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DTU Environment
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PhD Thesis
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The synopsis part of this thesis is available as a pdf-file for download from the DTU research database ORBIT: http://www.orbit.dtu.dk

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Preface

This PhD thesis is based on the work conducted from 2011 to 2015 at the Department of Environmental Engineering, Technical University of Denmark (DTU), and at the Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences (CAS). All field work was completed during an external stay at the Chinese Academy of Sciences, Beijing, China, from October 2012 to March 2014. The project was supervised by Professor Poul L. Bjerg (main supervisor), Senior Researcher Rasmus Jakobsen (co-supervisor), and Professor Xianfang Song (co-supervisor).

The research was funded by the Sino-Danish Center for Education and Research (SDC), Aarhus, Denmark, and the Department of Environmental Engineering, Technical University of Denmark, Kongens Lyngby, Denmark.

The PhD thesis is organized in two parts: The first part puts the finding of the PhD into context in an introductive review, including a field report from the study area. The second part consists of the papers listed below. These will be referred to in the text by their paper number written with the Roman numerals **I-III**. Paper **I** has been accepted in the Journal of Environmental Sciences (JES). Papers **II** and **III** have been submitted, but not yet accepted.

I Brauns, B., Bjerg, P.L., Song, X., Jakobsen, R. 2015. Field scale interaction and nutrient exchange between surface water and shallow groundwater in the Baiyang Lake region, North China Plain. In press.

II Brauns, B., Jakobsen, R., Song, X., Bjerg, P.L. 2015. Pesticide use in the wheat-maize double cropping systems of the North China Plain: Assessment, field study, and implications. Submitted.

III Brauns, B., Song, X., Jakobsen, R., Bjerg, P.L. 2015. Vocational training for farmers: A long-term approach to the alleviation of environmental pollution from agriculture in China. Submitted.

In this online version of the thesis, paper **I-III** are not included but can be obtained from electronic article databases e.g. via www.orbit.dtu.dk or on request from DTU Environment, Technical University of Denmark, Miljoevej, Building 113, 2800 Kgs. Lyngby, Denmark, info@env.dtu.dk.

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The installation and sampling at my Chinese field site could not have been done without the generous and sturdy support of my PhD colleagues from the Institute of Geographic Sciences and Natural Resources Research. Special thanks go to Wenjia, Xiangmin, Bing, Zhenyu, Yilei, and Baogang for working with me in the field with under all kinds of conditions. Thanks as well to farmer Ma in Dongxiangyang Village, for his agreement to use his land, and his creativity in solving any problem at the field site. For kindly sharing his expertise on the case study area, I am grateful to Ruiqiang Rui. Thanks also to my entire PhD office in Beijing for receiving me with open arms in your group. I will always keep our discussions, travels with the research group, and long dinners as a very warm memory, and I hope to see many of you again in the future.

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And last, but certainly not least, thanks to Jan for being there for me all this time, and to my sister and parents for all the encouragement, which carried me through the years of this study.

Summary

The North China Plain is one of China's major economic zones and one of the most densely populated areas in the country. It covers a broad expanse of eastern China, extending from just below Beijing in the north down towards the Yangtze River in the south. This alluvial plain region is also one of China's main agricultural production zones, accounting for about one third of the national grain output. The dominant crop system is a winter wheat and summer maize rotation. Beginning in the 1980s, in an effort to increase agricultural productivity, China's government heavily promoted the use of fertilizers and pesticides. Unfortunately, the lack of regulation or oversight has led to the overuse of these agrochemicals: current application rates (in kg/ha) are two- to threefold higher than in most developed countries, and this is taking its toll on the environment. Problems include severe surface water and groundwater pollution by nitrogen and pesticides, soil degradation, bioaccumulation of toxic compounds, and more. It is crucial for China to do improve the safeguarding of its water resources in order to sustain the livelihoods of its people and ensure safe supply of drinking water.

Recently, the Chinese government and the scientific research community have acknowledged the need for more sustainable production techniques, and increasing quantities of money and effort are being directed toward achieving this goal. There has already been a great deal of improvement in determining the appropriate amounts of agricultural inputs, such as irrigation and fertilizers, as well as the ideal times to apply them. In terms of pesticides, most studies have focused on pesticide residues, crop resistances, and on the efficient treatment of specific pests. Despite this groundwork, however, statistical records show that the application of agrochemicals per hectare continues to increase, and the water quality within the river basins of the North China Plain remains substantially worse than in other parts of the country.

Taking this background into consideration, this PhD study focused on four different objectives: (a) to quantify the nutrient loading in groundwater and surface water at a sample field site in order to understand their exchange and removal pathways; (b) to review the current use and monitoring of pesticides (and especially herbicides) in the NCP; (c) to assess the occurrence of selected herbicides at a sample field site in the NCP; and (d) to provide a new basis for discussion and guidance on how to address the issue of water pollution caused by the improper use of agrochemicals in China. For the field investigation, a study site located within the NCP with river water-groundwater interaction was chosen, and field work was performed between October 2012 and March 2014.

Results from the field study showed that fertilizer inputs were excessive, and could be reduced substantially. Contaminated river water was infiltrating – and carrying ammonium pollution – into the shallow groundwater. Additionally, nitrate was infiltrating from the surface of the field into the aquifer. Anammox, denitrification, and cation exchange were the suggested dominant removal processes in the soil-surface water-groundwater system examined in this study, which showed a very high nitrogen removal capacity. However, if the composition of the river water were to change (if, for instance, the ammonium concentration were to decrease) the removal processes in the system would also be altered. Consequently, further monitoring of nitrate pollution is suggested.

Regarding pesticides, a literature review and data assessment revealed that the most commonly applied herbicides in the North China Plain wheat-maize cropping system are 2,4-D, acetochlor, and atrazine. Although 2,4-D and atrazine are listed in the Chinese Drinking Water Guideline, there is currently no systematic monitoring of these compounds taking place, and most research studies have focused on the monitoring of legacy pesticides such as hexachlorocyclohexanes (HCHs) and dichlorodiphenyltrichloroethanes (DDTs).

In the river water and groundwater samples drawn during this study, mainly 2,4-D and atrazine residues, were discovered in concentrations of several µg/l (these results were consistent across all four sampling campaigns). Most of the pollution seemed to have been caused by the river water carrying pesticides into the groundwater system. This indicates that it may be important to pay more attention to the investigation of currently-used pesticides, especially in areas where surface water infiltrates into shallow aquifers.

The overall observation on agricultural activities in the North China plain was that much improvement is needed in educating farmers on sustainable production techniques and the proper application of agrochemicals. One way to increase farmers' understanding and knowledge of the environmental impact of agriculture would be to shift to a more formal training regime, for example vocational education. One possible side effect of such a change could be to raise the status and income opportunities enjoyed by agricultural workers, thereby giving the younger generation an incentive to choose farming as a profession.

In conclusion, this PhD study gave insights into a more systemic understanding of nutrient degradation and the occurrence of particular herbicides at a specific field site. The research into the use and monitoring of pesticides in the North China Plain was reviewed and new recommendations were developed to enhance the dissemination of knowledge from environmental researchers to farmers.

Dansk sammenfatning

Den Nordkinesiske Slette (DNS) er en alluvial region i det østlige Kina, som strækker sig sydover fra syd for Bejing til Yangtze floden. Regionen er et landbrugsområde, der producerer ca. en tredjedel af landets hvede. Den primære afgrøde er vinterhvede i rotation med sommermajs. Den Nordkinesiske Slette er desuden et af de tættest befolkede områder i Kina, og der er stor økonomisk aktivitet. Den kinesiske regering fremmede anvendelsen af gødning og pesticider i 1980'erne for at øge produktiviteten. Desværre er forbruget af disse landbrugskemikalier steget ukontrolleret, og i dag sker der et overforbrug (i kg/ha) på to til tre gange mere end i de fleste i-lande. Miljøpåvirkningerne til følge inkluderer alvorlig overfladevands- og grundvandsforurening med nitrogen og pesticider, ødelæggelse af jordbunden og bioakkumulering af toksiske stoffer. Bedre beskyttelse af vandressourcerne er afgørende for at opretholde levebrød og en sikker drikkevandsforsyning.

Den kinesiske regering og det kinesiske forskningssamfund har anerkendt behovet for bæredygtige produktionteknikker og der investeres for at nå dette mål. Med hensyn til passende anvendelse og timing af kunstvanding og kunstgødning er der sket fremskridt. Men hensyn til pesticider har de fleste studier fokuseret på pesticidrester, afgrøderesistens og effektiv behandling af specifikke skadedyr. På trods af dette arbejde stiger anvendelsen af landbrugskemikalier per hektar, og vandkvaliteten i de hydrologiske oplande på den Den Nordkinesiske Slette er stadig væsentligt ringere sammenlignet med andre egne i Kina.

Set i lyset af dette, har dette ph.d.-studie haft fire mål: (a) at kvantificere næringsstofbelastningen i grundvand og overfladevand med henblik på at forstå udveksling og primære fjernelsesveje gennem et feltstudie; (b) at redegøre for nuværende brug og overvågning af pesticider (særligt herbicider) på DNS; (c) at vurdere forekomsten af udvalgte herbicider på et case site på DNS; og (d) at fremsætte et nyt grundlag for diskussion og vejledning til håndtering af vandforurening forårsaget af uhensigtsmæssig anvendelse af landbrugskemikalier i Kina. Feltundersøgelserne blev udført på en lokalitet på DNS med grundvand-overfladevandsinteraktion, hvor feltarbejdet med blandt andet vandprøvetagning blev gennemført i perioden marts 2013 til marts 2014.

Resultater fra feltområdet viste et stort forbrug af kunstgødning, som kan reduceres betydeligt. Forurenet flodvand, med blandt andet ammonium, infiltrerede til det overfladenære grundvand. Derudover var der en nedsivning af nitrat fra jordoverfladen til grundvandsmagasinet. Anammox, denitrifikation og ionbytning så ud til at være de primære fjernelsesprocesser i jord-, overflade-, grundvandssystemet, som viste en høj nitrogenfjernelseskapacitet. Hvis overfladevandssammensætningen ændrer sig (for eksempel til at have en lavere ammonium koncentration) ændres også fjernelsesprocesserne og øget nitratovervågning anbefales derfor.

Dataanalyse og et litteraturstudie afslørede at herbiciderne 2,4-D, acetochlor, and atrazine er de mest anvendte i hvede-majs afgrødesystemet på DNS. Selvom 2,4-D og atrazine er angivet i "Chinese Drinking Water Guideline" findes der ingen systematisk overvågning af stofferne, og de fleste videnskabelige studier forkuserer på overvågning af gamle pesticider, som for eksempel hexachlorocyclohexanes (HCHs) and dichlorodiphenyltrichloro-ethanes (DDTs).

Vandprøver af flod- og grundvandet samlet under dette ph.d.-studie viste rester af 2,4-D og atrazine på flere μg/l (fra alle fire vandprøvesamlingskampagner). Forureningen vurderes at være forårsaget af overførsel af pesticider fra flodvandet til grundvandet. Det indikerer, at det er vigtigt at undersøge den nuværende pesticidanvendelse, særligt i områder med infiltration af overfladevand til overfladenære grundvandsmagasiner.

Generelt observeres for landbrugsaktiviteter på DNS et udtalt behov for uddannelse af landmænd indenfor bæredygtige produktionsteknikker og hensigtsmæssig anvendelse af landbrugskemikalier. En måde at opnå dette kunne være at øge forståelsen for og viden om landbrugets miljøpåvirkninger ved at indføre formel uddannelse af landmænd, for eksempel faguddannelse. Uddannelse kan også føre til forbedret status og indtægtsmuligheder for landbrugsarbjedere, og på den måde være incitament for den yngre generation til at vælge landbrugssektoren som en profession.

Dette ph.d.-studie har givet indsigt og forståelse af kvælstofstofkredsløbet og forekomsten af udvalgte herbicider på en feltlokalitet. Der er redegjort for pesticid -anvendelse, -overvågning og -forskning på Den Nordkinesiske Slette, samt udviklet nye anbefalinger til at forbedre vidensdeling mellem miljøforskere og landmænd.

摘要

中国北部平原指的是位于中国东部地区的冲积平原区域，从北京的南部延伸至南部的长江。它是一块主要的农业生产区域，占据了全国谷物产量的三分之一。主要的庄稼系统是冬小麦和夏玉米的更替。中国北部平原也是主要的经济区，以及中国人口最密集的区域之一。中国政府在 1980 年代大力地推广化肥技术和农药技术来提升产值。不幸的是，这些农业化学品的使用逐渐地失去控制，以至于目前过度使用的状况（使用量比大多数发达国家高出两到三倍）。这以环境作为了代价，负面的影响包括了地表水和地下水遭到了氮气和农药的严重污染，土壤退化，有毒化合物的聚集，以及更多。对水资源采取更好的安全措施，对于民生的可持续发展以及饮用水的安全供给至关重要。

中国政府和学术领域都已经认识到了对于更加可持续性的生产技术的需要，并且正在投入越来越大的资金和努力来实现这个目标，其中包括了在灌溉和化肥使用方面规定更加合理的使用次数和数量。在农药使用方面，越来越多的研究关注于农药的残留，农作物的抗性，特定害虫的有效处理。虽然这些基础性的工作都在进行，但是统计数据记录显示农业化学用品在每公顷上的使用量仍然在上升，中国北部平原的江河流域内的水质量仍然持续地比中国其他地区更加恶劣。

根据这些背景情况，这份博士研究关注于四个不同的目标，(a) 量化地下水和地表水中的营养负荷，以了解它们在给定的实验区域内的交换和主要去除途径；(b)调研在中国北部平原地区内目前农药的使用和监管（尤其是灭草剂）；(c) 在中国北部平原地区的实验区域内，评估选取的灭草剂的使用状况；(d)为中国不适当地使用农业化学用品导致的水污染现状提供一个新的探讨依据和指导方针。关于实地考察方面，选取了一个中国北部平原地区的实验地点，来研究河流地下水的交互情况，在这个河流中，对河床的生物带，土壤水以及地下水进行了采样，实验时间从 2013 年 3 月持续到 2014 年 3 月。

实地考察的结果显示化肥施用过度，其实可以被大幅度降低。 污染的河水正在向浅层地下水渗入，并且携带了铵污染。另外，硝酸盐正在从场地表层向地下蓄水层渗入。 被研究的土壤-地表水-地下水体系表现出非常高的氮去除容量，厌氧氨氧化，反硝化作用以及阳离子交换被认为是主要去除途径。但是，如果河流水的成分改变了（比如更低的铵浓度），那么整个系统的去除程序就会有相应变化，因此硝酸盐污染就需要被更加深入地进行监控。
在农药方面，文献和数据的分析显示在中国北部平原的小麦 - 玉米庄稼系统中最通常使用的灭草剂是二氯苯氧乙酸（2,4-D）， 乙草胺（Acetochlor）和莠去津（Atrazine）。虽然二氯苯氧乙酸（2,4-D）和莠去津都被在列中国饮用水标准中，但是并没有系统性对于这些复合物的监管，大部分的研究只是关注于农药残留的监管，比如，六氯环己烷（HCHs）和 二氯二苯三氯乙烷（DDTs）。

在这份博士研究的河流水和地下水的样本中，主要发现的是二氯苯氧乙酸（2,4-D ）和 莠去津的残留（通过四次采样）。大多数的污染是由河流水携带农药进入地下水系统导致的。这表明了更多地关注于现用农药的监管，尤其是地表水渗入浅含水层的区域尤为重要。

对于中国北部平原的农业活动的总体观察来看，在可持续的生产技术以及农业化学用品的合理运用方面，对于农民的教育是必须的。实现这一点的一个途径可以是提升对于农业的环境影响方面的知识和理解，向农民提供更加正式的培训，比如职业教育。这将带来农业从业者在社会地位和收入上的提升，也为年轻的一代选择农业作为职业提供了激励。

综上所述，这份博士研究对于养分降解，在特定实验场地的专门灭草剂的使用方面提供了观点。对于中国北部平原地区的农药使用、监管和研究进行了调研，并且在加强农业研究和对农业从业者的知识提升和教育普及方面提出了新的观点和建议.。

Table of contents

Abbreviations

Anammox	Anaerobic ammonium oxidation
BY Lake	Baiyang Lake (Baiyangdian)
COD	Chemical oxygen demand
DDT	Dichlorodiphenyltrichloroethane
DO	Dissolved oxygen
EC	Electric conductivity
HCH	Hexachlorocyclohexane
MEP	Ministry of Environmental Protection
N	Nitrogen
NCP	North China Plain
NECP	Northeast China Plain
OCP	Organic chlorinated pesticide
P	Phosphorus
P_{CO_2}	Partial pressure of carbon dioxide
SI	Saturation index
T	Temperature
TC	Total carbon
TIC	Total inorganic carbon
TOC	Total organic carbon

1 Introduction

1.1 Motivation

Within the People's Republic of China (from here on referred to as China), "an adequate degree of self-reliance in food" is considered an issue of state security, which is a premise for economic development and social stability (Li et al., 2014). It is therefore an integral part of the national agenda to maintain the balance between agricultural production and demand. However, China has 20% of the world's population to feed, but only 7% and 6% of the world's arable land and freshwater resources, respectively (World Bank Group, 2015). Furthermore, the distribution of the agricultural production zones across the country is very uneven, mainly because of regional differences in topography, geography and climate. Therefore, China relies heavily on efficient agricultural production in specific regions, which results in high local pressures.

Cereals like rice, wheat, and maize are the most important staple foods produced in China. The main agricultural zones are the Northeast China Plain (NECP, cultivation of different grains in the warm months of the year), the North China Plain (NCP, perennial farming activities, mostly in form of irrigated winter wheat and rain-fed maize), and the Yangtze region (the most important region for rice production). Among these three regions, the NCP experiences the highest pressures because of a) its high population density, b) its intense use for farming, industry and human settlement, and c) its strong limitation in water resources. These pressures have led to an ongoing observed environmental decline showing soil degradation (Ye and van Ranst, 2009), soil pollution (Wang et al., 2007), declining groundwater levels at rates of up to 1 m per year (Liu et al., 2003), and deterioration of both groundwater and surface water qualities (Economy, 2007). Especially industrial fertilizers and pesticides, and discharge of animal manure into open waterways show negative effects. According to the 2007 national pollution census, 57% and 67% of the national nitrogen and phosphorus pollution (respectively) are caused by agricultural inputs (Qiu, 2011).

Studies have shown that nitrogen fertilizers are often applied in amounts 30-60% higher than recommended (Ju et al., 2009). Additionally, application times and methods are often inappropriate (such as surface application of urea, which causes high volatilization), leading to low nitrogen use efficiencies (Zhang et al., 2013). The Chinese government and research community increasingly acknowledged and addressed these negative impacts in recent decades, and environmental protection became one of the key points on the national five-year plan from 2001 on.

Much research focus has been given to water, crop, and fertilizer management in regions heavily impacted by agriculture (e.g., the NCP). Levels of nitrate pollution have been assessed in different environmental compartments, such as surface water and groundwater (Chen et al., 2005; Huang et al., 2011; Sun et al., 2012). Numerous studies give recommendation on appropriate fertilizer applications in different regions of China based on their previous assessment of nitrate leaching (Chuan et al., 2013; He et al., 2013; Ju et al., 2007; Xue and Hao, 2011). Additionally, a variety of in-situ studies tested different cropping systems and/or different fertilization amounts under field conditions in order to define best farming practices for increased yields (Dai et al., 2013; Fan et al., Gao et al., 2014; Li et al., 2007; Liu et al., 2010; Sun et al., 2007; Tan et al., 2013). Some studies aimed to identify the role of volatilization and denitrification within the soil system (Hartmann et al., 2014; Liu et al., 2003; Zou et al., 2006). However, the share of studies that assess the dynamics of nitrogen removal processes within the soil system is generally low compared to the vast amount of studies on nitrogen application and leaching. Though it is certainly useful that the level of research on implementation measures is high, more knowledge on the fate of pollutants in the Chinese environment, and especially the identification of interaction pathways between soil, groundwater and surface water would build a useful premise for vulnerability assessment. This could enhance China's progress in expanding the national water monitoring network in the right places.

Comprehensive data on current pesticide applications and monitoring data—if existing—is difficult to obtain. Research on pesticides seems to have a strong focus on legacy pesticides, such as dichlorodiphenyltrichloroethanes (DDTs) and hexachlorocyclohexanes (HCHs), which have been investigated in numerous studies, and especially in surface waters and sediments (Dai et al., 2011a; Dai et al., 2014; Wang et al., 2009a; Wang et al., 2013). Pesticide accumulation e.g., by means of bioaccumulation of DDTs and HCHs in the food web, has been given great attention (Dai et al. 2011b; Dou and Zhao, 1997; Hu et al., 2010; Yang et al., 2006; Zhu et al., 1999). Only few (and oftentimes international) studies include a larger set of pesticides (Deuerlein and Hurle, 2001; Domagalski, 2001; Xue and Xu, 2006), or even focus on currently-used pesticides only (Geng et al., 2013; Ren and Jiang, 2002a; Ren and Jiang, 2002b). This narrowly focused research on legacy pesticides seems to neglect the study on transport and fate of currently-used pesticides in the Chinese environment. Additionally, there is a strong focus on surface water and soil sampling, and less focus on groundwater systems. This makes it difficult to assess the dynamic interaction and contaminant transport between groundwater and surface water.

1.2 Aims

Based upon the background of the overuse and high environmental impact of agricultural inputs in China, this PhD research focused on selected aspects of agricultural practise and interactions of agricultural pollutants between surface water and groundwater. The aims of the research were:

- To improve the understanding of nutrient (especially nitrogen) exchange between surface water and groundwater, and to identify major removal pathways at an exemplifying field site in the NCP (Paper **I**).
- To review current use of pesticides in the NCP, and to assess research (and monitoring) of pesticides in groundwater and surface water in the NCP (Paper **II**).
- To assess the occurrence and temporal variation of selected herbicides at an example field site in the NCP (Paper **II**).
- To provide a new base of discussion and guidance on how to address the issue of water pollution caused by improper use of agrochemicals in China (Paper **III**).

1.3 Structure

General characteristics of agriculture in China and in the NCP are presented as background in Chapter 2. Chapter 3 introduces the Baiyang Lake (BY Lake) area, including a report of a field excursion for the initial area assessment, and a description of the study site, at which the field work for Paper **I** and Paper **II** was performed. Chapter 4 outlines the data collection an assessment of the nutrient and pesticide study that was performed within the framework of this PhD study. Chapter 5 discusses the results and implications from the field study and summarizes insights that on agricultural management and farmer’s knowledge in China as well as research challenges, such as data acquisition and analysis. Conclusions and perspectives are given in Chapter 6 and Chapter 7, respectively.

2 Background: Agriculture in China and in the North China Plain

This chapter gives an overview over the division of the different agricultural regions in China. The limiting factors for grain production in the main agricultural regions will be discussed. Special focus is given to water challenges in the NCP.

2.1 Agricultural regions in China

China is a very diverse country with different climates ranging from arid to humid, and with distinct topographic regions. Accordingly, agricultural areas are unevenly distributed. Little arable land is located in the dry western, and the mountainous central region of China, while more fertile lands aggregate towards the densely populated eastern part of China (Figure 1).

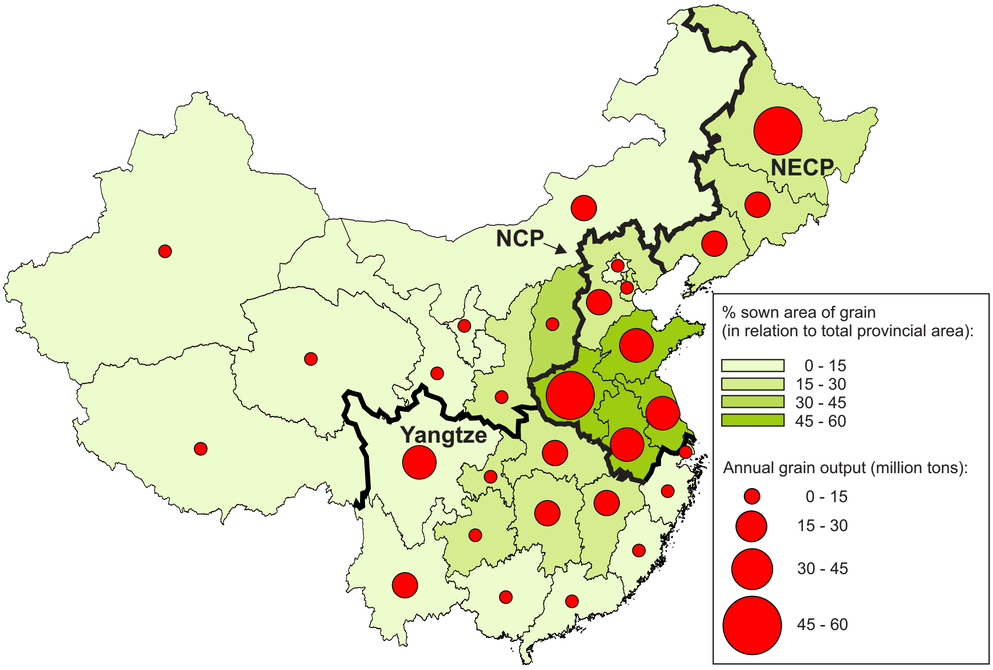


Figure 1. Percentage of sown area of grain with respect to the size of each province (indicated by colored area) and provincial grain production (indicated by circles) in mainland China. Delineation of the provinces that constitute the Northeast China Plain (NECP), the North China Plain (NCP), and the Yangtze region are printed in bold. "Sown area" corresponds to arable land times the Multiple Cropping Index (the frequency of planting a crop on the same arable land in one year). According to the definition used by the Chinese National Bureau of Statistics "grain" refers to cereals, legumes, and also tuber crops. (The figure was produced based on data from the National Bureau of Statistics, 2011.)

Most of China's arable land is concentrated in the central and eastern part of the country, which exhibits more favorable conditions for agriculture than the western region (characterized by extreme aridity, mountains, and/or low fertile soils). The eastern, arable, region of China can be divided into 3 distinct agricultural zones: The Northeast China Plain (NECP), the North China Plain (NCP), which reaches from the southern part of Beijing to towards the Yangtze River, and the lower Yangtze region (Figure 1). These three regions are considered *China's grain baskets*, and they alone accounted for 72.3% of the national grain output in 2011 (Li et al., 2014).

The NECP is dominated by fertile lands and sufficient water. Different cereals are grown, but low temperatures and long winters limit the productivity in this region. The NCP on the other hand offers good opportunities for the cultivation of wheat and maize in a double-cropping system due to its flat terrain, shorter frost periods, and its continental monsoon-type climate. However, available water resources and the intensity of solar radiation are not sufficient for extensive paddy rice fields. Instead, these fields can be found in the Yangtze Region, which accounts for around 85% of the national rice production. In summary, the limitations in water resources are the constraining factor for agricultural production in the NCP, whereas grain production in the water-abundant Yangtze region is rather restricted by the area of arable land.

2.2 The North China Plain

The NCP is the most productive agricultural area in China, and produces about one quarter of the total national food production (Yang et al. 2014). Its area (approximately the size of Germany) encompasses the southern parts of Beijing and Hebei, the entire provinces Tianjin and Shandong, the eastern part of Henan, and the northern parts of Anhui and Jiangsu. It is delineated by the Yan and Taihang Mountain range in the North and West (respectively), the Bohai Sea in the East, and it merges into the Yangtze River Plain in the South. Three out of the seven major river basins lie (partially) in the NCP, namely the southern part of the Hai River Basin in the North, the easternmost part of the Yellow River Basin (which extends past the NCP into arid north-central part of the country), and the Huai River Basin in the South (Figure 2).

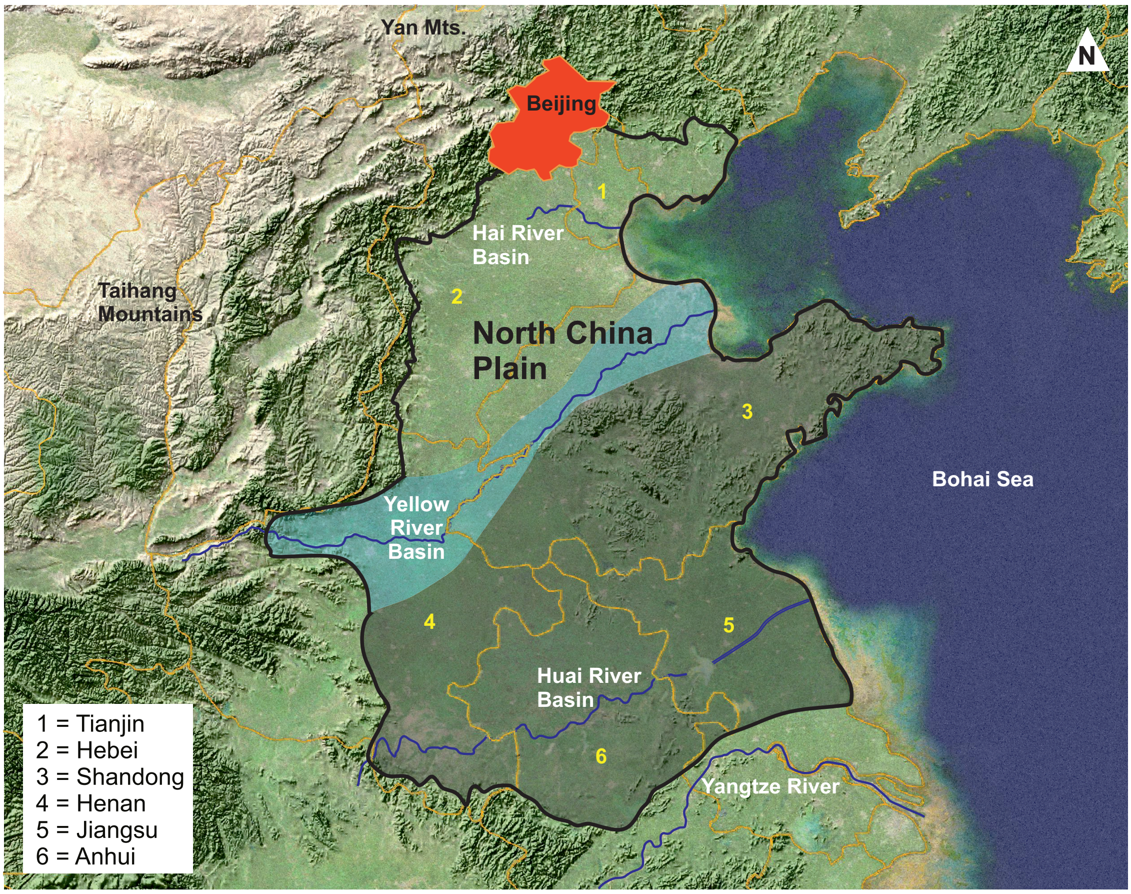


Figure 2. Location and topography of the North China Plain and approximate delineation of the Huai River Basin, and the parts of the Yellow and Hai River Basins that constitute the NCP (background map modified from Stewardship, 2015)

Most soils derive from silty and clayey alluvium, with a natural pH range of 7.5-8.5, and are very suitable for agriculture (FAO, 1978). The aquifer system consists of a 2-layered structure, with an unconfined and/or seimiconfined upper aquifer, and a lower aquifer with a confining boundary at 100-150m depth. Recharge occurs mainly from the mountain and piedmont plain, potentially by some fracture recharge from the lower layer into the upper layers (Yuan et al., 2013). The climate in the NCP is a continental monsoon climate with 70-80% of the rainfall occurring during the summer months, and very dry, but relatively mild winters. This allows for double cropping systems, such as the winter wheat-summer maize rotation, which is the most dominant agricultural production system in this area, covering 16 million ha of land (Liang et al. 2011).

2.2.1 Water challenges

Water resources, especially surface water, have a high spatial and temporal variability in the North China Plain. 70-80% of the rainfall occurs from July-September, which leads to peak flows (and sometimes flooding) in the summer and early fall. Most of the surface water recharge originates in runoff from the adjacent mountainous region of the NCP (Liu et al., 2001). About 60% of the water resources come from groundwater. The NCP experiences extremely high pressures on its water resources because of:

- High population density. The provinces of the NCP are not only some of the most agriculturally productive, but also some of the most densely populated provinces. With only 3.6% of China's land area, the two provinces Hebei and Shandong alone hold 12.6% of the country's population (National Bureau of Statistics, 2011, Figure 3).
- Water pollution. The high population density and high agricultural use of the region also increase pollution sources, while there is comparatively little renewable freshwater available. Taking the Shandong-Hebei area again as an example, it can be seen that 1.6% of the national renewable freshwater resources are subject to the effects of 13.6% and 14.2% of the national fertilizer and pesticide input, respectively (National Bureau of Statistics, 2011, Figure 3).
- Irrigation agriculture. The predominant type of cultivation is a winter wheat-maize double cropping system. Winter wheat in the NCP has an average crop water requirement of 450mm, of which the sparse rainfall during the crops growing period (typically October-June) can only meet 25-40% (Liu et al., 2001). Therefore, irrigation is a necessity in the NCP to achieve sufficient yields, and 70% of the available water resource is used for irrigation (Liang et al., 2011).
- Groundwater overexploitation. Surface water resources are often not sufficient, or not of acceptable quality for the intended water use (especially for domestic use). This led to heavy exploitation of the aquifer system, and water table declines range up to 1 m/year (Wang et al., 2002). The investment in massive projects—like the drilling of two ultra-deep wells in Beijing Province with a depth of about 1800 m and 1500 m that are intended to secure safe drinking water for the area—can be seen as an indicator of the severe water shortage in the area (Aswathanarayana, 2008).

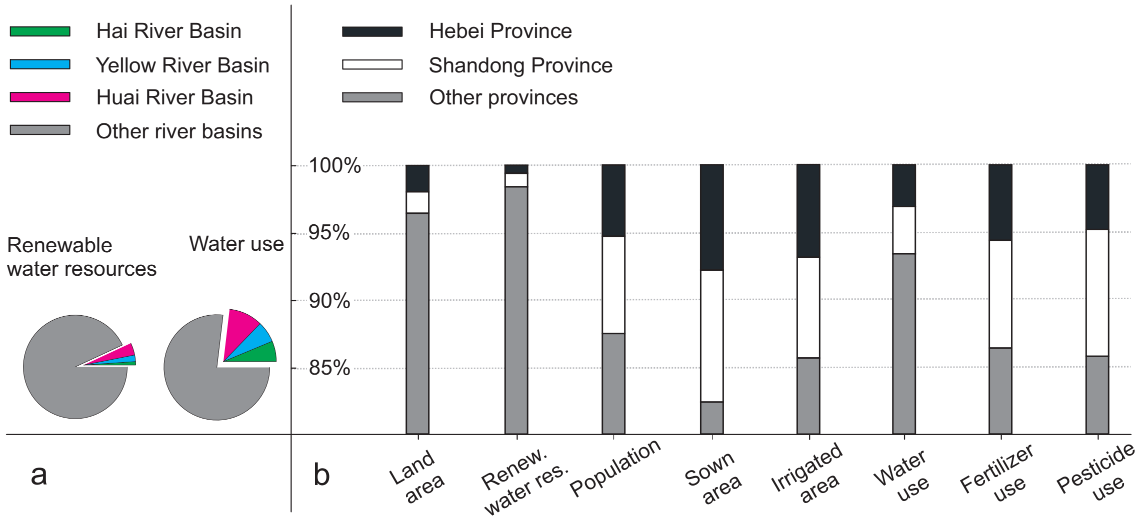


Figure 3. (a) Share of renewable freshwater resources and water use of the three river basins in the NCP in comparison with the national renewable freshwater resources and water use (Data according to the Ministry of Water Resources, 2008); (b) the share of the two provinces Hebei and Shandong of national resources (land area, renewable water resources), demand drivers (population, sown agricultural area, irrigated area), and resource use (water, fertilizer and pesticide input) (The figure was produced based on data obtained from National Bureau of Statistics, 2011).

2.2.2 Water level monitoring, and water quality

In order to assess and monitor regional groundwater levels, a first monitoring network with about forty monitoring stations was installed in the NCP in the 1950s (CIGEM, 2006). However, monitoring was often infrequent, e.g., caused by deterioration of sampling wells, and no measurement of water quality criteria was included (Wang et al., 2009b). Today, surface water quality is monitored nationwide at 972 state-controlled monitoring points. These include a total of 415 rivers and 35 lakes. 145 of the monitoring station are automatic monitoring systems that are distributed over 25 of the provincial-level subdivisions, and they monitor 63 rivers (including the seven major river basins), and 13 lakes and reservoirs. Measurements include temperature (T), pH, dissolved oxygen (DO), chemical oxygen demand (COD), electric conductivity (EC), turbidity, permanganate index, total dissolved organic carbon (TOC), ammonia and (only for lake water) total nitrogen (N) and phosphorus (P). According to the "Surface Water Quality Standards" (GB 3838-2002), these measurements are then used for water classification (CNEMC, 2009). Surface water quality reports are published weekly and monthly (National Surface Water Quality Report) by the Ministry of Environmental Protection (MEP).

As mentioned earlier in the text, there are seven main river basins in China: Songhua, Liao, Hai, Yellow, Huai, Yangtze, and Taihu River Basin (Figure 2). Figure 4 shows the results of the surface water quality monitoring by the MEP for the years 2009 and 2012 for the Hai River, Yellow River, and Huai River basin (the basins of the NCP), compared the other four main river basins in China. It can be seen that the average surface water quality in the river basins of the NCP is substantially worse than the average water quality status of the other four main basins in China (with the exception of the Yellow River Basin in 2009). Especially the surface water in the Hai River Basin is of very poor quality: About 40% of the Hai River Basin's water is classified as unsuitable for domestic, or even industrial and agricultural use and very little quality improvement occurs between the years 2009 and 2012. This means in theory, that only 2/3 of the surface water in the Hai River basin are suitable as irrigation water for agriculture. However, it will be seen in the following chapters of this PhD thesis that water from rivers classified with status lower than V is still often used for irrigation in the NCP.

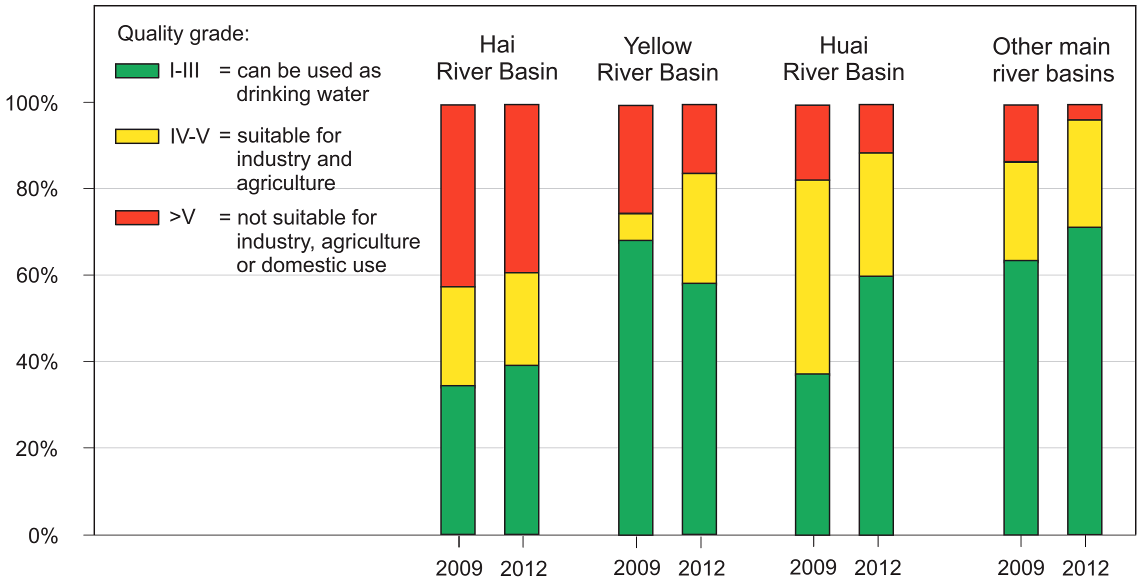


Figure 4. Water quality status of the three rivers basins in the North China Plain (Hai, Yellow, and Huai River Basin) for the years 2009 and 2012 in comparison with the quality status of the other main river basins (Songhua, Liao, Yangtze, and Taihu River Basin). (The figure was produced based on data from the National Reports on the State of the Environment in China, MEP, 2010 and MEP, 2013).

3 The Baiyang Lake study region

The BY Lake region was chosen as the study region for this PhD research. Reasons for this were a) the availability of perennial rivers and the lake region itself, enabling the study of interactions between surface water and groundwater, b) the shallow water table, which presumably coincides with a good connection between surface water and groundwater, c) the assumption—based on the national water quality reports—that water pollution would be severe, d) the close proximity to the research institute in Beijing. Within this chapter, the BY Lake study region will be presented. This will be done first by a brief introduction to the location, setting, agricultural use, and water resources of the region. Secondly, a field report of the area assessment will presented.

3.1 Location and setting

BY Lake is located in the central part of Hebei Province, approximately 150 km south of Beijing, and 40 km east of Baoding, Hebei's largest city by prefecture-level (Figure 5). BY Lake is considered to be the largest lake in northern China, even though it is rather a system of approximately 100 small lakes with an average depth of only 2 m that are connected by reed-covered, wetland-type channels.

Figure 5. Location of the Baiyang Lake Area, and of the different sites (1-4) that were visited for the area assessment. Site 2 is also the location of the field study that will be presented in chapter 4 (background map obtained from Google Inc., 2013).

The lakes encompass an area of about 362 km^2, and they play an important role in the livelihood of the approximately 243 000 residents of the nearby villages. The main economic value of the BY Lake region derives from tourism, fishing, aquaculture, and the harvest of reed and lotus. Other benefits of the lake area include the regulation of the local climate and flood protection (Guo et al. 2011). Most of the land area surrounding BY Lake is used for agricultural purposes. As it is typical for China, most fields are small in size (less than 1 hectare), but neighbouring farmers often agree to grow the same crop, resulting in extended areas with the same type of cultivation. The main cropping system in the NCP is a winter wheat-summer maize double cropping (Fang et al., 2010). This also is also the case in the BY Lake region.

The catchment of BY Lake is a subcatchment of the Hai River basin covering an area of about 31,200 km^2. It is composed of a mountainous region in the western part (Taihang Mountains), and alluvial fans and plains in the eastern part. The plain area has an underlying unconfined shallow aquifer system with an average depth of 50 m. The water level lies only few meters below the ground surface in many areas. The area belongs to the temperate zone with semi-humid monsoon climate and a mean annual precipitation around 550 mm.

3.2 Water pollution and abstraction

Surface water qualities at BY Lake are impacted by localized human activities, but also by contaminant loads carried in by the lake's major (and only perennial) tributary, Fu River (Hu et al., 2010). Fu River springs near Baoding city, where it is fed by industrial and urban effluents (Qiu et al., 2009). It also receives high inputs of fertilizers and pesticides along its flow through the more rural areas approaching BY Lake. Fu River greatly affects the water quality of the wetland area (Qiu et al., 2009), and both Fu River and the BY Lake area are subject to seasonal eutrophication. Furthermore, anoxic events have been reported for BY Lake in the past. The water table is generally shallow, but increased withdrawals (mostly for agricultural purposes) have locally decreased both groundwater and surface water at BY Lake (Zhuang et al 2011).

3.3 Area assessment – field report

Figure 6. Typical agricultural area in the Baiyang Lake region.

3.3.1 Motivation of the field trip

From May 29-31, a field trip was undertaken to the BY Lake area. The team (Bentje Brauns, DTU Environment; Xiangmin Sun, CAS IGSNNR) was supported by a local driver. Several farmers and local inhabitants were briefly interviewed to obtain basic information at the different sites. The goal of the field trip was to obtain an impression of the BY Lake area and the different pollution sources, and to select a monitoring site for the field study on surface water-groundwater exchange of agricultural pollutants.

3.3.2 Overview of the visited area

The field trip focused on the eastern side of the BY Lake area (see Figure 5), which is known to be more severely polluted, and to have higher groundwater tables. This was important in order to ensure that the selected site has good contact between surface water and groundwater.

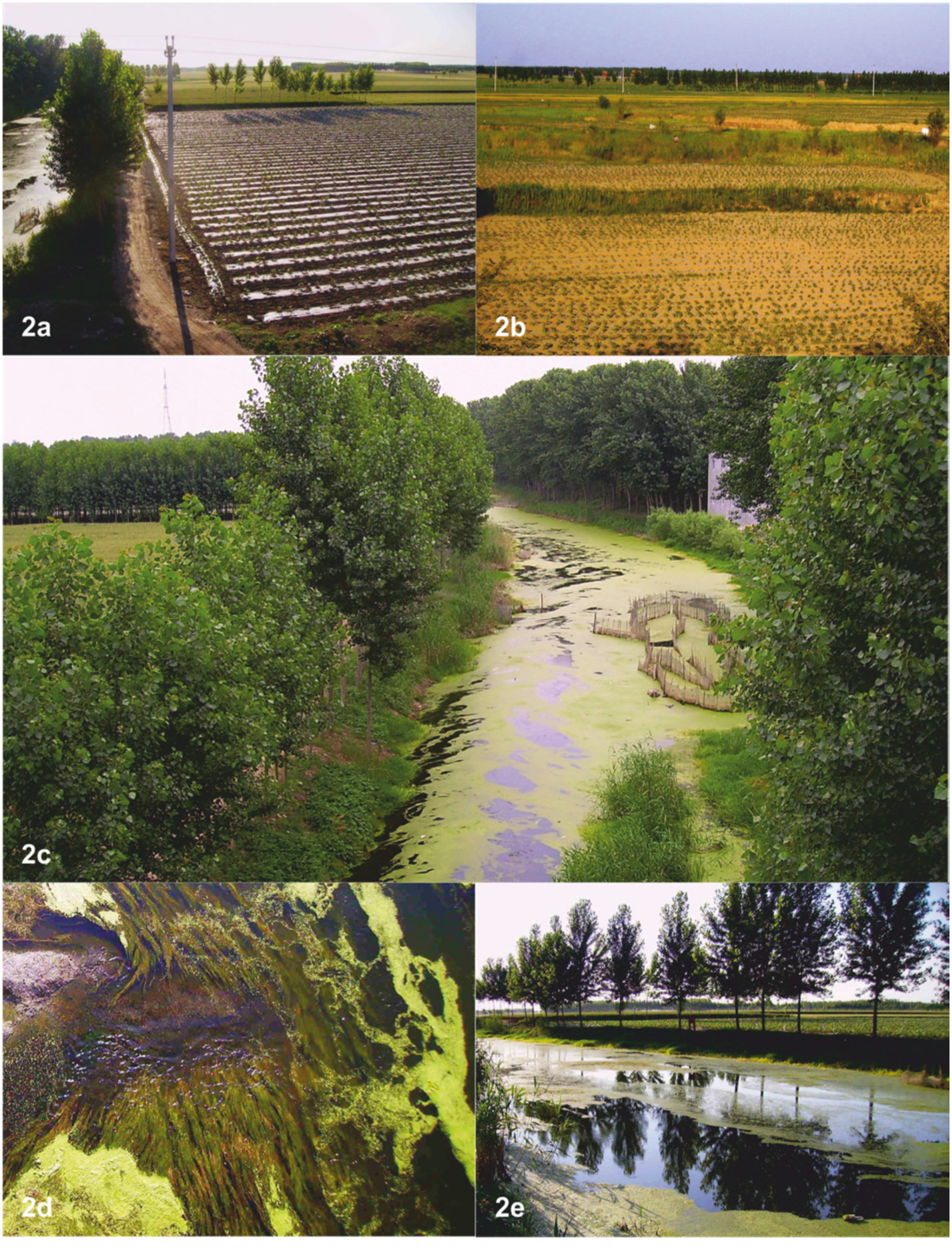
The starting point was in Anxin Town, which is one of the largest settlement areas in the lake's proximity and adjacent to Fu River.

Photos 1. Site 1 – Canal north of Anxin Town. (1a-b) Canal and adjacent field, (1c) canal at the sampled location, and (1d-e) canal further downstream, covered by a thick film of sludge, resulting from extensive pollution.

3.3.3 Site 1 – Canal north of Anxin Town

The first visited site was an open canal at the northern border of Anxin Town (see Figure 5 for the location, and Photos 1 for pictures of the site). The canal was fed from several draining ditches between agricultural fields, and additionally by inflowing sewers that carried municipal wastewater. The canal was about 20 m wide and had an estimated depth of about 5 m. The eutrophication status was visibly very severe, and the water was close to stagnant and very smelly. A surface water sample was drawn from the canal itself, and a groundwater sample was obtained from a test-drilling about 2 m next to the river. The groundwater level was reached at 3.5 m depth, which corresponds to approximately 2 m below the elevation of the surface water. Both samples (canal water and groundwater) showed a very high EC with values of 1217 and 1122 μS/cm, respectively. The high temperature (16.5°C) of the groundwater indicated that the canal water seems to recharge (and carry pollution) into the unconfined shallow aquifer at this site. The canal water had a concentration of DO of 7.3 mg/L, while the groundwater was anoxic.

No farmer could be interviewed at this site, but a passing resident indicated that the canal water was used for the irrigation of some of the adjacent fields, which did not have a groundwater well nearby. He also stated that—in response to complaints about the smell of the canal water—the local government made plans to dry up the canal, dig out some of the bottom soil, and build a concrete-lined canal.



Photos 2. Site 2 – Fu River. (2a-b) Agricultural fields adjacent to Fu River, (2c) Section of Fu River with fence from previous duck-farming and (2d-e) algal bloom at instreaming drainage ditch from agricultural fields. (All photos are from the location that was closer to Anxin Town.)

3.3.4 Site 2 – Fu River

The second visited site was Fu River, which is the only perennial instreaming river into the BY Lake Area. Fu River springs northwest of Baoding City, flows through Baoding, and then mostly passes agricultural areas until it discharges into the BY Lakes. Fu River is known to receive contamination from domestic and industrial wastewater, and is further contaminated by agricultural runoff. It is a major pollution source for the BY Lake Area.

The river was sampled at two different sites. The first location was south of Anxin Town in a rather remote agricultural section briefly before the river discharges into one of lakes. The second location was about five kilometres further to the West, near the village Dongxianyang that has about 5000 inhabitants. The reason for choosing a second location was that the river was considered as the field study site for the nutrient and pesticide study, and it was therefore better from a logistic point of view to move closer to settlement.

Again, eutrophication was a dominant characteristic, but the water was not as smelly (and also less stagnant) than the canal north of Anxin (Site 1). The elevation of the adjacent fields north of the river was barely higher than the river water level, while the southern bank generally had a steeper gradient. Water samples were taken from the river in both places and from a hand-drilled borehole 2 m from the northern river bank at the river segment near Dongxianyang (the groundwater level at 1.6m depth below ground). Field parameters were similar to the ones taken at the Site 1, with high EC (up to 1675 µS/cm), and indications from the groundwater temperature that the river water may recharge the groundwater. However, there was considerably more algal growth in Fu River than at Site 1, especially in places where drainage ditches from the adjacent agricultural field entered (see Photos 2d-e).

Photos 3. Site 3 – Small stream near the lakes. (3a) Duck-farming in the stream, (3b) aquaculture in the stream, (3c) stream and adjacent winter wheat field, (3d) fishing boats in the stream, (3e) oil pollution in the stream from motor-boats.

3.3.5 Site 3 – Small stream near the lakes

The third visited site was an unnamed stream that flows from near the mouth of Fu River towards the southern region of the BY Lakes (Figure 5). The river is extensively used for transportation by boats (Photos 3c-d), as well as for aquaculture and some duck-farming (Photos 3a and 3b, respectively). The reason for the more extensive use for aquaculture may be that the stream is very close to the scenic area of the lakes. Many inhabitants earn their livelihood with small fish restaurants, in which they cater for locals and tourists, and the stream is therefore a convenient location for farming of fish and shrimps. Additionally, small agricultural fields were adjacent to the river at places where there was no settlement directly leading up to its bank.

At first glance, the water at this site seemed less polluted than at the canal north of Anxin, or at Fu River. However, surface water samples revealed similar parameters as before, with a high EC (1313 µS/cm) that indicated pollution. Additionally, it could be observed that some of the motor boats used for transportation were leaking oil (Photos 3e), which caused pollution from petrochemicals.

No groundwater sample was taken at this site because it was found to be too busy to be considered as a study site for the field investigation on nutrients and pesticides. When asking a local resident for permission to sample his shallow well, access was denied (the reason for this was that he did not have the ownership of the well). However, the resident indicated that he believed the shallow groundwater was unsuitable for drinking (because of its smelliness, and due to local rumors that the shallow aquifer is contaminated). For this reason, he received his tap water from a deeper well. Unfortunately, he nevertheless used the water from the shallow well without any further quality assessment for the lined fish tanks, in which he breeds the fish for his restaurant. This demonstrated a lack of information within the rural population on possible reception pathways and on bioaccumulation of toxic compounds in the environment (and fish).

Photos 4. Site 4 – The Baiyang Lake recreational area. (4a-c) Healthy looking section of the Baiyang lakes, and (4d-e) severe eutrophication in smaller bends of the lakes.

3.3.6 Site 4 – The Baiyang Lake recreational area

The last visited site was part of the recreational area of the BY Lakes itself. This area is mainly used for tourism, but also for aquaculture and reed harvest. It was assumed that due to the many tourists visiting the area, it could be difficult to set up a field site too close to the lakes, but the visit to this site was nevertheless undertaken to get some brief visual impression on the impact that the inflow of polluted streams.

Overall, the impression on the water quality and eutrophication status of BY Lake was very was very mixed. At places where more dilution took place, no eutrophication of the lake water could be observed. There was an abundance of reed growth, which would be harvested in the fall by local residents (Photos 4a-c). In smaller bends of the lake—where water was more stagnant—high growth of algae and duckweed was observed (Photos 4d-e). In addition to eutrophication, pollution from partially very old motorboats that take tourists out onto the lake area is very likely, but could not be observed on a large scale. Fish and wildlife (e.g. birds and many insects) were spotted in several locations.

Nevertheless, water samples that were drawn from the lake in three different places during a boat ride revealed that EC values ranged between 998 and 1217 μS/cm (no test-drilling was done in this area because no suitable location for the planned field work could be found). This was only slightly lower than the observed values at the other sites. The indication of this was that even though some dilution and mixing takes place, the lake area is highly impacted by inflowing rivers.

Photos 5. Other sites – (5a-c, 5e) Waste deposition into ditches and its effect on water quality; (5d) outlet of a wastewater pipe into a river, discharging untreated wastewater.

3.3.7 Overall impressions and site selection

The general impression of the overall area was that most water bodies seemed to be highly impacted by a variety of human activities:

- Agricultural runoff of nutrients (obviously leading to eutrophication in some places), and potentially also pesticides.
- Excrements from duck farming.
- Petrochemicals from leaking boats.
- Waste deposits of household waste, mostly into ditches (Photos 5a-c, 5e).
- Direct discharge of untreated wastewater into water bodies (Photos 5d).

A striking observation was that the behavior of the local population towards the environment seemed often ambiguous: Some local residents were disturbed by the smell of polluted water body (Site 1), but others (or seemingly a majority of them) would discard their household waste into ditches, or even directly into water ways. Also, as in the case of the fish farmer/restaurant that was interviewed at Site 3, there seemed to be some awareness that not all the available groundwater within the region was suitable for drinking and household purposes. Nevertheless, the water was still used for other purposes (in this case for fish tanks and irrigation of the adjacent garden) without further consideration of potential impacts from this, and also without undertaking efforts to obtain a detailed water quality analysis.

Based on the area assessment, it was decided to choose Fu River as the site for the field study on nutrients and pesticides. In addition to the reasons introduced in the beginning of section 3, this decision was taken because of:

- Perennial flow: At some of the other sites, it was not clear if there would be perennial flow throughout the entire study period.
- Previous studies: Because Fu River has been a pollution source throughout the past decade; the river mouth has usually been included in studies on BY Lake. Therefore, some previous data on water quality could be collected (and later on compared to the own findings).
- Accessibility: Easy access to the site by vehicle was a premise for the study, so that the installation equipment, heavy batteries, etc. did not have to be carried too far. At the same time though, it was important that the installed wells would be safe from too much commotion.

4 The Baiyang Lake nutrient and pesticide study

As discussed in section 3.3.7, Fu River (Site 2 from the field excursion) was chosen as the study site for the nutrient and pesticide study. The exact location is on the northern bank of Fu River near Dongxiangyang Village, which is located few kilometres southwest of the town Anxin, Hebei Province (see Site 2 on Figure 5, and Figure 7). The village itself extended from the south up to the banks of Fu River and had an approximate population of about 5000 inhabitants. This was very convenient, because it implied to have easy access to equipment, and very little traffic at the field site itself (the village does not extend north of the Fu River). More detailed information on the setting of the field site can be found in **I**.

Figure 7. Picture of Fu River at the field site location during the installation of the wells for sampling the hyporheic zone in March 2012.

4.1 Agricultural activities

The fields at the BY study site extend very close to the adjacent Fu River. Distances between the agricultural area and the river bank are only 2 to 5 meters, and aside from sparse tree lines, no hedges or similar boundaries are present that would protect the surface water from runoff or wind drift that may contain agricultural pollutants. Almost all of the annual rainfall occurs in the summer, especially from June to September (80%). Throughout the remaining time of the year, only very few rain events take place. This leads to a need for irrigation especially from April to June when temperatures are rising and the winter wheat starts growing, but the weather is dry. Accordingly, irrigation of the winter wheat took place at the field site once in April and twice in May 2013 (see **I** for more detailed information).

The field site is agriculturally used without lag periods for a winter wheat–summer maize rotation. The neighboring fields are used for the same cropping system, and the farmers follow the same farming schedule and share equipment. Based on interviews, it seems however, that the farmers often take different decisions regarding fertilizer and pesticide applications. Detailed information on fertilizers and on pesticides used on the studied field can be found in **I** and **II**, respectively.

4.2 Pollution sources

Fu River has been classified by the Chinese Ministry of Environment as a river with water quality inferior to grade V (MEP, 2013), which means that it should not be brought into direct contact to humans, and is unsuitable for any purpose, including agricultural irrigation, fishing, duck farming, etc. In fact however, the river was widely used for irrigation, duck farming and fishing by the local population.

Pollution sources for Fu River are instreaming domestic and industrial wastewaters (especially in Baoding City), and agricultural runoff. At the field site itself the main pollution sources were:

- Agricultural inputs, such as fertilizers and pesticides. In addition to the direct application, "empty" containers were often left in the field directly, or discarded into the river (Figure 8a).
- Excrements from nearby duck farming (which took place regardless of the rivers low water quality) (Figure 8b).
- Domestic waste that was discarded directly into Fu River and into drainage ditches adjacent to the field site (Figure 8c).

Figure 8. Different pollution sources at the field site. (a) Runoff of fertilizers (and pesticides), (b) duck farming, (c) household waste that is discarded into the river and accumulates in areas with low flow.

4.3 Water flow

Water levels in the BY Lake catchment have been declining with increasing rates up to 1 m/yr from 1974-2007 due to over-exploitation of groundwater, and an increase in drought years (Lü et al., 2010). However, the study site had a very shallow groundwater level just few meters below ground. A comparison of the water level from pre-site assessment in May 2012 and the installation period in October 2012-March 2013 indicated an annual fluctuation of the groundwater level of about 1m. Due to the high water table and the perennial flow of Fu River, groundwater and surface water were assumed to be well connected during all times of the year at this study site.

4.4 Data collection

The aim of the water sampling was to monitor nitrogen species in river water, groundwater and soil water over the course of one year (with sampling intervals of approximately six weeks), and to take samples for selected pesticides during four of the sampling times (May, July and October 2012, and March 2013) from river water and groundwater. The dataset was supplemented by ion analysis and measurements of stable isotopes (hydrogen and oxygen). In order to do this, two monitoring lines were installed at the field site (Figure 9) that were comprised of groundwater well (screened at

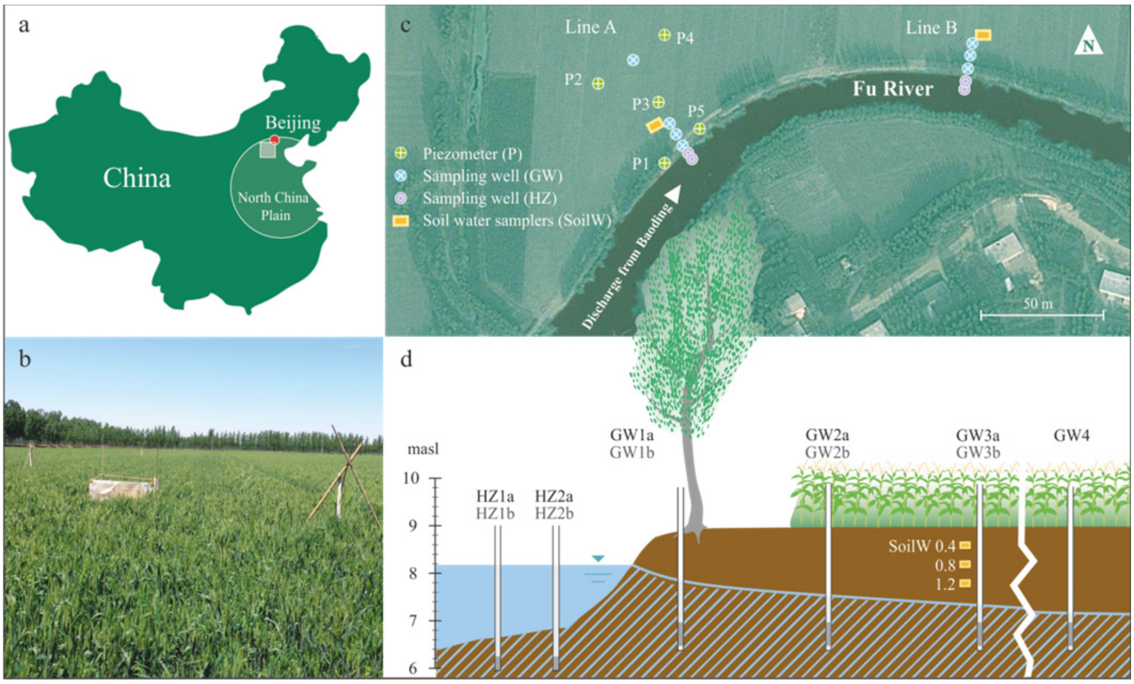


Figure 9. (a) Location of the study site; (b) installed sampling wells and the box for the soil water samplers; (c) aerial view of the setup of the monitoring network; (d) cross-section of monitoring network (modified from **I**).

a depth of approximately 2.5m below ground), four wells in for sampling of the hyporheic zone, and soil water samplers. For soil water sampling, PTFE/quartz suction cups (Prenart Super Quartz Mini, Prenart Equipment ApS, Denmark) were placed at 0.4, 0.8, and 1.2 m depth at GW3a and GW3b. With this, nutrient leaching from the field surface should be measured. For ion analysis and isotopic measurements all water samples were collected using a low-flow peristaltic pump (~200mL/min), to which a flow cell was attached. The sampled water was filtered immediately through a 0.45 μm cellulose-ester membrane into three 20 ml polyethylene bottles. This were filled to overflowing, capped, and immediately cooled to 4°C and transported to the Institute of Geographic Sciences and Natural Resources Research for analysis (transport took place on the same, and analysis on the following day). Additionally, the samples for cation analysis were acidified immediately (pH=2) using concentrated HNO_3. All samples were transported and stored at 4°C. For pesticide analysis, 1 L samples were filled to the rim into brown glass bottles. They were immediately cooled to 4°C, and then shipped by courier to the laboratory (Eurofins, Glostrup) in Denmark because no suitable location for desired range of analytes was found in China. For more details on the sampling, and on limitations in soil water sampling that occurred after heavy rainfalls in the late summer 2013, refer to **I** and **II**.

In addition to the water analysis, hydraulic heads of river water and groundwater were monitored via automatic manometers (Mini and Micro Diver, Schlumberger, Netherlands), so that detailed information about the flow patterns at the site could be obtained. The manometers were installed in five piezometers (P1-P5, see Figure 9) on the agricultural field and in one location in the river. The automatic monitoring was complemented by additional manual hydraulic head measurements which were taken at each sampling campaign from the piezometers, as well as from the sampling wells. An application of 6 moles of LiBr (dissolved in 10 L of water) was made near M3a and M3b in April 2013 to obtain an estimate on vertical and horizontal subsurface water velocities. For determination of particle size distribution, soil samples over a depth from 0–260 cm were taken to measure total carbon (TCs), total organic carbon (TOCs), and total inorganic carbon (TICs) of the sediment.

4.5 Data analysis

Water samples were measured in situ via electrodes in a flow cell for T, DO, pH, and EC with a field meter (Multi 3430, WTW, Germany). Alkalinity was determined via immediate titration in the field. NH_4-N was analyzed using a discrete analyzer (Smartchem 300, AMS Alliance, Italy), and for all other cation analysis, an ICP-OES (Optima 5300DV, PerkinElmer, USA) was used. Anions were analyzed via an integrated IC System (ICS-2100, Thermo Scientific™, USA) and δD and δ18O via an Isotope Ratio Mass Spectrometer (MAT 253, Thermo Scientific™, USA). The charge balance error for each of the 114 water samples was calculated and lay within the permissible limit of ±5% (**I**). Pesticide samples were analyzed by the company Eurofins (Glostrup, Denmark) for a standard package of different pesticides, which included some of the most broadly applied herbicides in the NCP (and at the BY Lake study site). Analysis was done via gas chromatography mass spectrometry (GC/MS) for chlorphenols and dichlobenil, and via liquid chromatography tandem mass spectrometry (LC-MS/MS) for all other pesticides. The detection limit was 0.01 µg/L for all compounds (**II**).

Soil texture was analyzed using a particle-size analyzer (Analysette 22, Fritsch GmbH, Germany). TCs was measured via elemental carbon analyzer (CS 200, LECO Corporation, USA), and the TOCs was determined via the Walkley-Black method (Lettens et al., 2007). TICs was then calculated by subtracting TOCs from TCs. Further geochemical analysis of the water samples, saturation indices (SI) for different mineral phases and partial pressure of carbon dioxide (P_{CO_2}) was done using the geochemical speciation and modelling code PHREEQC Version 3 (Parkhurst and Appelo, 2013). The PHREEQC code was also used to simulate hydrogeochemical effects of the river water flowing into the aquifer in a 1-D horizontal transport. The model was set up in order to identify dominant removal processes for nitrogen. More information on the setup of the PHREEQC model is given in publication **I**.

5 Results and discussion

In this chapter, the main results and implications of the BY nutrient and pesticide study are briefly outlined and discussed. The results of the pesticide study include the outcome of the literature review on pesticide use and research in the NCP. Following this, there will be a discussion of additional observations and insights obtained during the PhD work in China, namely the lack of farmer education (which is also discussed in detail in **III**), challenges in literature research, and limitations in access to data. More detailed information on the different studies, their outcomes, and on the 1-D model simulation with PHREEQC can be found in **I** (nutrient study and PHREEQC simulation, and results on grain size analysis, bromide tracer application, and stable isotope measurements), and **II** (pesticide study), and **III** (recommendations on increased farmer training to reduce excessive inputs of agrochemicals in China).

5.1 Nutrient exchange and removal

The BY Lake nutrient study (**I**) showed that the groundwater (as well as the soil system) at the studied field site is under strong influence of the instreaming river water. Even though sufficient oxygen was available, the river system could not nitrify its ammonium load of up to 29.8 mg/L NH_4-N. Ammonium was therefore transported into the shallow groundwater. Peak concentrations of 134.8 mg/L NO_3^-N were measured in the soil water. This implied a substantial amount of nitrate leaching from the surface of the agricultural field into the aquifer.

Both nitrogen species were only detected in low concentration in the shallow groundwater (averaging at 3.6 mg/L NH_4-N and 1.8 mg/L NO_3-N) despite the high inputs from the river water and from the field surface. Measurement results supported by PHREEQC-modelling indicated cation exchange (the soil was very clayey, and therefore had a high cation exchange capacity), denitrification, and anaerobic ammonium oxidation (anammox), coupled with partial denitrification, as major nitrogen removal pathways. It is highly recommended to improve the water quality of Fu River. The use of the currently polluted for irrigation is not recommended due to its high pollution. Monitoring of nitrate in the aquifer is important because a difference in surface water quality could the conditions for the current degradation processes (especially for anammox), potentially leading to increased nitrate concentrations in the aquifer (**I**).

5.2 Pesticide use in China

The findings of the literature study on pesticides in China show that the three most widely used herbicides in the NCP are 2,4-D, acetochlor, and atrazine. All of these compounds are suspected to have potential negative impacts on the environment, and two of them (acetochlor and atrazine) have recently been banned for use in Europe. Research studies in the NCP are focused very much on pollution by the legacy pesticides DDT and HCH. These had been phased out in China in the 1980s, but are still present in many waterways.

In the BY field study, 2,4-D, atrazine, and atrazine metabolites could be detected in river water and groundwater throughout the year, with peak values of 3.00 µg/L, 0.96 µg/L, and 9.84 µg/L, respectively, which occurred in May 2013. This indicated that monitoring of these compounds might be important in Chinese surface waters and groundwaters. Nevertheless, results from literature review showed that research studies in the NCP are focused very much on pollution by the legacy pesticides DDT and HCH, which were phased out in China in the 1980s, and biased towards surface water and soil sampling. Even though it is certainly important to monitor legacy pesticides, a systematic monitoring of currently-used pesticides should also be encouraged. The monitoring program should include both surface water and groundwater, and it should be able to catch temporal variations (**II**).

5.3 Lack of farmer education

Overuse and improper handling of pesticides have been observed in China for a long time, and studies have been pointing out since the 1980s that better management needs to be implemented. This has been confirmed by observations and own results during this PhD study. Farmer education is still poor in many areas, and infrastructure for proper pesticide disposal is missing (also discussed in **II**). Accordingly, empty pesticide containers were found at several locations (on the field itself in the surface water) at the field site.

Better farmer education, enhanced quality control, and a sufficient disposal system for pesticides are highly recommended to avoid occupational risks, pesticide overuse, and environmental pollution. Furthermore, enhancing the training of agricultural workers could potentially increase environmental awareness and critical thinking and reflections about currently unsustainable farming practices. One way of doing this could be to increase the investment in alternative forms of education, such as vocational farmer training (discussed in depth in **III**).

5.4 Challenges on data acquisition

Obtaining sufficient and appropriate data for China can be a challenge for several reasons:

- Language challenge. More than in some other countries, language can be a barrier in data acquisition for non-native speakers. Even though many governmental webpages are also available in English, the amount of available information often differs, and in some cases, the needed information is only available on the Chinese website. A basic knowledge of the language is therefore very useful to be able to scan websites, or also data tables in papers that are only published in Chinese.
- Use of different units. Even in English documents, Chinese data is often represented in Chinese units, such as "mu (亩)" as area, and—more often—"wan (万)"as a quantifier of "10 000". Therefore, much of the Chinese data needs to be re-calculated, and a critical look at data from non-Asian authors is often needed, as it is an easy mistake to confuse "wan" with the internationally used quantifier 1000.
- Difference in definitions. Whereas some non-Asian publications (e.g., FAO) use "grain" referring to "cereal grains" only, it is commonly used in China (e.g., the National Bureau of Statistics) as a more broad term for cereals, legumes, and tuber crops (Li et al., 2014).
- Insufficient accuracy of data. In some cases, no accurate data is available due to systematic problems. One example of this is the exact number of full-time farmers. In Chinese statistics, household registrations are based on the *hukou* system. The *hukou*, which is determined on hereditary basis and not on the actual place where a person currently lives, is divided in rural and urban *hukous*. Therefore, a person from a rural background normally maintains the *hukou* from the village he or she comes from. This means, for example, that many construction workers who work in different cities most of the year, (but occasionally farm their land in their hometown) will statistically be considered the same as full-time farmers (Yang, 2013). As it is often difficult to record the exact number of rural migrant workers, also the data on the pure farming population might be skewed.

5.5 Challenges in chemical analysis

Due high governmental funding of research, many Chinese universities and research institutions are extremely well equipped. Well-trained staff usually runs specific pieces of equipment. However, there are some challenges to the foreign researcher. One difficulty is that laboratory procedures are often based on Chinese standards, which are tedious to obtain, and often not translated into English. Another factor is that some compounds are not as frequently researched as others, and therefore it might be difficult to find a place for analysis. During this PhD study, for example, the samples for pesticide analysis were shipped to Denmark. This was done for two reasons:

- Even though the research network between universities was well connected, no institute could be found that would analyse for pesticides other HCHs and DDTs. (This underlines the heavy research focus on these compounds discussed in section 4.2.1.).
- Commercial laboratories have to follow governmental guidelines of data disclosure. In case of atrazine, for example, the exact concentration could be given if it exceeded the allowed drinking water level of 0.2 µg/L. However, the laboratory was not allowed to disclose the exact concentration if it fell below the threshold value.

6 Conclusions

The goal of this PhD study was to improve the understanding of transport and fate of agricultural nutrients (with a focus on nitrogen) between a surface water-soil-groundwater system at the BY Lake study site in the NCP; to review pesticide use, research and monitoring in China; to assess occurrence and exchange of pesticides at the BY Lake study site; and—based on the field observations and the research results—to identify and discuss gaps and shortcomings in Chinese agriculture practice.

Based on the results of the one-year field study at BY Lake, and supported by extensive literature review, the following conclusions can be drawn:

- The investigated surface water-soil-groundwater system at the BY Lake study site showed a good removal capacity for nitrogen. However, if—in addition to denitrification of nitrate and cation exchange of ammonium—anammox is the dominant removal process (as indicated by PHREEQC modelling), then the conditions for degradation might change if surface water changes (for example by decreasing the amount of ammonium). Surface water quality improvements are therefore recommended, but in conjunction with simultaneous monitoring of nitrate in the aquifer. Additionally, reduced agricultural N-inputs should be considered.
- Results from the literature research indicated that sampling was biased towards surface water monitoring. Furthermore, most studies focused on legacy pesticides like DDTs and HCHs. The three most commonly applied herbicides in the NCP (2,4-D, atrazine, and acetochlor) were only addressed in few studies, and investigation of groundwater was seldom. A more balanced research of both, legacy and currently-applied pesticides, and increased monitoring of groundwater (especially in shallow aquifers) is therefore recommended.
- Pesticide analysis of water samples from the BY Lake study site showed that current herbicide application may lead to surface water and groundwater contamination. 2,4-D and atrazine were the pesticides with the highest concentrations, ranging up to 3.00 and 0.96 µg/L, respectively. Peak values occurred after the spring application of the pesticides on the winter wheat. Surface water had higher concentrations than groundwater. In conclusion, the monitoring in groundwater and surface water of currently-used pesticides should be given more importance.

- The observed use of agrochemicals at BY Lake, and numerous research studies indicate that fertilizers are often used in excess, and that current knowledge on the active ingredients in pesticides and their potential environmental harm (or occupational risk factor) is not well understood. Better farmer education, enhanced quality control, and an effective disposal system for pesticides are highly recommended to avoid occupational risks, pesticide overuse, and environmental pollution.

7 Perspectives

China has been doing extremely well over the last three decades in maintaining food self-sufficiency for staple foods (such as grain) on a national scale. However, the high toll on the environment that is caused by the impact of agricultural pollutants needs to be more efficiently counteracted in the future to prevent further deterioration. In this PhD study, the following perspectives for future research have been identified:

- Increasing the process understanding of soil-groundwater-surface water interactions, especially the transport and fate of pollutants. This would be valuable information to understand how water qualities in one compartment are affected by changes in the other, and to assess vulnerabilities. At the studied field site near BY Lake, it would be interesting to further test for the hypothesized removal processes of nitrogen. For example, isotope pairing technique with N-15 isotopes could be used to determine degradation pathways in more detail.
- Delineation of areas, in which surface water is infiltrating into groundwater. Because many surface waters are currently heavily polluted, it is important to identify groundwaters that might be impacted by pollutant transport from surface waters.
- Development of improved monitoring strategies, especially for pesticides. Even though improvements in water quality monitoring of nitrogen species, such as ammonia, have been made in China, no systematic approach could be identified to determine the occurrence of pesticides on regional or national scales. Collecting and disseminating information on pesticide occurrence, and including such test results from the main river basins in the “Annual Report on the State of the Environment” (published by the MEP), could guide researchers in their study focus. Furthermore, it could increase public and farmer’s awareness of pesticides that accumulate, and cause problems in the environment.
- Evaluation of different approaches to knowledge dissemination from research results to field application. A more efficient and sustainable way of knowledge transfer to farmers is urgently required.
- Identification of incentives for the younger generation to choose farming as a profession. Full-time agricultural workers are more likely to have the required knowledge, motivation and time to farm in most optimized ways; therefore it would be good to promote professional farming.

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9 Papers

I Brauns, B., Bjerg, P.L., Song, X., Jakobsen, R. 2015. Field scale interaction and nutrient exchange between surface water and shallow groundwater in the Baiyang Lake region, North China Plain. In press.

II Brauns, B., Jakobsen, R., Song, X., Bjerg, P.L. 2015. Pesticide use in the wheat-maize double cropping systems of the North China Plain: Assessment, field study, and implications. Submitted.

III Brauns, B., Song, X., Jakobsen, R., Bjerg, P.L. 2015. Vocational training for farmers: A long-term approach to the alleviation of environmental pollution from agriculture in China. Submitted.

In this online version of the thesis, **paper I-III** are not included but can be obtained from electronic article databases e.g. via www.orbit.dtu.dk or on request from.

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