments. The framework can help to achieve the Sustainable Development Goals (SDGs) for Clean Water and Sanitation (SDG 6) and Zero Hunger (SDG 2) at the same time. For example, targets for food production (related to SDG 2) and water quality (SDG 6) can be used as multiple constrains in optimization analyses. This may help to identify possible synergies and trade-offs.

5.4 Concluding remarks

In this study, we argue that large-scale water quality assessments can learn from air pollution control to identify optimistic and optimal solutions. Both optimistic (e.g. technically feasible) and optimal (e.g. cost-effective) solutions are needed for effective reduction of future water pollution from food production. We draw three main lessons from air pollution control for water quality assessments, searching for optimistic and optimal solutions. These lessons are: 1) Integrated models for air pollution control have been more successful tools for international decision making than water pollution models; 2) Models can support the search for optimistic and optimal solutions for multiple pollutant problems in water, by assessing maximum technical feasibility and cost-effectiveness; 3) To account for societal feasibility in water pollution assessment participatory approaches may be needed. Next, we present a framework for exploring optimistic and optimal solutions for water quality problems. The framework combines optimistic scenarios and optimization approaches with water quality models to explore the optimistic and optimal solutions for water pollution. We show that current water quality studies focus on environmental targets and technical solutions. We argue that future studies could shift their focus to better account for societal and economic targets. Participatory approaches may be needed to ensure feasibility of future solutions to reduce water pollution from food production.

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Chapter 6

Discussion and conclusion

6.1 Introduction

This chapter provides a general discussion and conclusion of the thesis. I summarize the main findings and novelties of the thesis in Section 6.2. I draw lessons for modelling water pollution and exploring the future in Section 6.3. Finally, I discuss the future outlook in Section 6.4.

6.2 Findings and novelties

The objective of this PhD thesis was to explore control strategies for future water pollution in China, caused by nutrients and pesticides. To address this, I formulated four sub-objectives:

Sub-objective 1: to analyse future potential pesticide losses to Chinese waters under global change.

Sub-objective 2: to explore the possibilities to avoid coastal eutrophication in the North China Plain.

Sub-objective 3: to quantify desired nutrient pollution levels for sub-basins of the Yellow, Yangtze and Pearl Rivers from both an environmental and equality point of view.

Sub-objective 4: to draw lessons from air pollution control for large-scale water quality assessments, where multi-pollutant approaches are more common.

These four sub-objectives are achieved in Chapter 2 to Chapter 5 of this PhD thesis. I discussed main findings and novelties in Sections 6.2.1 and 6.2.4 by chapter. The general overview is illustrated in Figure 6.1.

6.2.1 Water pollution caused by pesticides under global change (Chapter 2)

The main findings are as follows:

- Pesticide runoff to surface water increased by 45% during 2000–2010, nationally
- In the future, 58% to 84% of the Chinese population will live in hotspots of pesticide pollution.

Chapter 2 provides novel insights into the trends and hotspots of potential pesticide runoff from agriculture to surface waters from the past to the future for China. I explore the future trends of the pesticide runoff for the SSPs and RCPs. From 2000 to 2010, the pesticide runoff from agriculture to surface water increased by 45%. The top five provinces (Hebei, Fujian, Sichuan, Hubei, and Henan province) contribute over half the total pesticide runoff. In 2010, over 70% of the population lived in hotspots of pesticide pollution. In the future, the pesticide runoff may largely increase in large parts of China, and the hotspot area could expand to different areas under global change. In the Economy first scenario (SSP5-RCP8.5), which assumes a rapidly developing world with intensive use of resources and with severe climate change, the pesticide runoff is projected to increase considerably from 2010 to 2099. Moreover, the pesticide runoff in the top five province increased by 85%. The population in the hotspots is projected to increase to 84% in the Economy-first scenario, In the Sustainability scenario (SSP1-RCP2.6), which assumes sustainable socio-economic development with low climate change, the population in the hotspots could reach 76% in 2050 and 65% in 2099. In the "Middle of the Road" scenario, the hotspots are calculated to accommodate 77% of the total population in 2050 and 58% in 2099.

6.2.2 Controlling water pollution caused by nutrients (Chapters 3 and 4)

The main findings of Chapter 3 are as follows:

- To avoid coastal eutrophication without crop yield losses in the future, reducing 50–90% of N and P inputs to the seas is required.

- To achieve the required reduction, replacing synthetic fertilizers with animal manure is the most effective option.

Chapter 3 is novel in that it explores possible pollution control strategies for the NCP including reducing river export of dissolved N and P to the desired target, so that the potential for coastal eutrophication is low. This chapter is also novel in providing an illustrative example of applying the back-casting approach with the MARINA to identify effective water pollution control strategies to avoid future coastal eutrophication in China. In 2050, the Yellow, Huai and Hai Rivers (in the NCP) are projected to export large amounts of nutrients, which could pose a high risk for coastal eutrophication. To avoid the coastal eutrophication meanwhile maintaining the crop yield in the NCP, 50-90% of river export of N and P need to be reduced. I developed 54 scenarios by combining three different pollution control strategies to different extends: improving N and P removal rates in sewage systems, improving animal feed to reduce N and P in animal manure, and replacing synthetic fertilizer with directly discharged animal manure. The results indicated that coastal eutrophication could be avoided in the NCP, and replacing synthetic fertilizer with directly discharged animal manure is the most effective option.

The main findings of Chapter 4 are as follows:

- It is possible to avoid coastal eutrophication while considering equality in socioeconomic development in the Yellow, Yangtze and Pearl Rivers.
- To reduce N to allowable levels, reducing both point and diffuse source pollution may be needed
- Allowable levels for P could be reached by reducing point source pollution from manure and sewage

The main novelties of Chapter 4 are exploring the opportunities for controlling nutrient pollution in the Yellow, Yangtze and Pearl Rivers while considering socio-economic equality in 2050. I combined the MARINA with a Gini optimization approach to account for equality in socio-economic developments. For 2050, we defined allowable levels for the river export of TDN and TDP that imply reductions by about 90% for the Yellow River, about 80% for the Yangtze River, and about 85% for the Pearl River. The required reductions in TDN and TDP were quantified for each sub-basin, considering equality in socio-economic developments. The required reductions range from 60% to 96% for

TDN and 62% to 97% for TDP among the sub-basins. For TDN, this implies a need to reduce pollution from both point and diffuse sources. For TDP, the targets can be met by reducing point source pollution, particularly from sewage effluents and direct discharges of animal manure to surface waters.

6.2.3 Water pollution caused by multiple pollutants (Chapter 5)

The main findings are as follows:

- Integrated models are useful to explore solutions for water pollution accounting for environmental, societal and economic targets
- A framework is developed that combines optimistic scenarios and optimization approaches with large-scale multi-pollutant assessment of water quality

This chapter is novel in proposing a framework for combining optimistic scenarios and optimization approaches with integrated large-scale multi-pollutant models for water quality assessment. Integrated models have been applied to many regions (e.g. Europe, China, and India) to control air pollution, and these can serve as examples for controlling water pollution using integrated water quality models. The proposed framework enables exploration of optimistic and optimal solutions that can simultaneously reduce multiple pollutants in waters. By integrating it with participatory approaches, the framework can assist negotiation among stakeholders and provide scientific evidence to explore feasible solutions for water pollution in large regions. Additionally, there is a potential to link targets for people, planet and profit with the framework to explore technical, economic and social solutions for water pollution.

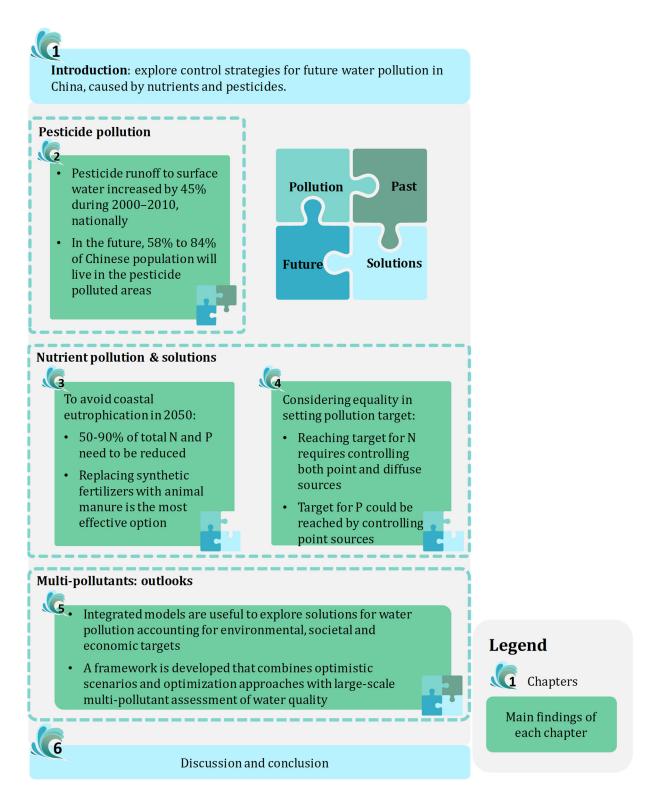


Figure 6.1 An overview of the main findings of this PhD thesis by chapters.

6.3 Lessons for modelling water pollution and exploring future solutions

In this section, I briefly introduce the models that are used in this thesis and their strength and weakness in Section 6.3.1. In Section 6.3.2, I compare other models that are widely used to quantify water pollution with models that I used in this thesis. I summarize lessons of this PhD thesis in Section 6.3.3.

6.3.1 Models used in this thesis

Pesticide model

In this thesis, I updated the existing global insecticide model developed by Ippolito et al. (2015) to assess pesticide pollution in China. The pesticide model developed in this study calculates the potential pesticide runoff from near-stream agriculture to surface waters in China at a 30 by 30 arcminutes grid for two past years (2000 and 2010) and two future years (2050 and 2099). The potential pesticide runoff reflects the possible maximum pesticide losses to surface water through runoff after the maximum daily rainfall event within the year (Table 6.1). The model considers nine factors that can largely affect potential pesticide runoff to surface waters: 1) the area of the near-stream agricultural land, 2) pesticide application rate, 3) soil carbon content, 4) soil texture, 5) average slope, 6) proportion of agricultural land, 7) maximum daily precipitation, 8) soil organic carbon-water partitioning coefficient of the pesticide, and 9) plant interception.

The pesticide model developed in this study is an improvement in three main ways, which I consider strengths of the model: 1) pesticides, 2) model inputs and 3) my future analyses. First, the pesticide model of this study can quantify the potential for pesticide runoff, while the original insecticide model only calculates the potential for insecticide runoff. Second, the pesticide model developed in this study uses local information and updated global datasets as a basis for the model inputs. For example, the area of the near-stream agricultural land for the pesticide model is calculated based on a more detailed dataset with river network information. The model by Ippolito et al. (2015) assumes that the near-stream agricultural land (stream corridor) for each grid is constant. The most important improvement in the pesticide model is improve estimation of the pesticide application rate. Based on the uncertainty analyses of

Ippolito et al. (2015), the pesticide application rate results in the largest uncertainty of the model. The pesticide model developed in this study uses the province-based pesticide application in the National Statistical Yearbook and grid-based land-use data to estimate the pesticide application rate. The global insecticide model estimate the insecticide application rate using the country based the Food and Agriculture Organization (FAO) dataset. Third, the pesticide model developed in this study can estimate the pesticide runoff for the past (2000 and 2010) and future (2050 and 2099). This is another novelty of this pesticide model. To forecast the potential for pesticide runoff under global change, I developed three scenarios based on the combined SSPs and RCPs that are the Sustainability, "Middle of the Road", and Economy-first scenario. To our knowledge, this is the first attempt to explore the pesticide pollution in waters under global change for the whole of China.

The pesticide model developed in this study has limitations. It is challenging to validate this type of model output because the model does not calculate the exact pesticide concentration. The potential pesticide runoff can be integrated as the maximum pesticide losses to surface water under the worst case scenario. No such measurements exists in China. Consequently, it is currently impossible to validate the model for the whole of China. Therefore, we need to build trust in the model in via an alternative approach. I argue that the model is fit for purpose, because the modelling approach has been applied and validated to assess pesticide runoff (Schriever and Liess 2007; Schriever et al. 2007) or insecticide runoff for Europe (Kattwinkel et al. 2011) and worldwide (Ippolito et al. 2015). Additionally, the model inputs I used are from reliable and publicly available Chinese statistical or widely-used global datasets (Fischer et al. 2008; Hempel et al. 2013b; Hurtt et al. 2020a; NBSC 2001). I thus combined a validated model with reliable input data for China in my study. This builds trust in the model. However, monitoring data for testing and further improving the model is required, especially for local analyses. This data should account for the effects of extreme rainfall on pesticide runoff.

The pesticide model developed in this study quantifies the potential for pesticide runoff but does not specify the pesticide type. This simplification can lead to under- or overestimation of the model outputs. In this thesis, I focus on pesticides as a broad group to provide initial insights into spatial variability in pesticide pollution in response

to heavy rainfall events. Thus, the results should be interpreted as a worse case and an indication of the spatial variability (where the problem is) and future trends (what can be expected). Despite the model's limitations, it enables estimation of the potential for pesticide runoff for data-poor regions. I consider this an elegant modelling approach as a first indication of pesticide pollution in the environment. Moreover, the model can be adjusted for specific pesticides when detailed data, such as the sown area and application rate of the specific types of pesticide, are available.

Another limitation is that the model misses some sources of pesticide in waters such as from urban areas. The model assumes that urban areas are not sources of pesticides. This is incorrect, especially for local analyses such as those for cities. However, it can be argued that urban areas contribute a small proportion of the total pesticide losses, which can be considered negligible. I realize that the above mentioned limitations and simplifications could lead to over- or underestimation of the output. I believe that the messages of my thesis are not largely influenced by these limitations. My analysis focuses on the whole of China. The model is specially explicit and quantifies the potential pesticide runoff at 30 by 30 arcminutes grid for China. For local analyses, such as those for cities, the model must be checked for missing sources and further validated.

MARINA

The first version of the MARINA was developed in 2016 for China at a sub-basin level for six major rivers (Strokal et al. 2016a). The model quantifies river export of nutrients to Chinese coasts based on human activities on land (e.g. agriculture, sewage), the main characteristics of sub-basins (e.g., soil retention, land-use type) and nutrient retention in the river and during export towards the river mouth (coastal waters). MARINA 1.0 quantifies nutrient pollution in Chinese waters for past years (1970 and 2000), and it also projects the future nutrient pollution in Chinese waters for the year 2050 under MEA scenarios (Table 6.1). When I started my PhD project, MARINA 1.0 was the latest version of the MARINA. When I finished my PhD project, more versions of the MARINA had been developed (see Chapter 1).

MARINA 1.0 quantifies nutrient export from land to seas in China by diffuse and point sources. Diffuse sources includes synthetic fertilizers, animal manure and human excretion on land, atmospheric N deposition (for dissolved inorganic N (DIN)), and

biological N fixation (For DIN), weathering of P-containing minerals (for dissolved inorganic P (DIP)), and leaching of organic maters (for dissolved organic N (DON) and P (DOP)). Point sources include direct discharges of animal manure, uncollected human waste from rural and urban populations, sewage effluent, and detergent use (only for DIP and DOP).

MARINA 1.0 has limitations and uncertainties. The model uncertainty is associated with the model inputs, scale and sources. The model inputs are from different sources. For example, inputs related to land-use, river network, and river discharges are from different global datasets (model description can be found in Chapter 3). The inconsistency of these data can create uncertainties in the model results. However, the model already incorporates available local information as much as possible, especially for the major sources of nutrient pollution. For example, many inputs related to animal manure excretion on land and animal manure directly discharged to rivers are derived from the Nutrient flows in Food chains, Environment and Resources use (NUFER) model, and this information is estimated based on Chinese statistics and surveys. Using local information increases trust in using the model for water pollution analyses.

The model calculates nutrient inputs to waters on a sub-basin scale. This is relatively coarse for analyzing local water pollution. Another limitation of MARINA 1.0 is associated with sources of nutrient losses to waters. MARINA 1.0 does not account for some nutrient sources, such as aquaculture and sewage effluent from industry. These missing sources were important for local nutrient pollution problems. However, this thesis strongly focus on nutrient pollution in large rivers, such as the Yellow, Yangtze and Pearl Rivers. Therefore, I believe the main conclusion will not change if these missing sources are included. The sub-basin scale is useful for studying large rivers, such as the Yellow, Yangtze and Pearl Rivers. These rivers are important study areas for this thesis. Additionally, this is a rather simple, elegant and transparent approach. This approach increases the opportunities for model application in data-poor regions and enables scenario analyses on nutrient pollution for large rivers in China. Hence, this approach can be integrated with other modelling approaches, such as back-casting and optimization. This is the focus of the Chapter 3 and 4.

MARINA 1.0 is the first comprehensive model that quantifies river export of nutrients to Chinese seas. The model use a simple, elegant and transparent approach that has many

potential functions. For example, it can be integrated with other modelling approaches. Hence, the results of MARINA 1.0 are relatively easy to understand and this increases the opportunities for model application. Moreover, Strokal et al. (2016a) used three different approaches to build trust for MARINA 1.0. First, the nutrient fluxes of MARINA 1.0 were compared with published measurements and the results of other models (e.g. Global NEWS-2). These comparisons indicate that the performance of the MARINA is good (Strokal et al. 2016a). Second, MARINA 1.0 models the trends of nutrient fluxes and this was compared with existing empirical studies. These empirical studies confirm the findings of MARINA 1.0 that nutrient inputs to Chinese waters have largely increased since the 1970s. Third, the model inputs are compared with independent datasets. The outputs of MARINA 1.0 are sensitive to changes in animal manure production, the direct discharge of animal manure, use of synthetic fertilizer and water discharge. To increase the confidence of the model outputs, inputs were compared with independent datasets at the county level for animal manure, synthetic fertilizers, and sub-basin areas. These comparison indicate the inputs of MARINA 1.0 is reliable (Strokal et al. 2016a). Therefore, I consider MARINA 1.0 a suitable model for this study based on the above mentioned strengths and its proven good performance.

Table 6.1 Overview of the models that are used in this thesis for pesticide and nutrient pollution in Chinese waters.

Models		Pesticide model in this thesis	MARINA 1.0 for nutrients (Strokal et al. 2016a)	
Purpose		Calculate the potential for pesticide runoff in response to heavy rainfall events	Analyze human affected nutrient losses to seas from sub-basins	
Model type		Screening model ^a	Processes-based, lumped empirical model	
System boundary		From agricultural land to surface water	From land to surface water, rivers and seas	
Pollutant sources		Diffuse sources: agricultural runoff	Point sources: collected and uncollected human waste b, the direct discharge of animal manure. Diffuse sources: agriculture, N deposition & fixation, P weathering, leaching of organic matter	
Considered pathways to surface water		Surface runoff that is generated by rainfall events	Direct discharge of treated and untreated pollutant, Leaching and runoff,	
Future projections		Shared Socio-economic Pathways and Representative Concentration Pathways	Millennium Ecosystem Assessment scenarios	
Model inputs		Environmental factors, human controlled agricultural practices	Human activities, economy & demography, sub-basin characteristics	
Outputs details	Outputs	Upper bound estimate of pesticide runoff to surface waters	River export of nutrients, source attribution Indicator for coastal eutrophication (ICEP) Sub-basin contribution	
	Spatial level of detail	30 × 30 arc-minutes	Sub-basins	
	Spatial extent	China	China	
	Temporal level of detail	Annual	Annual	
•	Temporal extent	2000, 2010, 2050, 2099	1970, 2000, 2050	

^a Screening model use the simple mathematic equations to estimate the extreme upper bound concentrations. It estimate the concentration under the "worst-case" scenario.

^b human excretion that is not collected by sewage system and can potentially be discharged directly to rivers without treatment.

6.3.2 Comparison with other models

Many models exist to assess the water quality for large regions. Most water quality models quantify single groups of pollutants. They differ in focused pollutants, modelling approach, and in spatial level of detail. To illustrate a general overview of the large-scale water quality models, I selected models that have been widely used in recent decades (Table 6.2). These are Global NEWS-2 (Mayorga et al. 2010), IMAGE-GNM (Integrated Model to Assess the Global Environment-Global Nutrient Model) (Beusen et al. 2015), WorldQual (Fink et al. 2018), SAPRROW (SPAtially Referenced Regressions On Watersheds) (Schwarz et al. 2006), NUFER (Ma et al. 2010), RTM (River Transport Model) (Liu et al. 2019), GloWPa (Global Waterborne Pathogen Model) (Vermeulen et al. 2017), GlobalTCS (Global Triclosan model) (Van Wijnen et al. 2018), Global insecticides model (Ippolito et al. 2015), SWAT (Soil and Water Assessment tool) (Arnold et al. 2012), and BASINS (Better Assessment Science Integrating Point and Non-point Sources) (EPA 2019).

The Global NEWS-2 model calculates the river export of nutrients to seas by point and diffuse sources for over 6000 rivers (Mayorga et al. 2010). The influence of human activities, such as sewage and agriculture, on river export of nutrients to seas are considered. Major characteristics of the basins, such as hydrology and land-use, are also included. The model accounts for nutrient retention in soil and rivers and nutrient removal during the transportation using the steady-state approach. The model outputs include dissolved inorganic forms, dissolved organic forms and particulate forms of N, P, Si and C exported by rivers to seas. The source attributions of these river exports of pollutants are also calculated. Point sources include sewage and detergents. Diffuse sources are synthetic fertilizer, animal exertion on land, atmospheric deposition (for N), biological fixation (for N), weathering of minerals (for P) and leaching of organic matter. The model uses the ICEP to represent the environmental impact of nutrient pollution in coastal seas. The Global NEWS-2 model provides outputs for two past years (1970 and 2000) and two future years (2030 and 2050). To project future trends, the model use the MEA scenarios. This model has been widely used for the world, Europe, Africa, and China. MARINA 1.0 is based on Global NEWS-2 (Strokal et al. 2016a). The improvements of MARINA 1.0 include 1) down-scaling to sub-basin scale, 2) adding missing sources (animal manure and human waste), 3) updating reservoir information and 4) updating

model inputs with local information. The most important improvement is adding the direct discharge of animal manure as point sources, which accounted for over 50% of TDP and 20% of TDN in 2012 (Wang 2020).

IMAGE-GNM calculates river export of nutrients for the world at the grid scale (30 by 30 arcminutes) (Beusen et al. 2015). The model considers the nutrient retention in soil and rivers using a dynamic and spiraling approach. Besides sewage and agriculture, this model considers pollutant sources from aquaculture, industrial wastewaters, allochthonous biomass inputs to rivers, and direct N deposition on waters. The model outputs are the total N and P to seas from 1990 to 2010 at annual level. This model has been applied to the world (Vilmin et al. 2018), Yangtze basin (Liu et al. 2018), and China (Wang et al. 2019). The IMAGE-GNM can be used to analyze local nutrient pollution problems; however, the data-demand is very high. It requires gridded inputs, which is challenging for data-poor regions such as China. Hence, the outputs of the IMAGE-GNM are difficult to check since the model is not validated or calibrated for China.

Some other models, such as RTM (Liu et al. 2019) and SPARROW (Schwarz et al. 2006), also calculate the river export of nutrients; however, they can only calculate some forms of the nutrients. RTM calculates the DIN in rivers and DIN flows to seas considering the agriculture, water regulation and human waste for the world at grid scale (30 by 30 arcminutes) (Liu et al. 2019). The model takes the dynamic approach to account for the N transportation from land to seas. The current version can calculate the annual DIN fluxes from 1991 to 2010. SPARROW model can estimate the N transported from land to seas by combining empirical and process-based mass-balance approaches (Schwarz et al. 2006). The model calculate the annual nutrient fluxes for basins in the US from 1999-2014. It only considers the diffuse sources. The modeling approach of RTM and SPARROW could potentially be applied to China; however, this would be challenging. The model requires a large number of inputs on an very detailed spatial scale and this is almost impossible for China. Hence, these models do not specify source attribution of nutrient fluxes as detailed as MARINA 1.0. Without source attribution, it is difficult to identify opportunities to reduce nutrient pollution and this is one of the objective in Chapter 4 of this thesis.

Table 6.2 Summarized overview of the selected large-scale water quality models

Models		Global NEWS-2 Nutrient Export from WaterSheds	IMAGE-GNM Integrated Model to Assess the Global Environment-Global Nutrient Model	SPARROW SPAtially Referenced Regressions On Watersheds	NUFER Nutrient flows in Food chains, Environment and Resources use	RTM River Transport Model
Purpose		Estimate nutrient export from basins to seas under human activities	Quantify the nutrient	Estimate the pollutants transported from watersheds to large water bodies	Quantify nutrient flow in the food chain	Estimate nitrogen flows to seas under human influence
Polluta	ınt group	Nutrients	Nutrients	Nutrients	Nutrients	Nutrients
Model type		Lumped, quasi- empirical model	Distributed, physically based model	Distributed, physically based model	Lumped model with combined approach	Lumped model with combined approach
Modelling approach		Steady-state	Dynamic	Steady-state	Steady-state	Dynamic
Pollutant sources		Point sources and diffuse sources	Point sources and diffuse sources	Diffuse sources	Point sources and diffuse sources	Point sources and diffuse sources
Outputs details	Spatial level	basins	30 × 30 arcmin	1 km grid	County	30 × 30 arcmin
	Spatial extend	Global	Global	National (U.S.)	China	Global
	Temporal level	Annual	Annual	Annual	Annual	Annual
	Temporal extend	1970, 2000, 2050	1990-2010	Long term average from 1999-2014	1989-2010, 2030	1991-2010
References		(Mayorga et al. 2010)	(Beusen et al. 2015)	(Schwarz et al. 2006)	(Ma et al. 2010)	(Liu et al. 2019)

Table 6.2 (Continue) Summarized overview of the selected large-scale water quality models

Models		GloWpa Global Waterborne Pathogen Model	GlobalTCS Global Triclosan model	Global insecticide model	WorldQual	SWAT Soil and Water Assessment tool
Purpose		Estimate pathogen concentrations in surface water	Estimate triclosan export from land to seas	Estimate the potential for insecticide runoff	Assess water quality level under human influence	Simulate water quality and quantity for small watershed
Pollutant group		Pathogens	Triclosan	Insecticide	Nutrients, Pathogens	Nutrients & Pesticides
Model type		Lumped, quasi- empirical model	Lumped, quasi- empirical model	Screening model	Distributed, physically based model	Distributed, physically-based model
Modellin	g approach	Steady-state	Steady-state	Steady-state	Dynamic	Dynamic
Pollutant sources		Point sources	Point sources	Diffuse sources	Point sources and diffuse sources	Point sources and diffuse sources
Outputs details	Spatial level	30 × 30 arcmin	30 × 30 arcmin	5 × 5 arc-minutes	5 × 5 arcmin	Flexible (depends on model inputs)
	Spatial extend	Global	Global	Continental	Global	Flexible (depends on model inputs)
	Temporal level	Annual	Annual	Monthly	Annual	Flexible (depends on model inputs)
	Temporal extend	2010, 2050	2000, 2050	1990-2010	Average 2000-2010	Flexible (depends on model inputs)
References		(Vermeulen et al. 2017)	(van Wijnen et al. 2018)	(Ippolito et al. 2015)	(Fink et al. 2018)	(Arnold et al. 2012)

Some models are not developed to assess the water quality, while others can also calculate pollutant emission to waters. For example, NUFER was developed to analyze the nutrient flows in the food chain for China (Ma et al. 2010; Wang et al. 2017). One of the main NUFER outputs is the nutrient use efficiency in different parts of the food chain. NUFER can also calculate annual nutrient emissions to air and waters at a county level. The model combines a process-based and mass-balance approach with local information as inputs. However, its approach to quantifying nutrient emissions into waters is rather simple. It does not calculate river export of nutrients to Chinese seas and this is a focus of this thesis.

Recently, interest has increased in models to quantify the concentration of new emerging pollutants in waters. The GloWPa model quantifies the concentrations of annual waterborne pathogens from sewage systems and livestock at a grid scale (30 by 30 arcminutes) for the world (Vermeulen et al. 2017). The GlobalTCS model calculates the river export of triclosan to seas at the grid scale (30 by 30 arcminutes) (Van Wijnen et al. 2018). The global insecticide model quantifies the potential for insecticide runoff at a grid scale (5 by 5 arcminutes) (Ippolito et al. 2015). These models account for major sources of the pollutants. The findings of these studies implies that new pollutants are polluting Chinese waters, and these merit more attention.

In fact, models that can quantify multiple pollutants simultaneously in waters exist. For example, WorldQual can assess the water quality for large regions at the grid scale (5 by 5 arcminutes) (Fink et al. 2018). This model uses simple equations and consistent available data to calculate the monthly biochemical oxygen demand and total dissolved solids considering the anthropogenic influence and flow dilution. The model has versions to calculate the total P in some large lakes of the world and the pathogen concentration in rivers for Africa and Europe for the past and present. The model considers point and diffuse sources, such as agriculture, urban surface runoff, uncollected waste, weathering, vegetation and soil, collected and uncollected human waste and manufacturing. However, the currently available version of the WorlQual model does not calculate the different pollutants in surface water, simultaneously. A similar example is the SWAT (Yang et al. 2008). The SWAT can calculate the pollutant

fluxes at hydrological response units, and then the outputs are further aggregated to basins. The spatial and temporal scales depend on the model inputs and the input demands are very high. The SWAT has been widely used to analyze the water pollution caused by nutrients (from diffuse sources) and pesticides for both large and small basins. The model can provide high-resolution results at monthly or seasonal scales, but only when it has been accurately applied. These models are currently only applied for past and present years. It is challenging to project the future using these models. Hence, none of the above mentioned models considers the interaction of different pollutants in waters. This can potentially lead to increases or decreases of pollutants in waters. Therefore, to better understand water pollution caused by multiple pollutants, there is a need to develop a model that quantifies losses of multiple pollutants to waters for large regions using a transparent approach. Strokal et al. (2019) propose a global multipollutant modelling approach that can identify hotspots for water pollution.

6.3.3 Exploring the future

In Chapters 2, 3 and 4, I study the future trends and explore potential solutions for water pollution. I apply forecasting, back-casting and optimization with water quality models (Figure 6.2). The forecasting and back-casting are scenario based approaches that describe likely, possible (forecasting) or desired (back-casting) futures (Robinson 2003). Examples of the scenarios with the forecasting approach are socio-economic development scenarios of the MEA (Alcamo et al. 2005a) and SSPs (O'Neill et al. 2014). An example of future climate change scenarios are the RCPs that were published by the Intergovernmental Panel on Climate Change (IPCC) (Van Vuuren et al. 2011). The optimization approach is often a mathematical approach to identify the "optimal" solutions to reach the targets with given constraints. The trade-offs within the studied system are automatically considered by the mathematical algorithm. Optimization models are used to assess optimization problems (Bashar et al. 2018). For example, costoptimization models search for the cheapest solutions to meet certain targets, such as reducing nutrient pollution levels. In the following paragraphs, I introduce the application of scenario-based forecasting and back-casting, and the mathematical model based optimization in this thesis.

Scenario-based forecasting

Scenario-based forecasting aims to generate a forecast under different assumptions for the future. Such forecasts can describe likely futures, the worst-case futures or futures under given conditions (Bunn and Salo 1993). Chapter 2 provides an example that explores the trends in the potential for pesticide runoff to Chinese waters under three global change scenarios. The three global change scenarios represent a sustainable future (Sustainability – representing SSP1-RCP2.6), a future with moderate progress towards sustainability ("Middle of the Road" – representing SSP2-RCP4.5), and a rapidly developing future with intensive resources use (Economy-first scenarios – representing SSP5-RCP8.5). Parts of Chapters 3 and 4 also use the forecasting approach with MARINA 1.0 to quantify river export of nutrients to seas under the Global Orchestration (GO) scenario.

Combining a forecasting approach and water quality models obtains further insights into the future trends of water pollution. The findings in Chapter 2 provide better quantitative understandings of pesticide pollution in the future. In Chapters 3 and 4, I quantify the river export of nutrients under the Global Orchestration scenario of the MEA and this is considered a likely future for nutrient pollution in Chinese waters. However, when I began Chapters 3 and 4, China had not yet launched many policies to improve the water quality and promote sustainable food production. This gap has been fulfill by other studies using a forecasting approach with water quality models. For example, Wang et al. (2018a) present the effectiveness of current policy, which restricts synthetic fertilizer use in China (Zero growth in fertilizer use after 2020), on reducing nutrient pollution in waters using scenario analysis with the NUFER model. Yang et al. (2019) assess the future impacts of two events, which are the 2022 Olympic Winter Games in Guanting basin and the development of Xiong'an in the Baiyangdian basin, on river export of nutrients. The above mentioned studies provide some snapshots of a plausible future for the nutrient pollution in parts or the whole of China using forecasting with water quality models.

Scenario-based back-casting

Back-casting aims to explore the possible pathways towards a desired future (Holmberg and Robert 2000; Robinson 1982). It begins by formulating a desired goal for the future. The desired future is preferably described as a quantitative target. Back-casting can be combined with scenario analysis. An increasing number of studies apply a set of

normative scenarios with water quality models to explore the possible scenarios that can reach the quantitative target. Chapter 3 of this thesis provides an illustrative example of the application of scenario-based back-casting with a water quality model. I used back-casting with MARINA 1.0 to explore pollution control strategies that could avoid coastal eutrophication at the mouth of the Yellow, Huai and Hai Rivers in the NCP. The desired future is defined as a future with low risks for coastal eutrophication at the mouth of Yellow, Huai and Hai Rivers. I used the ICEP to quantify this desired future, that is, an ICEP value below zero (with a uncertainty range from +1 to -1). Based on the ICEP, the quantitative target is calculated: the maximum level for the river export of dissolved N and P when the ICEP is below zero. I developed 54 alternative scenarios that comprise three pollution control strategies with different extents of implementation. In this study, I identified 7 to 25 scenarios with low risks for coastal eutrophication at the mouth of Yellow, Huai and Hai Rivers. This back-casting approach can be applied with other water quality models for other regions with different environmental targets.

Mathematical model based optimization

Mathematical model based optimization is used to obtain optimal solutions for reaching targets under given constraints. Other than in back-casting, the model itself is used to find the optimal solution. Optimization models can help to answer questions such as "What are the most cost-effective solutions to reach certain targets?", or "What is the most equitable plan for pollutant discharges permitted in large regions?". Optimization models are successfully applied in air pollution assessments searching for optimal policy solutions. One example is the Global Air Pollution Information and Simulation (GAINS) model (Wagner et al. 2013). The GAINS model has been used to search the cheapest or most cost effective mitigation strategies to achieve air pollution targets, particularly for Europe. Integrating such optimization approaches with water quality models can help to search for optimal solutions for water pollution problems under given constraints. Some applications of the mathematical model based optimization exist in the water pollution field. For example, Strokal et al. (2020) integrated cost-optimization with MARINA 1.0 to identify cost-effective options to reach the target for river export of N and P. Another example used the Gini optimization model to equally allocate the pollutant discharge permit to 13 cities in Jiangsu province (Yuan et al. 2017). Chapter 4 of this thesis provides another example of such an application. I combined Gini optimization with

MARINA 1.0 to explore the required reduction levels of nutrient fluxes in sub-basins of in the Yangtze, Yellow and Pearl Rivers with minimized inequality in socio-economic development. By this approach, the opportunities to reach the required reduction levels of nutrients were also identified. Results of Chapter 4 show the allowable levels for nutrient pollution, which ensure low risks for coastal eutrophication and meanwhile account for the socio-economic equality for the sub-basins of the Yellow, Yangtze and Pearl Rivers. The modeling framework of Chapter 4 can be adapted to the other versions of the MARINA or other water quality models to account for equality in exploring solutions for water pollution.

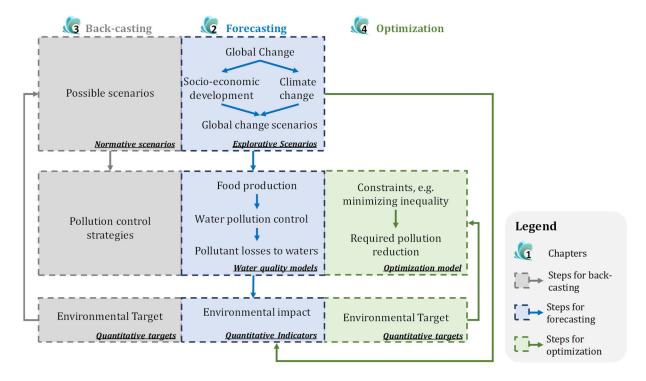


Figure 6.2 Three approaches to explore possible future or possible solutions for water pollution.

6.3.4 Lessons

Based on my study, I draw four main lessons for modeling the water pollution caused by nutrients and pesticides in China.

Lesson 1: The pesticide and nutrient models of this thesis are useful tools to identify hotspots and project future trends for water pollution in China and elsewhere.

I applied the existing MARINA model and developed the pesticide model to better understand the pollution distribution in China in the future. These two models provide spatially explicit outputs, which helps to identify the hotspots for nutrients and pesticide pollution (Chapters 2, 3 and 4). Additionally, these two models effectively balances the transparency and uncertainty. Therefore, I can use these two models to project water pollution under global change and further explore the potential options to control water pollution caused by nutrients (Chapters 3 and 4). These two models are transparent and the inputs demands are rather low. This gives the two models high potential for application in any other large region with limited information.

Lesson 2: Food production is often a common source of nutrients and pesticides in waters, calling for a need to develop multi-pollutant approaches in research and pollution control.

In this thesis, I used two separate models to analyze two groups of pollutants. According to the results, I found some common aspects when modelling these two groups of pollutants. First, the major sources of both nutrients and pesticides are mainly from the agricultural sector. Second, from the modeling aspects, these two models require the same inputs, such as land use. Third, I found that high levels of water pollution caused by pesticides and nutrients are in the same regions, for example, in the NCP. Fourth, policies to promote sustainable food production and green agriculture development focus on both pesticides and nutrients. Based on the above, I consider that a model must be developed using multi-pollutant approaches to simultaneously quantify pesticides and nutrient pollution in waters. Examples of such models using multi-pollutant approaches exist (Strokal et al. 2019).

Lesson 3: Combining back-casting and forecasting approaches can support the search for effective strategies to reach desired future targets for clean water.

Combining back-casting and forecasting approaches with the MARINA model helps to identify effective pollution control strategies to avoid coastal eutrophication. I provide an illustrative example to show the usefulness of combining the back-casting approach with the MARINA model. In this example, the desired future targets are producing sufficient food and avoiding coastal eutrophication. These two qualitative targets are translated into quantitative indicators. To produce sufficient food, the nutrient input to land should be larger than the nutrient uptake by crops and animal grazing. To avoid coastal eutrophication, the ICEP value should not exceed 0. Based on these, I quantified the maximum level for river export of N and P. I developed 54 scenarios by combining the different pollution control strategies and then used the forecasting approach by applying these scenarios in MARINA 1.0. By comparing the river export of N and P with these 54 scenarios with the maximum level, I identified the pollution control strategies that could reach the desired future. Translating a qualitative future into quantitative targets and pollution control strategies enables conversations between scientists and non-experts, such as policymakers. There is a potential to involve policymakers in the process to explore the pollution control strategies for water pollution problems. This methods can also applied with other targets (e.g. national standards or Sustainable Development Goals) for other pollutants.

Lesson 4: Combining optimization analysis with substance flow analysis in the MARINA model is useful support if we want to minimize socio-economic inequality in water pollution control.

Combining the MARINA with an optimization model can help to account for socioeconomic equality in water pollution control for large regions. In Chapter 4 of this thesis, I provide an example of combining the Gini optimization model with the MARINA; the former is a mathematical model and the latter is a substance flow model. The required reduction for river export of TDN and TDP is quantified using a back-casting approach with the MARINA and ICEP. Socio-economic equality is often omitted in pollution control for large regions. Here, I use the Gini optimization approach to explore a reduction plan for sub-basins of the Yangtze, Yellow, and Pearl Rivers with minimized inequality.

6.4 Outlook

6.4.1 Future recommendations for science

The water pollution models developed in this thesis can be further improved in several ways.

First, the potential for pesticide runoff can be quantified by crop type. The current version of the pesticide model classify the land-use into agricultural land and others. The agricultural land can be further classified into more details classes. Thus, the contribution of pesticide runoff can be identified by crop type. This can help to identify pollution control strategies for over use of pesticides. Second, the potential pesticide runoff can be quantified by pesticide type. Due to the limited available data, the pesticide model can only consider a pesticide as a whole. Pesticide pollution could be assessed by pesticide type when more explicit data on pesticide application rates became available. Hence, the model can be modified for specific types of pesticide and this depends on the research objectives.

For nutrient modeling, the following next steps are recommended. First, the performance of MARINA 1.0 can be further improved in several ways. Adding missing sources, such as aquaculture and direct N deposition, are preferred. To model nutrient pollution on a local scale, region-specific sources, such as industry or mining, need to be considered in MARINA 1.0. Recalibrating model parameters for China is required when water quality data become available. Second, nutrient models, such as MARINA 2.0 and 3.0, can be combined with the back-casting and Gini optimization approaches to explore optimal and optimistic options that reduce nutrient pollution to a sustainable level in the future. Involving stakeholders and policymakers in future studies on nutrient pollution could increase the feasibility of implementing pollution control options. This can be achieved by integrating the participatory approach, back-casting and optimization approaches with the MARINA. Third, models that assess nutrient pollution in Chinese lakes and groundwater are lacking. Finally, scenario analyses aiming to explore pollution control options considering social and economical feasibility are preferred in future studies.

I used two separate models to quantify pesticide and nutrient pollution in waters in this thesis. Because of the similarities in pollutant sources and distribution, a multi-pollutant model could also be used or developed to simultaneously quantify these two pollutants in waters with consistent spatial and temporal levels of detail. More sources of pesticide losses could be considered to achieve this. For example, washing equipment for

pesticide applicators and sewage systems in the urban region can also transfer pesticide to the sewage system. A modeling approach to account for losses of pollutants from sewage exists in the MARINA. This can serve as a basis for modeling pesticide pollution from sewage systems in urban region.

In this thesis, I use back-casting, Gini optimization with the ICEP and the MARINA to explore options for nutrient pollution control. These modeling approaches can also be combined with the participatory approach. This will help to reach out to stakeholders and policymakers, and thus identify the most promising strategies for pollution control.

6.4.2 Future recommendation for Agricultural Green Development

Currently, China is facing big challenges in balancing rising food demand and worsening water quality. In 2020, China launched a new national policy focusing on promoting Agriculture Green Development (AGD) (Davies and Shen 2020; Shen et al. 2020). The AGD aims to transform the current high resource consumption and high environmental cost agriculture to "green agriculture". The "green agriculture" reflects a new agriculture system with high productivity, high resource use efficiency and low environmental impact in a green countryside. Therefore, I consider that my thesis is relevant and can contribute to AGD. One of the four themes of AGD is integrating animal-crop production, which is in line with the main findings of Chapter 3: replacing synthetic fertilizer with animal manure is an efficient option to reduce nutrient pollution in water. In Chapters 3 and 4, I provide illustrative examples to use integrated water quality models to explore social and technical solutions for water pollution in China. Some objectives of AGD can be translated as future desired targets. The back-casting approach and optimization models can be combined with water quality models to explore strategies to achieve some objectives of AGD. To reduce the environmental impacts, AGD emphasizes efficient use of synthetic fertilizer, animal manure, pesticides, plastics and antibiotics in the food production system. This is relevant to Chapter 5 of this thesis. In Chapter 5, I proposed a framework that combines optimistic scenarios and optimization approaches with an integrated large-scale multi-pollutant model for water quality assessment. This framework can serve as a starting point to explore solutions for water pollution caused by multiple pollutants in food production.

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Summary

China is the largest consumer of pesticides and synthetic fertilizers of the world. These agrochemcials are essential for Chinese food production. However, this results in losses of pesticides and nutrients to surface waters. The increasing use of pesticides and fertilizers is associated with an increasing demand for food for an increasing population in China. In the future, the demand for agrochemicals may remain high as a result of socio-economic developments. Moreover, global change may affect water pollution.

Models can help to better understand water pollution. MARINA 1.0 (Model to assess River Inputs of Nutrients to seAs) is the first comprehensive model that quantifies river export of nutrients to Chinese seas by various types of point and diffuse sources, and the Indicator for Coastal Eutrophication Potential (ICEP) for six large Chinese rivers. Modelling studies for pesticide pollution in Chinese waters are scare. Existing pesticide models use empirical based approach and their results are relatively uncertain.

Models can help to explore options to reduce water pollution. Earlier studies often use scenario analyses to quantify the technical potentials of available options for reducing river export of pollutants. Models can be used for scenario-based forecasting, but also for back-casting and mathematical-based optimization. Back-casting can be used to identify combination(s) of options that can reach the given environmental targets. Optimization can help to explore solution(s) to reach the environmental target under given constrains. So far, back-casting and optimization analyses have not been applied to a large extent in studies of water pollution.

Food production is the major sources of both nutrient and pesticide pollution in surface waters in China. Current water quality models often focus on one single pollutant and ignore interactions between pollutants. Clearly, there is a need for large-scale water quality models to take a multi-pollutant perspective.

Therefore, the main objective of this PhD thesis is to explore control strategies to reduce future water pollution in China, caused by nutrients and pesticides. To this end, I formulated four specific research sub-objectives and these four sub-objectives form the four research chapters of this thesis:

- 1. To analyze future potential pesticide losses to Chinese waters under global change (Chapter 2)
- 2. To explore the possibilities to avoid coastal eutrophication in the North China Plain (Chapter 3)
- 3. To quantify desired nutrient pollution levels for sub-basins of Yellow, Yangtze and Pearl river from both an environmental and equality point of view (Chapter 4).
- 4. To draw lessons from air pollution control for large-scale water quality assessments, where multi-pollutant approaches are more common (Chapter 5).

In Chapter 2, I analyzed the potential for pesticide losses from near-stream agriculture to surface waters in the past and future under global change. To this end, I developed a pesticide model for China based on an existing global insecticide model with updated local information. My model runs for 2000, 2010, 2050 and 2099 at a 30 by 30 arcminutes grid. For the future, I developed three scenarios by combining the Shared Socio-economic Pathways and the Representative Concentration Pathways. My scenarios are the Sustainability (SSP1-RCP2.6), "Middle of the Road" (SSP2-RCP2.6) and Economy-first (SSP5-RCP8.5) scenarios. I concluded that the total pesticides runoff to surface waters increased by 45% from 2000 to 2010 in the past. In the future, total pesticide runoff may remain at the level of 2010 in the Sustainability scenario or be considerably higher in the Economy-first scenario. In the future, 58% to 84% of the Chinese population is projected to live in pesticide pollution hotspots. This range is for the scenarios and years. These pesticide hotpots are mainly in the Yangtze, the North China and Chengdu Plains, where agricultural production is more intensive and/or where precipitation levels are relatively high.

In Chapter 3, I focused on pollution control strategies to reach environmental targets for river export of nutrients to avoid coastal eutrophication in the future. To this end, I combined two scenario approaches using MARINA 1.0: forecasting and back-casting. I set environmental targets for nutrient levels at the river mouths of the Yellow, Huai and Hai Rivers. These targets were derived from Indicator for Coastal Eutrophication Potential (ICEP). Next, I focused on the question how to reach these targets (back-casting). To this end, I developed 54 possible combinations of control strategies that reflect different levels of improving animal feed, recycling animal manure, and increasing wastewater removal efficiencies. I ran the model to analyze the impact of

these 54 control strategies on water pollution, compared to a baseline scenario (forecasting). I concluded that 50 to 90% reductions in river export of nutrients are required to meet the environmental targets and thus to avoid coastal eutrophication in 2050. This range is for rivers and nutrient forms. It is possible to meet the targets in different ways. I show that 7-25 combinations of control strategies reduce river export of nutrients to the target levels. Recycling animal manure to replace synthetic fertilizer is the most efficient strategy in these combinations.

In Chapter 4, I focused on equality in pollution control. I explored solutions for coastal eutrophication in the Yangtze, Yellow and Pearl rivers considering socio-economic equality. To do this, I integrated a mathematical based optimization model (Gini) with MARINA 1.0 and ICEP to quantify allowable levels for river export of nitrogen (N) and phosphorus (P) at river mouths of sub-basins. The allowable levels for nutrient pollution are defined as the levels of river export of nutrients that ensure low risks for coastal eutrophication while accounting for socio-economic equality for sub-basins of these large river. It shows that it is possible to avoid coastal eutrophication while considering equality in socio-economic development in Yellow, Yangtze and Pearl rivers. I conclude that reducing both point and diffuse source pollution are required to reduce N to the allowable levels. Allowable levels for P could be reached by reducing only point source pollution from manure and sewage.

In Chapter 5, I drew lessons from air pollution control for large-scale multi-pollutant water quality assessments. I conclude that integrated models are successful tools to find solutions for transboundary air pollution problems. Models can help to explore optimistic and optimal solutions for multiple pollutants. A next step could be to integrate participatory approaches with environmental pollution models. Based on these lessons, I proposed a framework to integrate optimistic scenarios and optimization approaches with large-scale water quality models. I provide examples of integrating environmental, societal and economic targets with water quality models to search for solutions that are technically, economically and socially feasible.

In Chapter 6, I discussed the main findings and lessons of this thesis. These lessons are 1) The pesticide and nutrient models of this thesis are useful tools to identify hotspots and project future trends for water pollution in China and elsewhere, 2) Food production is often a common source of nutrients and pesticides in waters, calling for a need to

develop multi-pollutant approaches in research and pollution control, 3) Combining back-casting and forecasting approaches can support the search for effective strategies to reach desired future targets for clean water, 4) Combining optimization analysis with MARINA 1.0 is useful support if we want to minimize socio-economic inequality in water pollution control. I summarized improvements that are needed for future research in this chapter, and I recommend to develop models assessing water pollution caused by multiple pollutants. Additionally, I recommend to use this thesis as illustrative examples of combining back-casting and optimization analysis with water quality models, searching for options to reach the Agriculture Green Development targets.

Appendix

Appendixes contain additional information to the following chapters of the PhD thesis:

- Chapter 2 (to be submitted as Li et al.)
- Chapter 3 (published as Li et al., 2019)
- Chapter 4 (under revision as Li et al. under revision)

The text, figures and table of the Appendixes from the published article and the articles under revision and to be submitted have been adjusted to the PhD thesis format (e.g., the numbering and formatting). This includes editorial changes for the consistency of presentation in this PhD thesis. The adjusted PhD thesis version of the Appendixes is available on request (your email here). The published versions of the Appendixes are available online with the published articles.

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About the author

Ang Li was born on 27 October 1989 in Bayannur City in China. In 2008, she studied International Water Management as her BSc major in China Agriculture University. In 2010, she went to Van Hall Larenstein University of applied science in Arnhem, the Netherlands to continue her BSc program on the International Water management. In 2012, she continued her BSc minor at Wageningen University. Later that year, she started her MSc



in Environmental Sciences at Wageningen University. During her first four years in Wageningen, she was board member of the Chinese Association for Students and Scholars in Wageningen (CASSW). In 2014, she started her MSc thesis with Prof. dr Carolien Kroeze and dr Maryna Strokal. In 2016, she continued her research career with Prof. dr Carolien Kroeze and dr Maryna Strokal in Wageningen University. During her PhD, she joint the PhD council of Wageningen Institute for Environment and Climate Research (WIMEK). In addition to her research, she was also active in educating and supervising students. She strives for strengthening the Sino-Dutch collaboration on reducing water pollution caused by nutrients. Results of her study have been presented in many international conferences and symposiums as posters or oral presentations.



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The Netherlands research school for the Socio-Economic and Natural Sciences of the Environment (SENSE) declares that

Ang Li

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has successfully fulfilled all requirements of the educational PhD programme of SENSE.

Wageningen, 23 March 2021

Chair of the SENSE board

Prof. dr. Martin Wassen

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





The SENSE Research School declares that **Ang Li** has successfully fulfilled all requirements of the educational PhD programme of SENSE with a work load of 41.2 EC, including the following activities:

SENSE PhD Courses

- o Environmental research in context (2016)
- Research in context activity: 'Organizing Trip to China in 2017 including: I Model training in China Agricultural University, II Project kick off meeting in Nanjing Agricultural University, III Annual progress meeting for WUR-CAS collaboration in CAS' (2017)

Other PhD and Advanced MSc Courses

- o Data management planning, Wageningen Graduate School (2016)
- o Introduction to R for statistical analysis, Wageningen University (2016)
- o GIS in practice, Wageningen University (2016)
- o Competence assessment, Wageningen Graduate School (2017)
- o Scientific writing, Wageningen Graduate School (2018)
- o The essential for SW and presenting, Wageningen Graduate School (2018)

Selection of Management and Didactic Skills Training

- Co-organisation of trip to China including symposia, workshops and field trips (2017-2019)
- Supervising two MSc students with thesis entitled 'Modelling the potential risk of insecticide runoff to surface waters in China' (2018) and 'Seasonality in herbicides exports from land to aquatic systems in China: exploring the possibilities to link parts of the MARINA model and Insecticides model' (2019)
- o Assisting practical of the BSc/MSc course 'Introduction to Global Change' (2016)
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- The effect of 'Double High Agriculture' on nitrogen losses from crop production to coastal waters in China. The 7th International Nitrogen Initiative Conference, 4-8 December 2016, Melbourne, Australia.
- o Potential pesticide losses to Chinese waters: some preliminary results. SuRe Food Interdisciplinary seminar .4 December 2019, Wageningen, The Netherlands.

SENSE coordinator PhD education

Dr. ir. Peter Vermeulen

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