# Managing nitrogen, water and carbon flows and their interactions for circularity in agrifood systems

— a case study in Quzhou, China



Zhibiao Wei

### Propositions

1. Agrifood systems are net carbon sources.  (this thesis)
2. Full circularity through full self-sufficiency harms sustainability. (this thesis)
3. In water-abundant regions more trees should be planted to mitigate global water scarcity.
4. The expansion of paddy areas aggravates global warming.
5. Trade is not a threat but a solution to climate change.
6. A flexitarian diet is the best dietary choice.
7. Deadlines contribute to a meaningful life.
Propositions belonging to the thesis, entitled
Managing nitrogen, water and carbon flows and their interactions for circularity in agrifood systems - a case study in Quzhou, China
Zhibiao Wei
Wageningen, 15 May, 2024

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This research was conducted under the auspices of the C.T. de Wit Graduate School for Production Ecology and Resource Conservation (PE&RC)

# Managing nitrogen, water and carbon flows and their interactions for circularity in agrifood systems

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#### Zhibiao Wei

#### **Thesis**

submitted in fulfilment of the requirements for the degree of doctor at

Wageningen University

by the authority of the Rector Magnificus,

Prof. Dr C. Kroeze,

in the presence of the

Thesis Committee appointed by the Academic Board

to be defended in public

on Wednesday 15 May 2024

at 1.30 p.m. in the Omnia Auditorium

# Zhibiao Wei Managing nitrogen, water and carbon flows and their interactions for circularity in agrifood systems - a case study in Quzhou, China 162 pages

PhD thesis, Wageningen University, Wageningen, the Netherlands (2024)

With references, with summary in English

DOI: https://doi.org/10.18174/647978

ISBN: 978-94-6469-888-6

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### Chapter I

### General introduction

Zhibiao Wei

## I.I. Agrifood systems and their environmental and social impacts

Agrifood systems comprise the entire range of actors and interlinked activities that add value in agricultural production and related off-farm activities such as food storage, processing, distribution and consumption (FAO, 2023). They are responsible for providing sufficient nutritious food for a growing and wealthier global population (FAO, 2022; OECD and FAO, 2022). Agrifood systems account for more than a quarter of global total employment, which is crucial for social stability. Meanwhile, linear and inefficient agrifood systems have caused substantial losses of nitrogen (N), water and carbon (C) from the system, inducing severe environmental problems (Springmann et al., 2018; Wang et al., 2022a). They emit about 30% of anthropogenic greenhouse gases (GHGs), create about 32% of acid emissions, are responsible for about 80% of freshwater and marine eutrophication, and account for about 70% of the global freshwater withdrawals (Poore and Nemecek, 2018; FAO, 2021). Reconciling food security with the environment and social stability necessitates transitioning to circular, environmentally sustainable agrifood systems (Ambikapathi et al., 2022; FAO, 2022; Wang et al., 2022a).

Such a transition requires redirecting material and nutrient flows in agrifood systems, encompassing the compartments of crop production, livestock production, food processing, household consumption and waste management (Ma et al., 2010; Wolf et al., 2015; Bai et al., 2016; Zhang et al., 2020). However, our understanding of the system is hindered by the lack of insight into the direct and indirect linkages between different compartments. Crops feed livestock and humans while livestock and human excreta could be used to fertilize crops, but the latter does not always happen (Halpern et al., 2019). Considerable amounts of manure and human waste are lost from the agrifood system (Berendes et al., 2018; Harder et al., 2020). Most research has been focused on only one compartment of the agrifood system (Liu et al., 2013; Bai et al., 2016; Bai et al., 2018; Cui et al., 2018). The impact of a single modification within one compartment on the overall system and on another compartment is often ignored. Supporting a sustainable circular transition necessitates an integrated system-level analysis of material and nutrient flows and their interactions in the agrifood system, such as the loops of N, water and C and their interactions.

# 1.2. Nitrogen, carbon and water cycles and their interactions in agrifood systems

#### I.2.I. N cycles in the agrifood system

Anthropogenic N input plays a key role in sustaining the current agrifood system. More than half of the population today is nourished by crops produced with chemical N fertilizer (Zhang et al., 2015). The nitrogen use efficiency (NUE) is approximately 20% worldwide (Lassaletta et al., 2016), and most reactive N is emitted to the atmosphere and water bodies. The emitted reactive N into the air is a major precursor in forming fine particulate matter with an aerodynamic diameter less than 2.5 µm (PM2.5), posing a threat to human health (Domingo et al., 2021; Xu et al., 2022). The nitrate discharged into water causes eutrophication and biodiversity losses (Sutton et al., 2011; Gu et al., 2015). Nitrogen losses from agrifood systems can be quantified using loss ratios (LRs) which are defined as the percentage of N input that is lost via a specific pathway. The LRs are regional specific (Liu et al., 2017; Van Damme et al., 2018; Chaudhary and Krishna, 2019), and the LRs of nitrous oxide (N<sub>2</sub>O-N) and ammonia (NH<sub>3</sub>-N) have been widely quantified across variable crop types (Gaihre et al., 2019; Li et al., 2022), field management (Shcherbak et al., 2014; Gerber et al., 2016; Davis et al., 2019; Fan et al., 2022), soil and climate conditions (Duan et al., 2021; Harris et al., 2022; Li et al., 2022). In contrast, the LRs of N leaching and runoff are rarely documented, particularly regarding their reactions to field management conditions (e.g., fertilizer types), despite studies suggesting organic substitution for chemical fertilizer to alleviate N losses (Xia et al., 2017; Zhang et al., 2019).

With site- and management-specific LRs of various N losses, it would be possible to quantify N losses from agrifood systems both in current systems and in scenarios exploring measures towards N circularity based on N flows in the system. Current studies on N flows and losses focus primarily on crop and livestock production (Renard and Tilman, 2019; Xu et al., 2021) but to a limited extent on households or entire agrifood systems. Some models, such as the NUFER (Nutrient Flows in Food Chain, Environment and Resource use) and CHANS (Coupled Human And Natural Systems) model (Ma et al., 2010; Gu et al., 2015; Zhao et al., 2017), have been developed and used to quantify N flows in the food system. These models could help explore measures and the potential to reduce N losses and facilitate the transition towards N circularity in advanced management scenarios. However, due to lack of validations at the household level, the data they provide on household consumption is unreliable. Therefore, it is difficult to directly apply existing models to reconstruct N flows for variable households and guide N circularity management.

Nitrogen circularity could be promoted by the concept of circular agriculture. Circular agriculture was proposed to connect agricultural production with sustainable resource use, aiming to minimize pollution, optimize production and maximize resource reuse (Kirchherr et al., 2017; Velasco-Muñoz et al., 2021). The principles of circular N use in the food system include reducing external inputs, reusing waste streams, and reducing environmental losses (Muscat et al., 2021; Velasco-Muñoz et al., 2021). Restoring N circularity requires selecting the most relevant spatial scale to enable nutrient transport and thereby facilitate N exchange.

### I.2.2. Water balances and their interactions with N cycles in the agrifood system

Water is crucial for human life, food production and ecosystem services (Scanlon et al., 2023). It is projected that by 2050, global water consumption will be 20–30% higher than the current level (WWAP, 2019). Climate change also contributes to an increasing frequency of extreme weather events, such as droughts and floods (Pokhrel et al., 2021). These events worsen spatial and temporal mismatches between water resources supply and demand, presenting greater challenges for many societies in securing reliable water supplies (Scanlon et al., 2023). For example, over-exploitation of groundwater in the North China Plain has led to a decline in the groundwater table level at a rate of 0.6 meters per year (Qiu et al., 2018), which has caused phenomena such as drying streamflows, ground fissures and land subsidence (de Graaf et al., 2019; Luo et al., 2022). In addition, drainage water with high nutrient concentrations is discharged to rivers and groundwater, leading to water pollution (Grafton et al., 2018; Berbel et al., 2019). Poor water quality and low available quantities are both urgent problems to be solved.

Agrifood systems are key in ensuring global water security for both quantity and quality. They account for about 70% of the global freshwater withdrawals and 90% of the water consumption (Hoekstra and Mekonnen, 2012; FAO, 2021), mainly by means of irrigation (McDermid et al., 2023). They are also the primary contributors to water pollution and deterioration (FAO, 2021). Water-saving irrigation was claimed to save water and address water competition among agriculture, industry, and the energy sector (Zhao et al., 2015; Flörke et al., 2018; Chen et al., 2020). For instance, regulated deficit irrigation and drip irrigation were proved to be effectively improve water use efficiency at the field level (Kang et al., 2017; Grafton et al., 2018). However, the "savings" achieved through efficiency gains at field level may overstate actual impacts on water supply, because water that is not consumed at one site is often discharged into surface water or recharges groundwater, which can be later recovered and used downstream (Aeschbach-Hertig and Gleeson, 2012; Grafton et al., 2018; FAO, 2021). Thus, a

comprehensive accounting of water flows is necessary to ascertain whether gains in water use efficiency at the field level effectively conserve water on broader scales.

Water cycles are closely connected with N cycles. Water use efficiency in crop production tends to be low when NUE is high (Sadras and Rodriguez, 2010). Moreover, N discharge contributes to water pollution (Cao et al., 2019). The European Union and United States experience degraded water quality primarily due to agricultural runoff. Approximately half of their surface waters are rated poor due to high concentrations of N and in some cases phosphorus (P) (Ibáñez and Peñuelas, 2019; Scanlon *et al.*, 2023). In China, anthropogenic N discharge into freshwater averaged around 14.5 Tg per year between 2010 and 2014. This is about 2.7 times the estimated "safe" discharge level (Yu et al., 2019). Worldwide, some 1.8 billion people must rely on drinking water that is contaminated with faecal matter, which puts them at risk of cholera and other diseases (Scanlon et al., 2023). When water quality is factored into water scarcity assessments, the proportion of the global population experiencing severe water scarcity increases from 30% to 40% (van Vliet et al., 2021). However, few studies have integrated the water and N cycles to explore the potential impact of current water use and pollution on future water security.

### I.2.3. C cycles and their interactions with N cycles in the agrifood system

The UN Climate Change Conference reached an agreement to limit global warming by the end of this century to well below 2 °C, aiming ideally for below 1.5 °C compared to pre-industrial levels (UNDP, 2015). Although over 80 countries, representing approximately 70% of global greenhouse gas (GHG) emissions, have pledged to achieve net-zero emissions mostly by the current mid-century (Matthews and Wynes, 2022; Zhao et al., 2022), the current efforts are insufficient to achieve the climatic goal. The options to achieve net-zero CO<sub>2</sub> (carbon neutrality) or even GHG (climate neutrality) emissions are still unclear.

Agrifood systems are key to achieving the UN climatic goal (Crippa et al., 2021; FAO, 2022). They emit a third of global anthropogenic greenhouse gases and induce about 22% of global mortality due to air pollution (Poore and Nemecek, 2018; Crippa et al., 2021; Crippa et al., 2022). Understanding the net effect of agrifood systems on climate requires quantifying C fluxes and their CO<sub>2</sub> equivalent (Wolf et al., 2015), including both C flows and storage in different compartments of the entire agrifood system (Wolf et al., 2015). The C storage in agrifood systems mainly exists in soils which comprise an important C pool on earth (Sykes et al., 2019). A large fraction of agricultural lands has been degraded by soil organic matter loss (Smith et al., 2008). Considerable amounts of C are harvested as grain from agricultural land and consumed by livestock and humans,

but little C from animal and human waste is recycled for crop production (Gu et al., 2015; Lassaletta et al., 2016; Billen et al., 2021).

Enhancing soil C sequestration is often listed as a potential solution not only to combat climate change (Sykes et al., 2019; Walker et al., 2022), but also to regenerate degraded soils (Kopittke et al., 2022). To regenerate degraded soils, adequate amounts of organic carbon should be applied to increase the content of soil organic carbon (SOC) to 2% or higher (Maillard and Angers, 2014; Oldfield et al., 2019). The increasing SOC both enhances crop production (Oldfield et al., 2019) and improves soil resilience and health (Domingo-Olivé et al., 2016; Oldfield et al., 2019). However, soil C storage and changes pose challenges for direct monitoring in fields due to the difficulty in sampling and quantifying belowground plant biomass and rhizodeposition (Hirte et al., 2018; Hu et al., 2018). In addition, the alteration of soil carbon storage is a gradual process necessitating long-term observations. The annual change rate is contingent upon the initial soil C content (Lugato et al., 2014; Moinet et al., 2023). Modelling serves as a readily available option to forecast soil C storage and changes, but accurate predictions require rigorous validations, while more than 70% of the models simulating soil carbon storage lack proper validation (Garsia et al., 2023). Furthermore, C sequestration in soils originating from atmospheric CO<sub>2</sub> should be considered in light of CO<sub>2</sub>-C emissions caused by the energy and power consumption required for system functioning (Lorenz and Lal, 2018; Adetona and Layzell, 2019). These emissions may counteract or even outweigh CO<sub>2</sub>-C sequestration in soils and agrifood systems.

Apart from CO<sub>2</sub>, non-CO<sub>2</sub> GHG emissions contribute to a quarter of climate change, including CH<sub>4</sub> and N<sub>2</sub>O, which process higher global warming potential (GWP) than CO<sub>2</sub> (Feral, 2015; Smith and Gasser, 2022; Xing et al., 2022). Therefore, mitigating climate change should consider both C and N cycles and their interactions. The C storage of terrestrial ecosystems is significantly influenced by N availability (Kicklighter et al., 2019; Raza et al., 2020). Enhanced N availability through atmospheric deposition promotes the net primary productivity of ecosystems and increases the biological sequestration of C (Kicklighter et al., 2019). However, N-induced soil acidification, which is neutralized by soil inorganic carbon (SIC), could also cause dramatic loss of SIC in cropland (Raza et al., 2020). In addition, it may reduce crop productivity and thus decrease the biological C sequestration. Circular N use with optimal N inputs in the agrifood system could directly reduce N<sub>2</sub>O emission and mitigate climate change. It also indirectly reduces N<sub>2</sub>O emissions by decreasing NH<sub>3</sub> and leached NO<sub>3</sub><sup>-</sup> losses (Paustian et al., 2006). Nitrogen circularity and C neutrality could be mutually reinforced. Promoting synergies and minimizing trade-offs between them may help transition to C neutrality or even climate neutrality in the agrifood system.

# 1.3. Options towards sustainable circular agrifood systems and the optimal scale

Promoting circularity necessitates good management practices for both production and consumption (Gevik et al., 2023). Good crop production practices could reduce fertilizer inputs, improve nutrient use efficiency, mitigate nutrient losses, increase crop biomass and thus C inputs to cropland. Such practices include optimal fertilization (Cui et al., 2018; Horton et al., 2021), organic substitution of chemical fertilizer (Zhang et al., 2019; Young et al., 2021; He et al., 2023), and enhanced-efficiency fertilizer, such as polymercoated fertilizer, nitrification inhibitor, urease inhibitor, chemically altered fertilizer, bacterial fertilizer (Lam et al., 2022). Regarding livestock production, a better feed conversion ratio could reduce feed inputs depending on the livestock species and environmental conditions, such as the livestock's thermal environment (Patience et al., 2015; Bai et al., 2016; Sell-Kubiak et al., 2017). Furthermore, good practices in household consumption, i.e. consuming less animal and more plant protein than the Western diet (Resare Sahlin and Trewern, 2022), as recommended by the EAT-Lancet Commission (Willett et al., 2019), could alleviate malnutrition, support human health and reduce environmental emissions from the agrifood system (Errickson et al., 2021; Vaidyanathan, 2021; Wang et al., 2023).

Dietary changes affect land use and crop cultivation, especially when considering circularity (Muscat et al., 2020; van Selm et al., 2022). If residents switch to the EAT-Lancet diet, the demand for plant-based foods will increase, especially for legumes. Ideally, this demand is met with locally grown food to support circularity. Thus, crop switching will be necessary, requiring increased cultivation of soybeans and other legumes to meet the rising demand for plant-based protein. Soybeans and other legumes require fewer agrichemicals and less water input per hectare compared to other cash crops, such as cotton, thereby resulting in lower N and C emissions as well as reduced water consumption (Zhang et al., 2009; Guo et al., 2022). Recent research in China also confirmed that crop switching and redistribution can promote co-benefits, including reductions in environmental impact (GHGs: -5%; water: -9%; fertilizers: -8%; pesticides: -8%) alongside an increase in farmer income (+5%) (Wang et al., 2022b; Xie et al., 2023).

Dietary changes also necessitate adjustments in livestock production to align with healthier diets (Willett et al., 2019; van Selm et al., 2022). The demand for livestock products would decrease with residents adopting the Lancet EAT diet. Ideally, livestock breeding should be scaled down to align with these healthier dietary patterns. This proportionately reduces N and C emissions from livestock production, manure management and energy and power consumption. In addition, this reduces water and

nutrient requirements for feed production. This can potentially spare more land to grow food directly for humans or ecosystem services, considering that around 30% of the global cropland is cultivated to grow livestock feed (Muscat *et al.*, 2020).

Apart from the above options, waste recycling also contributes to closing cycles of N, C and water within agrifood systems, potentially enhancing their water and nutrient use efficiency (Springmann et al., 2018). Substantial amounts of human and animal manure, along with kitchen residue, are disposed of in dump pits or rivers, leading to the discharge of significant amounts of N and C from agrifood systems (Bai et al., 2016; Trimmer et al., 2017; Berendes et al., 2018; Spang et al., 2019). The recycling of organic wastes facilitates the reconnection between sanitation and agricultural practices. This includes the collection of household wastes and animal manure, converting them into fertilizer products, and repurposing food waste and losses as animal feeds (Sandström et al., 2022; Govoni et al., 2023). Moreover, utilizing livestock manure, crop residues, and potentially human waste is considered an efficient way to increase SOC (Maillard and Angers, 2014). Elevated SOC levels not only enhance crop production (Oldfield et al., 2019) but also promote soil resilience and health (Domingo-Olivé et al., 2016; Oldfield et al., 2019). In addition, this aids in reducing C and N emissions to the environment while increasing water retention (Bagnall et al., 2022), but the presence of contaminants, such as antibiotics or heavy metals, may restrict the widespread use of household and livestock wastes (Chee-Sanford et al., 2009; Cobo et al., 2018).

All these options contribute to decreasing N, C and water losses, thus promoting circularity within the agrifood system. However, their contributions may interact with one another. For example, reducing livestock means less available animal manure for crop production. Consequently, more chemical fertilizers may be needed to maintain the optimal fertilizer input as mentioned in good crop production practices. The overall impact on circularity remains unclear considering the range of available options, the varied contributions of different practices, and their interactions.

Promoting circularity necessitates selecting the most relevant spatial scale. Restoring circularity requires quantifying nutrient flows within certain spatial boundaries to describe the current state (Van der Wiel et al., 2020). The area should be sufficiently small to capture the local circumstances, enable transport and thereby facilitate nutrient exchange, but also large enough to include all components of the agrifood system. Field and farm scales (Zhao et al., 2017) are too small as they exclude practices to promote nutrient cycling among households and various farming systems (Giller, 2013). A national scale (van Zanten et al., 2023) is generally too large, obscuring underlying heterogeneity and hindering the optimization of nutrient cycles on smaller scales. Van der Wiel et al. (2021) claimed that the district scale was suitable for restoring nutrient

circularity in Germany because it meets the above requirements. Spiegal *et al.* (2020) proposed that the county level was one of the suitable research units to advance nutrient recycling in the US. A district or county may be a suitable scale for circularity provided that it encompasses the entire agrifood system, with its diverse components spatially close enough to form a network enabling easy exchange of agricultural products and nutrients. In addition, public agricultural extension services are typically organized at this scale.

#### 1.4. Research objectives

How to close N, C and water cycles simultaneously is unclear, particularly at a system level involving households and waste management. Moreover, options promoting circularity within the agrifood system may interact with one another. Furthermore, promoting circularity necessitates selecting the most relevant spatial scale. Therefore, the main aim of this thesis is to explore circularity options for N, C, and water in agrifood systems at the county scale.

We selected Quzhou county (China) as a case study. China is the largest producer and consumer of reactive N (Chen *et al.*, 2016), the largest emitter of CHGs (UNEP, 2020), and a hotspot of water scarcity due to issues with water quantity and quality driven by excessive N and C losses in agriculture (Rodell *et al.*, 2018; van Vliet *et al.*, 2021). The Chinese government pledged to achieve carbon neutrality by 2060 (Feng and Fang, 2022). Quzhou is a demonstration county for China's "Agricultural Green Development" strategy seeking to be a demonstration area for producing nutritious agricultural products while minimizing environmental impacts (MOA, 2020).

Quzhou is in the center of the North China Plain. It has a sub-humid, temperate, continental monsoon climate. The county covers an area of 677 km² and includes 10 towns and 342 village-size administrative units. The registered population was 527,000 in 2017, with over 90% of the inhabitants residing in rural areas (NBSC, 2018). It encompasses all relevant compartments of the agrifood system, with diverse components spatially close enough to enable the exchange of agricultural products and nutrients. The main crops are wheat, maize, cotton and vegetables, while the main livestock includes pigs, laying hens, sheep, and beef cattle. The crop and livestock production as well as human consumption varies among villages. To provide tailored recommendations to local farmers on agronomic technologies, China Agricultural University led to establish the Science and Technology Backyard platform in Quzhou. This platform involves agronomists living in villages among farmers and connects the scientific community with the farming community to facilitate information exchange and innovation. It has collected substantial data on the characteristics of the agrifood system in Quzhou.

Quzhou is a typical county in China which is in transition from extensive low-efficient to intensive high-efficient and high-emission production (Chen et al., 2016; Bai et al., 2018; Wei et al., 2023). Such a transformation is common in middle-income, rapidly-developing countries such as India, Indonesia, Brazil and Pakistan (FAO, 2022; OECD and FAO, 2022; Kang et al., 2023). Low-income regions (such as sub-Saharan Africa) are projected to experience similar transformations in the future with their economic development (FAO, 2022; OECD and FAO, 2022). The development paths of "emit first and treat later" followed by most high-income countries are not replicable for low- and middle-income countries in combating global warming. The options explored in this study provide insights for the global community on managing N, C and water cycles and transitioning towards sustainable circular agrifood systems.

The specific objectives of this thesis are as follows:

- to quantify the loss ratios of N leaching and runoff in croplands and assess N flows in an entire agrifood system (Chapters 2 and 3);
- to evaluate the impact of available options to promote N circularity and their synergistic effects (Chapter 3);
- to assess water security in an agrifood system, elaborated in terms of water quantity and quality, for both surface water and groundwater, along with their potential synergies and trade-offs with N circularity (Chapter 4);
- to explore the potential for achieving net-zero GHG emissions (climate neutrality) in an agrifood system, considering both C and N cycles (Chapter 5).

#### 1.5. Outline of this thesis

This thesis contains a general introduction (this chapter), four research chapters (Chapters 2-5), and a general discussion (Chapter 6). Figure 1.1 visualizes the framework of this thesis and the connections between the chapters.

Chapter 2 quantifies the losing rates of N leaching and runoff for various fertilizer types, which are rarely documented but crucial for both N and water cycles as well as their interactions in agrifood systems (the discharged N could contaminate water and threaten water security). The quantified factors aid in quantifying N cycles in the following chapter.

Combining the above loss ratios, farm surveys, statistical data, a nutrient flow model (NUFER) and parameters from literature review, **Chapter 3** presents N flows in an agrifood system, including not only crop and livestock production but also detailed household data. Key performance indicators (KPIs) in line with circular agriculture are selected to evaluate N circularity of the agrifood system. These KPIs are applied to a

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case study bridging the gap between village and county level N cycle in Quzhou, China. Based on the N cycles, this chapter further evaluates advanced scenarios to transition towards N circularity at the county level.

**Chapter 4** evaluates water security and N circularity, in terms of water quantity and quality, for both surface water and groundwater. The temporal-dynamic water balances are quantified by accounting for detailed water flows in the case study area of Quzhou, China, from 2010 to 2020. Scenario analysis was adopted to evaluates the possibility of achieving synergies between water security and N circularity.

Chapter 5 examines C cycles and net GHG emissions in the agrifood system by integrating the NUFER model with a soil C cycle model (RothC). Net CO<sub>2</sub> emissions from the system are quantified using CO<sub>2</sub>-C sequestration against CO<sub>2</sub>-C emissions from fossil fuels used in the agrifood system of Quzhou, China. Net GHG emissions from the system are quantified by considering both C and N losses. Subsequently, this chapter evaluates potential strategies to attain net-zero CO<sub>2</sub> and GHG emissions in the agrifood system.

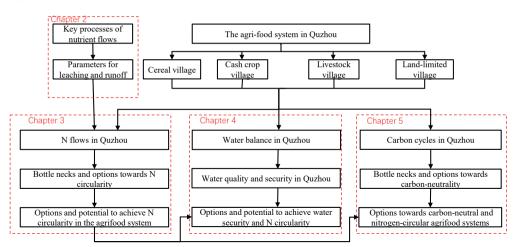


Figure 1.1 Framework for this thesis

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# Organic inputs to reduce nitrogen export via leaching and runoff: A global meta-analysis

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#### Published as:

Zhibiao Wei, Ellis Hoffland, Minghao Zhuang, Petra Hellegers, Zhenling Cui (2021) Organic inputs to reduce nitrogen export via leaching and runoff: A global meta-analysis, *Environmental Pollution* 291, 118176.

https://doi.org/10.1016/j.envpol.2021.118176

#### **Abstract**

Organic inputs as a substitution for, or addition to, chemical fertilizers can potentially mitigate N losses. However, it is not well known how their effects on N leaching and runoff depend on application approaches. We conducted a global meta-analysis of 129 studies to compare the effects of organic inputs on N export via leaching and runoff. We compared three application approaches: chemical fertilizer N substituted by organic fertilizer with: 1) equal amounts of total N or, 2) equal amounts of mineral N and 3) additional organic fertilizer N on top of chemical fertilizer. The meta-analysis showed that organic inputs reduced overall N leaching and runoff by 15% and 29%, respectively, without compromising crop yield, and that this effect was significantly influenced by the application approach taken. Organic substitution of chemical fertilizer N with equal amounts of total N decreased both leaching and runoff by more than 30% and hardly affected crop yield. Substitution with equal amounts of mineral N generally increased crop yield by 6% but also increased N leaching by 21%. Organic inputs in addition to chemical fertilizer N did not affect leaching and runoff. The differences between application approaches were reinforced with increased treatment duration. The loss ratios of leaching and runoff were 14% and 4.5%, respectively, from chemical fertilizer, and 9.2% and 2.6%, respectively, from organic fertilizer. The optimal substitution rates differed between leaching (40-60%) and runoff (60-100%) when substitution was based on equal amounts of total N. We conclude that substitution of chemical for organic fertilizer at equal amounts of total N is most effective in reducing N export via leaching and runoff without compromising crop production.

#### 2.1. Introduction

Overuse of reactive nitrogen (N) induces emissions to the environment, such as N leaching/runoff and other N losses (Sutton et al., 2011; Cui et al., 2018; Cassman and Dobermann, 2021), causing ecosystem eutrophication (Rivett et al., 2008), water quality degradation and loss of biodiversity (Yu et al., 2019). The anthropogenic N discharge (14.5 Tg per year) to freshwater in China was about 2.7 times the estimated 'safe' N discharge threshold (1.0 mg N L-1), of which more than 35% of N discharge was incurred from cropland fertilizer use (Yu et al., 2019). Organic materials (such as straw, manure and waste water products) could potentially be used to substitute for chemical fertilizer and mitigate N export via leaching and runoff (Xia et al., 2017; Malcolm et al., 2019; Tang et al., 2019; van der Wiel et al., 2020). Organic inputs can improve soil quality, increase its water retention capacity and promote crop N uptake (Bergstrom and Kirchmann, 2004; Oldfield et al., 2019), thereby reducing leaching and runoff.

Two organic substitution methods and one addition approach are commonly reported in the literature: substitution of chemical fertilizer with organic fertilizer based on the latter's total N content (Equal N<sub>total</sub>) or its mineral N content (Equal N<sub>min</sub>), and the addition of organic N to chemical fertilizer (Additional Norg). The Equal Ntotal approach is widely used in China (Zhang et al., 2019; Bah et al., 2020). Several meta-analyses have shown that the Equal N<sub>total</sub> approach could mitigate N leaching or runoff by approximately 26%, but either they did not differentiate between leaching and runoff (Wei et al., 2020) or had limited data sources (Xia et al., 2017). The results of the Equal N<sub>min</sub> approach have been reported in Europe and Americas (Bakhsh et al., 2007; Sorensen and Rubaek, 2012; Brockmann et al., 2018). Equal N<sub>min</sub> may increase N losses, especially when the organic fertilizer is surface applied (Bakhsh et al., 2007; Seidel et al., 2017). Most studies are single location trials (Bakhsh et al., 2007; Sorensen and Rubaek, 2012; Seidel et al., 2017), and a meta-analysis of the effects of Equal N<sub>min</sub> on N export via leaching and runoff is lacking. As an alternative to substitution, Additional Norg could reduce N losses either through soil quality effects or immobilization effects using a substrate with a high C/N ratio, or increase them due to extra N application. The addition of organic N to chemical fertilizer is widely practiced in Africa (and in some cases in China too) to increase crop productivity (Chivenge et al., 2011; Sileshi et al., 2019), while N losses were seldom analyzed.

The effect of organic substitution for, or additions to chemical fertilizers on N exports may also depend on the types of organic amendment (Hijbeek *et al.*, 2017; Xia *et al.*, 2018). The application to agricultural soils of organic fertilizers with a high C/N ratio is expected to increase microbial N immobilization capacity and reduce leaching and runoff (Zhang *et al.*, 2013). Using the Equal N<sub>total</sub> approach, Liu *et al.* (2021) analyzed

the responses to N losses using different types of organic fertilizer for vegetable systems in China and found that mixed animal-plant sources of organic N reduced N leaching more than single sources because mixed animal-plant N sources can optimize the C/N ratio to satisfy N and C demand, favoring vegetable yields and N uptake. Soil texture may affect organic matter decomposition (Xia et al., 2018) through effects on aeration and oxygen availability for mineralization, with knock-on effects on leaching and runoff (Ji et al., 2011). Soils with a sandy texture generally have poor retention of water and nutrients, and the increased water infiltration paired with N substrate from organic fertilizer mineralization could aggravate N loss (Blanco-Canqui and Lal, 2009; Xia et al., 2018). Field management, such as fertilizer application (Karimi and Akinremi, 2018) and crop cultivation (Klaus et al., 2018) could also affect the leaching and runoff response. N losses may increase rapidly when the N application rate increases and exceeds the optimum level of crop demand (Wang et al., 2019). Crop cultivation, such as flooding in paddy field, affects anaerobic conditions regulating organic decomposition and N transformations (Klaus et al., 2018).

N export via leaching and runoff can be quantified using the loss ratio (LR). This is defined as the percentage of applied fertilizer N that is lost via leaching or runoff. The Intergovernmental Panel on Climate Change (IPCC) uses a default LR of 30% for N leaching for fertilizers without distinguishing between fertilizer types or locations (Eggleston *et al.*, 2006). Wang et al. (2019) demonstrated that the IPCC overestimated the LR for leaching from (mainly) chemical fertilizers, which should range from 9-20% depending on the crop and fertilizer type. The LRs for leaching and runoff from organic materials have not been systematically analyzed, which could undermine the accuracy of loss inventories and databases of N export via leaching and runoff.

There could be trade-offs or synergies between N leaching/runoff and crop production (Xia et al., 2018; Zhang et al., 2019). Crop yield increased at lower levels of soil organic carbon (SOC) while leveling off or decreasing slightly at 2-3% SOC (Oldfield et al., 2019). Considering that about two-thirds of the world's croplands have a SOC content of less than 2% (Oldfield et al., 2019), we speculate that organic inputs improve SOC, increase crop yield, and thus reduce N surplus for leaching and runoff.

This study aims to: 1) quantify the effects of different organic application approaches on N export via leaching and runoff depending on the type of organic fertilizer; 2) compare the leaching and runoff LRs between chemical and organic fertilizers, and 3) demonstrate the potential trade-offs and synergies between N losses and crop production. To achieve these aims, we conducted a meta-analysis of 129 peer-reviewed articles.

#### 2.2. Materials and Methods

#### 2.2.1. Literature selection

Peer-reviewed papers were collected to evaluate the effects of organic fertilizer inputs on N export via leaching and runoff. Papers published before September 2020 were searched for in Web of Science, Google Scholar and China National Knowledge Infrastructure databases. 'Organic inputs', 'organic substitution', 'organic fertilizer', 'leaching' and 'runoff' were used as search keywords. We also included papers that cited or were cited by Cui *et al.* (2020) and Burkitt (2014) on similar research topics.

The papers included in our analysis met the following criteria: 1) They reported cumulative N leaching or runoff during a crop growth period. If the data were presented as annual rates, these were divided by the number of crops per year; 2) they reported the results of N leaching/runoff using lysimeter and suction cup methodologies which calculated leaching or runoff rate by N concentrations and volume of water flow. The sampling frequency was mostly once a week. After rainfall or irrigation events, samples were collected directly when discharge occurred; 3) the papers reported on both chemical fertilizer (CF, defined as control) and organic fertilizer (OF) treatments, including crop residues (such as green manures or straw; 17%), farm yard manure (29%), slurry (22%) and processed organic materials (composted manure or sewage sludge, digestate and commercial organic fertilizer; 32%), the amounts of which were specified; 4) phosphorus and potassium fertilizers were applied to ensure that these were not yield-limiting; and 5) only field or lysimeter studies were included. Papers that reported data from the same experiment were excluded. Observations conducted at the same site over sampling years were all recorded. In total this analysis included 129 peerreviewed articles on experiments performed worldwide (Asia, 53%; Europe, 31%; Americas, 12%; Africa, 4%), with 511 site/year observations for leaching and 125 observations for runoff. These observations were for various crop types: dryland crops, 77%; rice, 13%; vegetables, 10% (see supplemental database for details).

#### 2.2.2. Evaluation of variables and data treatment

Leaching and runoff were the main response/dependent variables used to evaluate the effects of organic inputs compared with chemical fertilizer only. Leaching and runoff included both organic and mineral N when reported (18%) and otherwise only mineral N (82%). The LRs of leaching and runoff as affected by organic input were calculated as a percentage of the applied fertilizer N lost through leaching or runoff. The responses for crop yield, nitrogen use efficiency (NUE), NH<sub>3</sub>, N<sub>2</sub>O and CH<sub>4</sub> emissions to organic inputs were also evaluated. N<sub>2</sub>O and CH<sub>4</sub> emissions were measured by static chamber, and NH<sub>3</sub> volatilization by dynamic or vented chamber. NUE was calculated as follows:

$$NUE = (U_f - U_C)/R \tag{1}$$

where Uf and Uc indicate the aboveground N uptake of treatment with or without fertilization. R means N application rate.

The effects of organic inputs were evaluated based on the application approach (Equal  $N_{total}$ , Equal  $N_{min}$  and Additional  $N_{org}$ ), treatment duration and substitution rate ( $R_s$ , which is defined as the ratio of organic N input to total N input) and organic types. In most cases the mineral N of organic fertilizers can be quantified by the sum of  $NO_3$ -N and  $NH_4$ +-N (plus uric acid N in some cases). Sometimes, mineral N is also calculated based on the total ammoniacal N retention and organic N mineralization. Table 2.1 summarizes the number of observations and N (mineral and organic) application rate for the different application approaches. Treatment duration was divided into 1 year and >1 year. The  $R_s$  was categorized in four classes ( $4 \le R_s \le 40$ ,  $40 < R_s \le 60$ ,  $60 < R_s < 100$ ,  $R_s = 100$ ). Organic types were grouped into crop residues, farm yard manure, animal slurry and processed organic materials. In addition to these variables, we also entered the following information about the experimental sites into our database, where applicable: fertilizer application rate; irrigation; soil properties (SOC, cation exchange capacity (CEC), soil slope and total N) and climate conditions (precipitation and temperature) (see supplementary data for details).

Table 2.1. Number of observations (n) for leaching and runoff, depending on application approach, and their mean N application rates (kg N ha<sup>-1</sup>) with 95% confidence interval (CI)

Category		n	Chemical fertilizer		Organic fertilizer			
				Mineral N		eral N	Org	anic N
			Mean	CI	Mean	CI	Mean	CI
Leaching	Overall	511	201	192-209	87	79-95	154	144-165
	Equal Ntotal	281	226	214-238	83	72-93	143	132-154
	Equal Nmin	156	162	152-174	51	41-60	216	195-240
	Additional Norg	74	185	166-203	185	166-203	59	47-73
Runoff	Overall	125	217	204-231	128	112-143	106	89-126
	Equal Ntotal	104	223	209-239	120	103-137	103	87-121
	Equal Nmin	6	139	119-159	68	35-101	310	201-446
	Additional Norg	15	205	184-226	205	184-226	47	33-64

Equal  $N_{total}$  means that chemical fertilizer N is substituted by organic fertilizer with equal amounts of total N; Equal  $N_{min}$  indicates substitution based on equal amounts of mineral N; Additional  $N_{org}$  refers to the effects of organic fertilizer N in addition to chemical fertilizer.

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#### 2.2.3. Meta-analysis

The effects of organic inputs on the response variable X (e.g., leaching or runoff) were quantified by effect size, which was defined as the natural log of the response ratio (lnRR) with the following equation:

$$LnRR = Ln\left(\frac{x_O}{x_C}\right) \tag{2}$$

where Xo and Xc represent the mean value of organic and chemical fertilizer treatment for response variable X. Natural log conversion of the response ratio was used to stabilize the variance (Hedges *et al.*, 1999). The results were exponentially backtransformed and presented as the percentage of changes (RR-1)×100% in the variables under organic application. Positive or negative percentage changes denoted an increase or decrease in the corresponding variable.

Effect sizes were weighted by the inverse of the pooled variance (Liu et al., 2019; Wei et al., 2020). For studies that did not report a standard deviation (SD) or standard error (SE), the approach of Bracken (1992) was used to estimate SD in the "metagear" R package (version 3.6.1). A random-effects model was used according to the significance of the residual heterogeneity of the observations in the "metafor" R package (version 3.6.1). A mixed-effects model was used to assess the variations in effect size according to several categorical and continuous factors. The mean effect sizes and the 95% confidence intervals (CIs) were presented in forest plots. Differences between organic fertilizer (OF) and chemical fertilizer (CF) treatment were considered to be significant when the CIs did not overlap zero.

Funnel plots were adopted to evaluate the publication bias (Makowski *et al.*, 2018; Ying *et al.*, 2019). We carried out a trim and fill analysis for the funnel plots in the "metafor" R package (version 3.6.1). Results (effects) were considered acceptable if there was no significant difference before and after trim and fill (Makowski *et al.*, 2018). In cases where the funnel plots test indicated publication bias, the bias-corrected effect size value was adopted, which was estimated using the trim and fill method (Figure A. 1 and 2).

#### 2.3. Results

# 2.3.1. Effects of organic application approaches on N export via leaching and runoff

Organic inputs, on average, significantly decreased N export via leaching and runoff by 15% and 29%, respectively (Figure 2.1). This overall result was dominated by studies in Equal  $N_{total}$  approach, because these were by far the most in terms of numbers (281 out of 511). Application approach had a significant impact on leaching and runoff: Equal  $N_{total}$  approach reduced both leaching and runoff by more than 30%, but Equal  $N_{min}$ 

approach increased N leaching by 21%, while it had no effect on runoff based on the few available observations (n = 6). Additional  $N_{\rm org}$  approach had no effect on leaching and runoff.

The effects of the application approach on N leaching and runoff varied with treatment duration. Equal  $N_{\text{total}}$  decreased N leaching after 1 year, but leaching increased when the substitution was based on Equal  $N_{\text{min}}$ , and was unaffected when organic N was added to chemical fertilizer. The differences between application approaches were reinforced with increasing treatment duration. Leaching was reduced by approximately 40% after more than 1 year at Equal  $N_{\text{total}}$ . However, Equal  $N_{\text{min}}$  increased leaching by 20%. While increased loss due to leaching remained consistent with different treatment regimes, with increased runoff, loss was visible only after more than a year at Equal  $N_{\text{min}}$ .

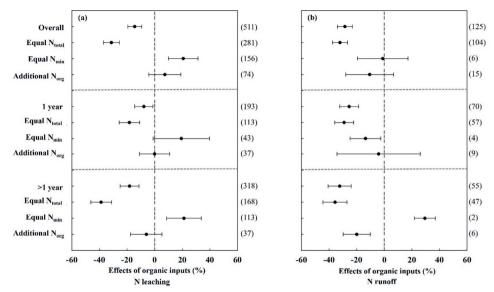


Figure 2.1. Effects of organic inputs on N leaching (a) and runoff (b). Error bars represent 95% confidence intervals. Numbers in parentheses indicate numbers of observations.

The rate of N fertilizer application did not correlate with the effect of organic inputs on leaching, regardless of the application approach (Figure 2.2a). It also did not lead to differences in leaching related to application approaches. Runoff was significantly more reduced (p< 0.01) with lower N application rates when the substitution was based on Equal  $N_{total}$  (Figure 2.2b). The relationship was not significant for Equal  $N_{min}$  (p = 0.28) or Additional  $N_{org}$  (p = 0.68) based on our limited data (n=6 for Equal  $N_{min}$ , 15 for Additional  $N_{org}$ ; Figure 2.2b).

# Organic inputs to reduce nitrogen leaching and runoff

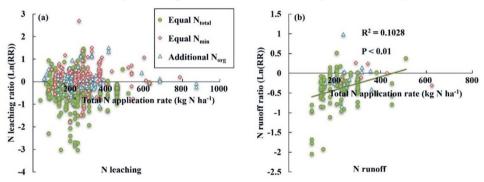


Figure 2.2. Effects of total N application rate on N leaching (a) and runoff (b) loss ratios of organic inputs. Negative and positive value indicates decreased or increased N leaching caused by organic inputs.

Organic inputs reduced leaching and runoff independently of the substitution rate in the Equal  $N_{total}$  approach. The optimal substitution ratio ( $R_s$ ) to mitigate N leaching was 40-60%, which decreased leaching by 45% (Figure 2.3). For  $R_s > 60\%$ , leaching did not decrease significantly compared to no substitution. Runoff decreased by 22% at a low substitution rate ( $4 \le R_s \le 40$ ), and the optimal  $R_s$  to mitigate runoff was in the 60-100% range. Runoff tended to decrease gradually as  $R_s$  increased.

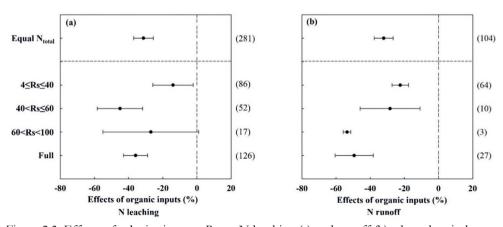


Figure 2.3. Effects of substitution rate Rs on N leaching (a) and runoff (b) when chemical fertilizer N is substituted by organic fertilizer with equal amounts of total N. Rs is defined as organic N input/total N applied (%). Error bars represent 95% confidence intervals. Numbers in parentheses indicate numbers of observations.

# 2.3.2. N export via leaching and runoff affected by organic material types and other factors

Organic material types affected N export via leaching and runoff under organic inputs (Figure 2.4). Crop residues with a higher C/N ratio (40) reduced N leaching and runoff by 22%. Farmyard manure with a low C/N ratio did not affect leaching but decreased N runoff by 23%. Animal slurry with a low C/N ratio (13) significantly affected neither leaching nor runoff. Processed organic materials reduced N leaching and runoff (33%) more than other types, while they had a significantly lower C/N ratio than crop residues. Organic inputs decreased leaching (Figure A. 3) and runoff (Figure A. 4) more when the SOC was low. The effects of organic inputs were not dependent on clay content and crop cultivation (Figure A. 5).

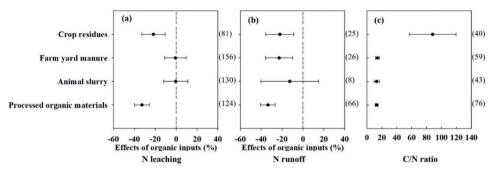


Figure 2.4. Effects of organic types on N leaching (a) and runoff (b), and their C/N ratio (c). Error bars represent 95% confidence intervals. Numbers in parentheses indicate numbers of observations.

The leaching and runoff LRs for chemical fertilizer were 14% and 4.5% of applied N, respectively. Organic inputs significantly reduced these LRs to 9.2% and 2.6%, respectively (Table 2.2). Additional  $N_{\rm org}$  reduced the LR for leaching most (7.0%) compared to other approaches, while Equal  $N_{\rm total}$  had the lowest LR for runoff (2.3%). The substitution rate  $R_s$  did not significantly affect either of the two LRs. Organic types did not affect the runoff LR but they did affect leaching. Processed organic materials showed a significantly lower leaching LR (6.1%) than animal manure (11%).

## Organic inputs to reduce nitrogen leaching and runoff

Table 2.2. Loss ratios (LRs) for leaching and runoff with 95% confidence interval (CI) depending on application approach, substitution rate and organic type.

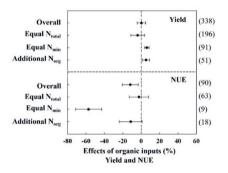
Category			Leachir	ng	Runoff			
		n	Mean	CI	n	Mean	CI	
Chemical fertiliz	zer	511	14	12-15	125	4.5	3.3-5.7	
Organic fertilize	er	511	9.2	8.2-10	125	2.6	1.9-3.3	
Equal Ntotal		281	8.8	7.3-10	104	2.3	1.8-2.9	
Rs (%)	0-40	86	6.9	5.1-8.9	64	2.3	1.6-3.2	
	40-60	52	6.1	3.7-8.9	10	2.2	0.8-3.9	
	60-100	17	10	5.4-16	3	1.4	0.3-3.4	
	100	126	11	8.4-14	27	2.5	1.5-3.6	
Equal Nmin		156	11	9.8-12	6	4.5	0.1-10	
Rs (%)	0-40	38	14	12-15				
	40-60	18	12	7.5-17				
	60-100	17	9.8	7.7-12				
	100	83	9.8	8.1-12				
Additional Norg	g	74	7	4.7-10	15	4.1	0.9-7.8	
Organic type	Crop residues	84	7.1	4.9-9.6	25	3.6	1.6-6.1	
5 /1	•							
	Animal manure	286	11	9.4-13	34	1.5	0.6-2.6	
	Processed manure	141	6.1	4.9-7.4	66	2.8	2.0-3.6	

R<sub>s</sub>, substitution rate, defined as organic N input/total N applied x100 (%).

#### 2.3.3. Effects of organic application approaches on crop production

Organic fertilizer application overall showed only minor effects on crop yield (Figure 2.5). Crop yield varied with organic application approaches. Equal  $N_{\rm total}$  did not significantly change crop yield, while Equal  $N_{\rm min}$  and Additional  $N_{\rm org}$  increased crop yield by 6% and 5%, respectively. NUE was decreased by organic inputs. Equal  $N_{\rm min}$  decreased NUE by 57%, while Equal  $N_{\rm total}$  and Additional  $N_{\rm org}$  did not significantly affect it.

The substitution rate  $R_s$  affected the crop response to organic inputs under the Equal  $N_{total}$  approach (Figure 2.6), and crop yield was significantly increased when  $R_s$  ranged from 40-60%, while it tended to decrease for other rates.



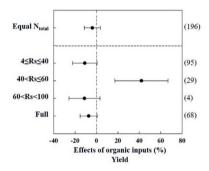


Figure 2.5. Effects of application approach on crop yield and Nutrient Use Efficiency. Error bars represent 95% confidence intervals. Numbers in parentheses indicate numbers of observations.

Figure 2.6. Effects of Rs on crop yield when chemical fertilizer N is substituted by organic fertilizer with equal amounts of total N. Rs, substitution rate, defined as organic N input/total N applied \* 100 (%). Error bars represent 95% confidence intervals. Numbers in parentheses indicate numbers of observations.

#### 2.4. Discussion

This study shows that reducing N export via leaching and runoff losses from agricultural fields through organic inputs depends on the application approach used. We demonstrated that partially replacing chemical fertilizer N by organic N based on equal amounts of total N can reduce leaching and runoff by approximately 30% across a wide range of environmental conditions. The leaching and runoff loss ratios of N were reduced from 14% and 4.5%, respectively, for chemical fertilizer to 9.2% and 2.6%, respectively, for organic fertilizer. The type of organic fertilizer applied is a major determinant of the loss reductions, with processed organic materials and crop residues being superior to animal manure. The reduced impact on the environment is achieved with no impact on yield. However, replacing chemical fertilizer N with organic fertilizers based on equal amounts of mineral N generally increases leaching. Adding organic fertilizer to chemical fertilizer did not significantly affect leaching and runoff.

# 2.4.1. Leaching and runoff and crop production responses to organic application approaches

To our knowledge, this is the first global meta-analysis to examine how the full suite of organic application approaches affects N export via leaching and runoff. Replacing chemical fertilizers based on Equal  $N_{\text{total}}$  reduced leaching and runoff by approximately 30% (Figure 2.1), which can potentially be explained by an improvement in the soil's water retention capacity (Bergstrom and Kirchmann, 2004), the slower release of mineral N from organic fertilizer or better synchronization of N supply from the

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organic fertilizer and crop N demand; (Sorensen and Rubaek, 2012; Fan *et al.*, 2017). This result is supported by previous meta-analyses which suggested that N losses were reduced by 26% under organic inputs (Xia *et al.*, 2017; Wei *et al.*, 2020). The optimal R<sub>s</sub> to reduce leaching was 40-60% (Figure 2.3). This corroborates our observation that crop yield was significantly increased when the R<sub>s</sub> was in this range (Figure 2.6). The optimal R<sub>s</sub> for runoff was 60-100%, but crop yield decreased at this R<sub>s</sub>.

N leaching significantly increased when chemical fertilizer was replaced by organic based on Equal N<sub>min</sub>. This is most likely because extra (organic) N was applied in the organic fertilizer (267 kg N ha<sup>-1</sup>) compared to the chemical fertilizer treatment (162 kg N ha<sup>-1</sup>) and, in most cases, there was extra N in excess of crop demand (150-210 kg N ha<sup>-1</sup>; Cui *et al.*, 2018; Liu *et al.*, 2021). This excess N was obviously not compensated for by improvements in soil quality that might have reduced the leaching and runoff. Organic N that is mineralized after the cropping season can contribute to leaching and runoff loads when drainage occurs (Yaguee and Quilez, 2015). This was supported by our observation that N export via leaching and runoff increased as treatment duration increased (Figure 2.1).

N leaching was not affected when organic fertilizer was added to chemical fertilizer (Additional N<sub>org</sub>; Figure 2.1a). Straw was used for this in the largest number of studies (71%) with the Additional N<sub>org</sub> approach. Crop residues with a higher C/N ratio (40) than farm yard manure (14) showed a greater potential to reduce N leaching (Figure 2.4). Previous studies also reported that organic sources differed in their potential to mitigate leaching and runoff (Xia et al., 2018; Zhang et al., 2019). Organic sources with a higher C content favor the N immobilization process and thus reduce leaching and runoff (Fan et al., 2017; Liu et al., 2021). Interestingly, processed organic materials could reduce leaching and runoff more while their C/N ratio (14) is lower than that for crop resides. This could relate to carbon conversion during composting or anaerobic digestion processes. For example, processed materials may contain more lignocelluloses and hemicelluloses which are more resistant to microbial decomposition (Xia et al., 2017). Any further underlying reason is unclear and would need further research. Obviously, the C/N ratio, as such, does not fully explain the effect of organic fertilizers on leaching and runoff.

Runoff was not significantly affected by Equal N<sub>min</sub> and Additional N<sub>org</sub> approaches (Figure 2.1b), although their total N application rates were relatively high (262 and 267 kg N ha<sup>-1</sup>, respectively; Table 2.1). On the one hand, organic inputs induce off-season N mineralization which could promote N runoff. On the other hand, organic inputs favor soil aggregation, which reduces soil erosion and runoff (Wang *et al.*, 2020). Organic inputs also loosen topsoil and thus reduce water flows laterally, which is the

prerequisite for N runoff (Tan et al., 2015). The above mechanisms counteract each other, resulting in an insignificant result for runoff for Equal N<sub>min</sub> and Additional N<sub>org</sub>.

Organic inputs did not affect crop yield (Figure 2.5). This result did not support our expectation that organic inputs would increase crop yield and N uptake by improving soil quality. A potential reason for this may be the slow release of mineral N from organic fertilizer decomposition (Bergstrom and Kirchmann, 2006; Sieling and Kage, 2006), which is offset by but not overcompensated for by improved soil quality and reduced losses. Crop yield is improved only when the treatment duration is more than 3 years (Xia et al., 2018; Wei et al., 2020), while our observation in 66% of cases was less than 3 years. Equal N<sub>min</sub> increases crop yield but also reduces NUE (Figure 2.5), which is at least partly due to increased leaching losses. Organic inputs decrease NH<sub>3</sub> emissions and increase CH<sub>4</sub> emissions, but have no significant effect on N<sub>2</sub>O emissions based on our limited data which does not distinguish between application approaches (Figure A. 6). Interestingly enough, application approaches are related to regional and country differences. In China, Equal N<sub>total</sub> was used in 77% of our observations, with >90% of them applied to substitute for urea. In Europe, Equal N<sub>min</sub> was adopted in 60% of cases, with animal slurry often used to replace ammonium nitrate or urea ammonium nitrate (see supplemental database for details).

The Equal  $N_{total}$  approach was more effective in mitigating N losses without sacrificing crop production than the Equal  $N_{min}$  or Additional  $N_{org}$  approaches (Figure 2.1 and 2.4). This may be partly due to the N application rate. The average N application rate was 226 kg N ha<sup>-1</sup> per season for the Equal  $N_{total}$  approach (Table 2.1). Therefore, crop demand (150-210 kg N ha<sup>-1</sup>) can still be met when substituting part of the active mineral N by slow-release organic N.

# 2.4.2. Factors determining how organic inputs affect leaching and runoff

Leaching (Figure A. 3) and runoff changes (Figure A. 4) affected by SOC, CEC, soil texture and water input were tested, independently. Leaching and runoff did not correlate significantly with the tested indicators, apart from SOC. Organic inputs decreased leaching and runoff more when the SOC was low, which could be explained by the effects of SOC on crop yield and N uptake: organic amendment increases the SOC more when the initial SOC is low. The increased SOC promotes crop yield and N uptake, and thereafter reduces N surplus and losses (Oldfield *et al.*, 2019; Waqas *et al.*, 2020). Organic inputs could also mitigate N discharge at high levels of SOC (> 30 g kg<sup>-1</sup>) with the mitigation potential promoted when SOC increases further. Such a result is probably related to water retention due to soil aggregation (Hoffland *et al.*, 2020).

## Organic inputs to reduce nitrogen leaching and runoff

We arrived at a leaching and runoff LR of 14% and 4.5%, respectively, for chemical fertilizer N (Table 2.2). This is similar to previous studies which reported LRs of 9-20% for leaching and 1-4.5% for runoff (Nesme *et al.*, 2010; Sørensen and Jensen, 2013; Wang *et al.*, 2019). Organic inputs overall reduced the leaching and runoff LRs to about half of that for chemical fertilizer N, so the default value set by the IPCC (30%) is too high, and this work emphasizes the relevance of LR calibration for leaching and runoff for different types of fertilizer.

#### 2.4.3. Limitations and implications

Although we looked for articles without imposing any geographic limitations, over 50% of the studies were carried out in Asia, especially in China. Since most of the Chinese experiments were designed on the basis of Equal N<sub>total</sub>, the "overall" results in Figure 2.1 and 2.5 were dominated by Equal N<sub>total</sub>. Even though P and K were not limiting factors for crop growth in the articles included, organic materials provide other essential nutrients, such as zinc (Zn) and boron (B), which may also affect crop yield and N losses (Franke *et al.*, 2008; Wei *et al.*, 2020). Moreover, when examining the effect of treatment duration on organic application approaches, the studies were only divided into two groups (1 year and >1 year) due to limited data, which might therefore not comprehensively reflect the long-term effect. Additionally, due to data limitations, we did not distinguish between application approaches when analyzing the leaching and runoff response to various other influencing factors.

Our manuscript reveals larger patterns at a global level and found substitution of chemical for organic fertilizer at equal amounts of total N could reduce leaching and runoff without compromising crop production. Scientists can gain some insight into application approaches and make large scale evaluations or model validations. Further research is needed to quantify the effect of organic application approaches on N losses with different treatment durations, because repeated application of organic fertilizers will lead to a build up of soil organic matter, which may result in increased soil fertility and losses in the long term. For managers working at higher scales of spatial resolution (i.e., field, village or county), more on-site experiments are required.

#### 2.5. Conclusions

This global meta-analysis looked at the effects of different organic fertilizer application approaches on N export via leaching and runoff. Partial substitution of chemical fertilizers by organic fertilizers with the same amount of total N generally reduces leaching and runoff without compromising crop yield. Processed organic materials with a lower C/N ratio reduce leaching and runoff more and have lower loss ratios compared with crop residues and animal manure. The C/N ratio does not fully explain the effect of organic fertilizers on leaching and runoff. Although we could not distinguish between application approaches when analyzing organic types due to data limitations, this work emphasizes the importance of loss ratio calibration for leaching and runoff for different types of fertilizers.

# **Acknowledgements**

We are grateful to Oene Oenema for providing some research ideas. This study was funded by Science and Technology Project of Yunnan Branch of China National Tobacco Corporation, Research and Demonstration of Non-point Source Pollution Prevention Technology in Tobacco-planting Area of Erhai Lake Basin (2020530000241005); China Scholarship Council (No. 201913043).

# **Supporting Information**

Supplementary information associated with this article can be found in the online version, at https://doi.org/10.1016/j.envpol.2021.118176

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# Towards circular nitrogen use in the agrifood system at village and county level in China

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#### Published as:

Zhibiao Wei, Minghao Zhuang, Petra Hellegers, Zhenling Cui, Ellis Hoffland (2023) Towards circular nitrogen use in the agrifood system at village and county level in China, *Agricultural Systems* 209, 103683. https://doi.org/10.1016/j.agsy.2023.103683

#### **Abstract**

Increased nitrogen (N) losses from linear agrifood systems result in severe environmental issues. These problems can be mitigated by circular N use. Yet. circularity is a scale-specific problem, and the feasibility of pathways towards N circularity at different scales is unclear. This study aimed to 1) evaluate N circularity of a complete agrifood system, including household compartment, at both the village and county scale; and 2) assess potential pathways towards N circularity. We used the county of Quzhou (China) with 342 villages as a case study and applied the modified version of a nutrient flow model (NUFER) to calculate N flows, using national statistical data and own survey data collected from farms and households. To evaluate N circularity, we selected four key performance indicators: N import, N loss, N use efficiency and N recycling rate. Our analysis showed significant variation at the village scale, depending on local production and consumption patterns. At county level, total N import was 546 kg ha<sup>-1</sup> yr<sup>-1</sup>, of which 54% was lost to the environment. The N use efficiency of the agrifood system and N recycling rate of excreta were both <30%. We investigated scenarios to increase N circularity, including application of good management practices in crop and animal production; household dietary change to more plant protein; recycling of organic waste; growing legumes instead of cotton; and reducing livestock breeding. All measures combined increased the system's N use efficiency by 172% and N recycling rate by 87%, while reducing N import by 68% and N loss by 77%. Recycling of organic waste was the most effective and most feasible strategy to promote circularity. Our study bridges the gap between village and county level N cycles, illustrates possibilities to transition towards circular N use, and can help policymakers worldwide to achieve more sustainable agrifood systems.

#### 3.1. Introduction

Nitrogen (N) is fundamental for human life, while excessive N induces environmental concerns. Across the world, agrifood systems suffer from substantial N losses, leading to major sustainability challenges such as waste of resources, heavy reliance on external inputs, air and water pollution, and water eutrophication (Poore and Nemecek, 2018; Springmann *et al.*, 2018; Domingo *et al.*, 2021). These problems will increase further with a larger and more affluent global population (Bodirsky *et al.*, 2014). Thus, there is an urgent need for transition to circular, environmentally sustainable agrifood systems in which the N cycle is closed (Billen *et al.*, 2021; Velasco-Muñoz *et al.*, 2021). Such a transition requires a redirection of agrifood system N flows, which is complex due to many interactions between the compartments of crop production, livestock production, food processing, and household consumption. To support this transition, it is important to quantify these flows and interactions, both in current systems and in scenarios exploring measures toward N circularity.

Nitrogen circularity should include N flows in the entire agrifood system, including the household component. However, literature on N flows and losses focuses mainly on crop and livestock production (Herrero et al., 2010; Renard and Tilman, 2019; Xu et al., 2021) and only to a limited extent on households or complete agrifood systems. The crop-grassland-livestock balance was reported to determine the robustness of food production in three types of European farming systems, assessed with a dynamic N flow model (Pinsard et al., 2021). The NUFER model (Nutrient Flows in Food Chain, Environment and Resource use) was used to quantify N flows in the food chain without detail human consumption data (Ma et al., 2010; Meng et al., 2022). Households N flows have generally received limited attention, but scientists increasingly point out the potential contribution of dietary change and food waste reduction to mitigate N losses (Willett et al., 2019; Chen et al., 2020; Vaidyanathan, 2021). Recycling N from human excreta is also receiving increased attention. The global mass of human excreta may surpass 1 Pg per year by 2030 (Berendes et al., 2018). Human excreta cause 1-3% of global N2O emissions (McNicol et al., 2020), while well-managed and recycled human excreta could substitute 15% of chemical N fertilizer and contribute to global long-term food and nutrient security (Rose et al., 2015; FAOSTAT, 2019; Harder et al., 2020).

The concept of circular agriculture was proposed to link agricultural production with sustainable resource use (Kirchherr et al., 2017; Velasco-Muñoz et al., 2021), and it has guided reshaping the agrifood system to eliminate pollution and reuse resources. For example, Zhang et al. (2019) proposed to recouple livestock production with cropland and feed livestock based on the manure carrying capacity of cropland, to mitigate agricultural pollution. Muscat et al. (2021) summarized several principles related to

circular N use in the food system, such as safeguarding the health of the agroecosystems, avoiding unnecessary products, using biomass efficiently, and recycling unavoidable byproducts. These principles of circular agriculture are abstract; therefore, indicators are needed to evaluate circularity (Moraga *et al.*, 2019; Velasco-Muñoz *et al.*, 2021). Specifically, we need some key performance indicators (KPIs) that reflect the main flows and processes, and which are relatively easy to collect and calculate.

Apart from selecting KPIs to evaluate N circularity, it is important to select the most relevant spatial scale. Restoring N circularity requires quantifying N flows within certain spatial boundaries to describe the current state (Van der Wiel et al., 2020; Papangelou and Mathijs, 2021). This area needs to be sufficiently small to capture the local circumstances, enable transport and thereby facilitate N exchange, but also large enough to include all compartments of the agrifood system (Van der Wiel et al., 2020). The national scale (Bodirsky et al., 2014; Luo et al., 2018) is generally too large as it masks underlying heterogeneity and impedes optimization of N cycles on smaller scales; the field or farm scale (Zhao et al., 2017) is too small as it excludes options to improve nutrient cycling between various farming systems (Giller, 2013). Van der Wiel et al. (2021) proposed that district may be a suitable scale in Germany, while in China, the county scale meets these requirements. It contains the full agrifood system, and its various compartments of the agrifood system are spatially close enough to form a network in which agricultural products and nutrients can be easily exchanged. In addition, public agricultural extension services are generally organized at county level and farm recommendations and agricultural policies are often implemented by a county agricultural bureau. However, to understand nutrient flows at the county scale, it is important to recognize the underlying heterogeneity at the scale of towns and villages that have different types of production and consumption patterns and population densities.

Based on gaps identified above, this study aimed to 1) evaluate nitrogen circularity of a complete agrifood system, including households, using KPIs to identify the main limitations for closing N cycles; 2) conduct this analysis by integrating village-level patterns to county-level flows; and 3) assess potential and feasible options towards circular N use bridging the gap between village and county scale, using scenario analysis. As a case study, we used Quzhou County (China). China is the largest producer and consumer of reactive N in the world (Gu et al., 2015; Chen et al., 2016) and its agricultural N flows are characterized by high input, high output, high surplus, and high dependency on synthetic fertilizer (Chen et al., 2016). In Quzhou, agricultural production is typical of rural areas across China, having evolved from low-input, low-output to high-input, high-output farming. Currently, Quzhou is a demonstration county for China's

"Agricultural Green Development" national strategy (MOA, 2020), aimed at high and nutritious production with low environmental impact.

#### 3.2. Materials & Methods

#### 3.2.1. Study area

The county of Quzhou is in the central part of the North China Plain. It covers an area of 677 km² and includes 342 village-size administrative units (hereafter as "villages"), including units in urban areas (NBSC, 2018). The distance between adjacent villages is less than 1 km on average. The county's registered population is 527,000, of which 491,000 live in rural areas. Farms generally consist of multiple plots of arable land, ranging from 0.03 to 2 ha, spread out across the village. The average arable land per household is 0.6 ha. Agricultural production in Quzhou has changed substantially; in the last 40 years, grain yield has increased from 1.5 to 7.1 Mg ha¹ due to increased use of chemical fertilizers and pesticides; meat production has increased fifty times from 1145 to 57440 Mg per year.

To account for the heterogeneity in N flows in a feasible way, we grouped the 342 villages in Quzhou into four types: (a) cereal villages, (b) cash crop villages, (c) livestock villages and (d) land-limited villages (Table 3.1). This approach proved to work well for villages participating in the Science and Technology Backyard platform in Ouzhou (Zhang et al., 2016). In cereal villages, the proportion of cereal crop areas is larger than average across Ouzhou, while in cash crop villages, the proportion of cash crop (including cotton, fruits and vegetables) areas is larger than average. In livestock villages, production of maize (main livestock feed in Quzhou) is lower than consumption, while in land-limited villages, less than 0.07 ha (1 mu) of arable land is available per capita and/or production of wheat (main staple food in Quzhou) is lower than consumption. These criteria were mutually exclusive for all except 13 villages, which met the criteria of both land-limited and livestock villages. We arbitrarily classified these 13 as landlimited villages, as their livestock densities were more like land-limited than livestock villages. The division is conceived particularly for this study and does not presume to suggest universal criteria for village classification. The characteristics of each village type (Table 3.1) are average values of all villages within the type. Villages in the same type are considered to be similar, so N flows from one village type to the same would not take place.

Table 3.1. Characteristics of the four village types distinguished for evaluating N circularity in Quzhou

Village type n		Main crops (ha/village)					Number of livestock (head/village)				Resident		
		Wheat	: Maize	Cotton	Vegetable	Fruit	Arable land	Pig	Cattle	Sheep /goat	Meat poultry	Layer poultry	population per village
"Cereal"	120	103	108	16	5	5	185	211	67	776	1101	2088	1542
"Cash crop"	87	75	88	27	24	7	161	241	69	914	5270	1562	1389
"Livestock"	82	82	84	24	7	5	145	2313	106	1087	10844	69668	1375
"Land- limited"	53	47	54	10	12	5	80	1377	49	570	4032	16436	1733

Note: n indicates the numbers of villages. Data source: Quzhou Statistical Yearbook 2017.

#### 3.2.2. Selection of KPIs

To evaluate N circularity of the agrifood system, we selected KPIs that reflect the main principles of circular agriculture: (1) reducing external inputs, (2) reusing waste streams, and (3) reducing environmental losses (Muscat et al., 2021; Velasco-Muñoz et al., 2021). For the principle of reducing external inputs, we used N import as KPI; for reusing waste streams, N recycling rate (NRR); and for reducing environmental losses, N use efficiency (NUE) and N loss. For the latter principle, we selected two KPIs, because neither NUE nor N loss alone is suitable for comparing open and closed systems. For example, intensive agricultural systems may have a relatively high NUE but are still characterized by an open N cycle with substantial N losses (Van der Wiel et al., 2020). The KPIs are defined below (section 3.2.3.3) in formulaic expressions.

#### 3.2.3. Calculation of N flows and KPIs

#### 3.2.3.1. Adapted NUFER model

The time scale of this research is one year, and the agrifood system based on the 2017 county statistical data and our own 2019-2020 survey data is considered as the baseline scenario (S0). N flows were quantified throughout the entire agrifood system, including the compartments of crop production, livestock production, food processing, and household consumption, at village (type) and county level. N flows were quantified both within and between the different village types (Table 3.1) using the NUFER model (Ma et al., 2010). While the original NUFER model was mainly used to assess N flows in crop and livestock production (Zhao et al., 2017; Bai et al., 2018; Meng et al., 2022), we added the household compartment, modified and expanded the model based on

detailed survey data on village-level household consumption and waste management (Supplementary Material). Nitrogen flows in the agrifood system were firstly quantified for each village type and then scaled up to county level based on N flows normalized by arable land and the average arable land area of each village type (Figure 3.1).

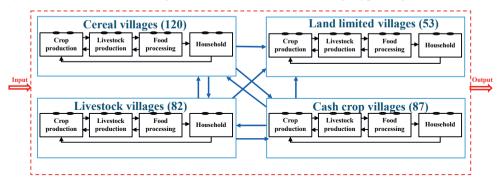


Figure 3.1. Adapted NUFER model, including households and integrating village and county level N flows. The blue and red lines indicate the boundaries at the village and county level, respectively; the black, blue and red arrows indicate N flows within villages of the same type, between different village types, and across county boundaries, respectively. The input and output are depicted at the village scale but are further detailed to each of the four compartments. The figures in brackets indicate the number of villages of each type. The flows are for all villages of the same type (Table 3.1).

#### 3.2.3.2. Village (type) level N inputs

For each village type, N inputs were calculated for the crop production, livestock production, and household compartment. For the food processing compartment, we only considered milling of wheat and maize due to limited data and supposed minor effect of this compartment. For each crop and livestock species, we used crop- and livestock-specific input data, and these inputs were integrated to village level using the average crop areas and livestock densities of each village type (Table 3.1).

Total N input into the crop production compartment of each village type (Ictotal) was calculated as:

$$I_{C_{total}} = I_{C_{chemical fertilizer}} + I_{C_{organic fertilizer}} + I_{C_{residue}} + I_{C_{deposition}} + I_{C_{BNF}} + I_{C_{Irrigation}} + I_{C_{seed}}$$
 (1)

Where the inputs are chemical fertilizer (Ic<sub>chemical fertilizer</sub>), organic fertilizer including livestock manure and human excreta (Ic<sub>organic fertilizer</sub>), residues from last crop (Ic<sub>residue</sub>), deposition (Ic<sub>deposition</sub>), biological N fixation (Ic<sub>BNF</sub>), irrigation water (Ic<sub>Irrigation</sub>), and seeds (Ic<sub>seed</sub>).

Total N input into the livestock compartment (Iltotal) was calculated as:

$$||_{total} = ||_{imported feed} + ||_{domestic feed} + ||_{young stock}$$
 (2)

where the inputs are imported feed (Il<sub>imported feed</sub>), feed produced within the village type (Il<sub>domestic feed</sub>), and imported young stock from outside the village type (Il<sub>young stock</sub>).

Total N input into the household compartment (Ih<sub>total</sub>) was calculated as:

$$Ih_{total} = Ih_{domestic food} + Ih_{imported food}$$
 (3)

where Ih<sub>domestic food</sub> is N input from food produced within the village; Ih<sub>imported food</sub> is N from food imported outside the village.

Total N input into the village agrifood system (Is<sub>total</sub>) was calculated by summing the abovementioned N inputs into the crop production, livestock production and household compartments, corrected for internal flows (N from crop residues, feed and food produced and used within the same village type), as follows:

$$Is_{total} = Ic_{total} - Ic_{residue} + II_{total} - II_{domestic feed} + Ih_{total} - Ih_{domestic food}$$
(4)

# 3.2.3.3. Calculating KPIs for evaluating N circularity at village (type) level

The KPI of N import into the agrifood system (NI<sub>s</sub>) was calculated as follows:

$$NI_{s} = I_{S_{total}} - I_{C_{deposition}} - I_{C_{BNF}} - I_{C_{Irrigation}}$$
 (5)

N deposition, BNF and N in irrigation water were substracted from N input (eq. 4), because these flows were not imported from a specific region and therefore did not compromise N circularity.

The KPI of N loss was calculated by adding the N losses in the crop production, livestock production, food processing, and household compartments. Nitrogen loss from the crop production compartment (NL<sub>c</sub>) was calculated as follows:

$$NL_{c} = Oc_{NH^{3}} + Oc_{N^{2}O} + Oc_{denitrification} + Oc_{leaching} + Oc_{runoff} + Oc_{erosion} + Oc_{discarded residue}$$
 (6)

where the losses are  $NH_3$  volatilization ( $Oc_{NH^3}$ ),  $N_2O$  emission ( $Oc_{N^2O}$ ), denitrification ( $Oc_{denitrification}$ ), leaching ( $Oc_{leaching}$ ), runoff ( $Oc_{runoff}$ ), erosion ( $Oc_{erosion}$ ), and discarded crop residues ( $Oc_{discarded\ residue}$ ).

Nitrogen loss from the livestock production compartment (NL<sub>I</sub>) was calculated as follows:

$$NL_{I} = OI_{manure H&S} + OI_{manure discharge} + OI_{manure treatment}$$
 (7)

where  $Ol_{manure\ H\&S}$  is N losses from manure during the housing and storage stage, where N is emitted as NH<sub>3</sub>, N<sub>2</sub>O and N<sub>2</sub> to air;  $Ol_{manure\ discharge}$  is N losses via manure discharge to water or landfill;  $Ol_{manure\ treatment}$  indicates N losses during manure treatment, in which N is lost as NH<sub>3</sub>, N<sub>2</sub>O and N<sub>2</sub> to air, or through leaching and runoff to water. In the present paper, these losses are collectively referred to as livestock manure losses.

Nitrogen loss from the food processing compartment (NL<sub>p</sub>) was estimated using a milling loss ratio of 5% for wheat and 2% for maize (Ma et al., 2010). Bran yield (20% in wheat milling, 3% in maize milling) was not considered a loss; all bran was assumed to be used for feeding local livestock, based on observations and the fact that local livestock consumed more bran than the amount produced within Quzhou.

Nitrogen loss from the household compartment was estimated based on our year-round survey data (see section 3.2.5).

Based on the above losses from the various compartments, total N loss of the agrifood system (NL<sub>s</sub>) was defined as follows:

$$N loss_s = NL_c + NL_l + NL_p + Oh_{food losses} + Oh_{excreta losses} + Oh_{living losses}$$
(8)

where NL<sub>p</sub>, Oh<sub>food losses</sub>, Oh<sub>excreta losses</sub>, Oh<sub>living losses</sub> represent N loss from food processing, household food waste (Figure S2), discharged human excreta, and human daily life (Ma et al., 2010).

The KPI of NUE was calculated both at the compartment level and system level. The NUE of crop production (NUE<sub>c</sub>) was calculated as follows:

$$NUE_{c} = (Oc_{main product} + Oc_{residue for feeding})*I 00/Ic_{total}$$
(9)

where  $Oc_{main\ product}$  is N output in main crop products;  $Oc_{residue\ for\ feeding}$  is N output in crop residues used for feeding livestock.

The NUE of livestock production (NUE<sub>1</sub>) was calculated as follows:

$$NUE_{l} = (OI_{livestock} + OI_{egg} + OI_{milk})*100/II_{total}$$
(10)

where Olivestock, Olege, and Olmik are N output in livestock, eggs, and milk, respectively.

The NUE of the agrifood system (NUE<sub>s</sub>) was calculated according to Ma et al. (2010), as follows:

$$NUE_{s} = (Oc_{export} + Ol_{export} + Ih_{total} - Ih_{imported food})/(Is_{total} - Ih_{imported food})$$
 (11)

where  $Oc_{export}$  is N output in crop product export (including crop residues for feeding);  $Ol_{export}$  is N output in livestock product export.

The KPI of NRR was calculated both at the compartment level and system level. The NRR of crop production compartment (NRRc) was calculated as follows:

$$NRRc = Oc_{recycled residue} * 100/(Oc_{residue} - Oc_{residue for feeding})$$
 (12)

where Oc<sub>recycled residue</sub> is residue-N recycled to cropland and Oc<sub>residue</sub> is total residue-N production. Crop residues used for feeding livestock were subtracted from total residue production because they are regarded as marketable products (as grains), rather than waste.

The NRR of livestock production (NRRI) was calculated as follows:

$$NRRI = OI_{recycled manure} * I 00/OI_{manure}$$
 (13)

where  $Ol_{recycled\ manure}$  indicates manure-N recycled to crop production and  $Ol_{manure}$  indicates total manure-N production.

Finally, the NRR of the agrifood system (NRRs) was defined as follows:

NRRs = 
$$(Oc_{recycled\ residue} + Ol_{recycled\ manure} + Oh_{recycled\ excreta})*100/(Oc_{residue} - Oc_{residue} \text{ for feeding} + Ol_{manure} + Oh_{excreta} + Oh_{food\ losses})$$
 (14)

where Oh<sub>recycled excreta</sub> indicates human excreta-N recycled; Oh<sub>excreta</sub> means total output of human excreta-N.

#### 3.2.3.4. N flows and agrifood system evaluation at county level

County level N flows were quantified as the sum of each village type multiplied by the number of villages within each type minus the internal flows between the four village types within Quzhou (Figure 3.1). The flows between villages were estimated based on our year-round farm and household survey (see section 3.2.5). The evaluation of N circularity at the county level was based on the same KPIs as used for the village level (see 3.2.3.3: N import, N loss, NUE and NRR).

#### 3.2.4. Scenario design towards circular N use in Ouzhou

Better management could increase crop and livestock production and reduce N losses (Li et al., 2017; Yang, 2019; Liu et al., 2020); healthy diets with less livestock protein, less carbohydrate and more plant protein have the potential to support sustainable circular agrifood systems, since plant protein production is more efficient in terms of energy and nutrients (Willett et al., 2019; Sheng et al., 2021). Based on these studies, we designed five scenarios towards achieving N circularity and evaluated their performance based on KPIs (see 3.2.3.3). The parameters used in these scenarios are based on literature review of local surveys and experimental data to ensure feasibility.

Good management practices (GMP). This scenario explored how N flows in the agrifood system can be made more efficient by good management practices in crop production (optimal N application, organic substitution of chemical N fertilizer, enhanced-efficiency fertilizer, which may reduce total N fertilizer input by 14-50% and increase crop yield by 4-38%, depending on crop species), and in livestock production (better feed conversion ratio to reduce feed input by 1-68% depending on livestock species). In addition, this scenario includes "good practices" in household consumption, i.e., a dietary change to reduce the intake of livestock protein as recommended by the EAT-Lancet Commission (Willett et al., 2019). All reduction and conversion parameters for this scenario were taken from the literature (Tables S4, S8 and S9).

Combination of GMP plus crop & livestock waste recycling (GMP+WR1). In this scenario, crop waste (straw, residues) and livestock waste (manure) are recycled and treated as organic resource. Residue disposal is avoided, and all crop residues are returned to the field within villages. All livestock manure (except for unavoidable losses) is recycled, i.e., applied to cropland within the same village or in another village within Quzhou if the source village does not have enough cropland to apply the produced manures. In line with recent policy, livestock manure is not discharged at the storage stage, and NH<sub>3</sub> emission from the manure management chain is mitigated by improved management practices such as solid-liquid separation and use of biofilters (Table S10).

Combination of GMP+WR1 plus household waste recycling (GMP+WR2). In addition to recycling waste from crop and livestock production, this scenario also recycles waste from households. Human excreta discharge and leaching losses at the storage stage are avoided by replacing dry latrines with septic tanks or other clean toilets, as promoted by recent local policy. All excreta are applied to cropland to substitute chemical fertilizer, and all kitchen residues and food waste are used to feed livestock to substitute concentrated feed.

Combination of GMP+WR2 plus switching to legumes (GMP+WR+STL). If residents switch to the EAT-Lancet diet as assumed in the three scenarios described above, the demand for plant-based food, legumes in particular, will increase. Ideally, this demand is met with locally grown food, rather than imported food, to support N circularity within Quzhou. Thus, in this scenario, soybean and other legumes are grown to substitute cotton or are planted as understory in orchards, to provide plant protein to the villages and reduce food import from outside Quzhou. Another benefit is that soybean and other legumes require less N fertilizer than cotton (60 compared to 210 kg N ha<sup>-1</sup> yr<sup>-1</sup>) to maintain the optimal yield of 2.7 Mg ha<sup>-1</sup> (Zhang et al., 2009).

Combination of GMP+WR plus reducing livestock breeding (GMP+WR+STL+RL). As assumed in the four scenarios described above, the demand for livestock products will decrease if residents switch to the Lancet EAT diet. Ideally, livestock breeding is reduced to adapt to the healthy diet. To avoid over production and reduce feed imports and livestock exports, all livestock species are reduced by 87% in this scenario.

#### 3.2.5. Data source.

Villages were grouped based on data in the Quzhou Statistical Yearbook 2017. Data on crop-specific application rates of chemical and organic N fertilizer, and N input from crop residue and seeds were collected in a survey among 1308 crop farmers throughout Quzhou (Table S2). Nitrogen inputs from deposition, BNF, and irrigation were based on a literature review (Table S3). Cropland N losses were from the parameters in the NUFER model and our literature review, which were calculated with specific loss ratios based on soil type, crop type, fertilizer type, N surplus and field management conditions (Tables S5-S7). The amount of discarded crop residues was based on data from our farm survey. Data on the feed composition and feeding rates for different livestock species were collected from a survey among 94 livestock farmers throughout Quzhou (Figure S1). Nitrogen losses from livestock production were calculated by multiplying a specific loss factor with the total amount of livestock manure N produced. The N loss factors in the manure management chain were taken from Bai et al. (2016). The amount and types of food consumed, kitchen residues, food waste, inter-village flows of crop and livestock products and manure were collected in a year-round survey (Table S4). In

this survey, we selected a total of ten households from the four village types, observed and recorded what these households bought, ate and wasted per meal for nine days each in December, April, July, and October 2019-2020. We collected kitchen residues from the surveyed households every three days during each 9-day observation period. The collected samples were used to measure their N content. The amount of human excreta was estimated based on mass balance (Supplementary Material).

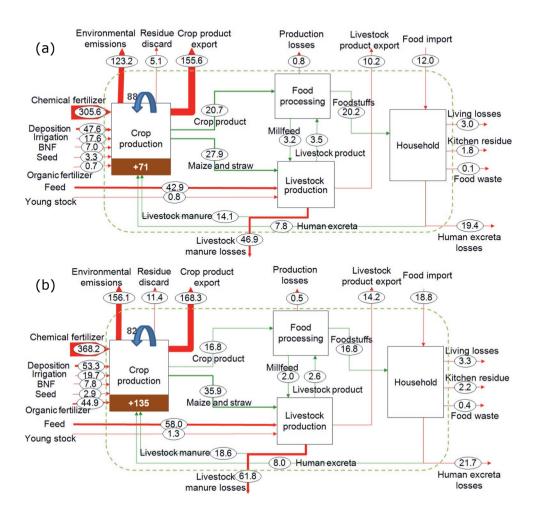
#### 3.3. Results

#### 3.3.1. Village-level N flows

Total N input at the village level, including all compartments of the agrifood system, were 439, 575, 1097 and 1024 kg N ha<sup>-1</sup> yr<sup>-1</sup> for cereal, cash crop, livestock, and land-limited villages, respectively (Figure 3.2). The highest N input was observed in livestock villages (Figure 3.2c), due to their substantial import of feed (652 kg N ha<sup>-1</sup> yr<sup>-1</sup>, compared to 43, 58 and 399 kg N ha<sup>-1</sup> yr<sup>-1</sup> in the cereal, cash crop, and land-limited villages, respectively). In all village types, chemical fertilizer input contributed substantially to total N input, amounting to 306, 368, 335 and 429 kg N ha<sup>-1</sup> yr<sup>-1</sup> in cereal, cash crop, livestock, and land-limited villages, respectively. N input through organic fertilizer, consisting of imported livestock manure and biofertilizer (microbial and humic fertilizers), was 45 kg N ha<sup>-1</sup> yr<sup>-1</sup> in cash crop villages, while this rate was less than 2 kg N ha<sup>-1</sup> yr<sup>-1</sup> in cereal, livestock, and land-limited villages. However, the latter two village types recycled considerable amounts of locally produced livestock manure and human excreta.

N output in the form of crop and livestock products (including crop residues for feeding) was 218, 238, 404 and 370 kg N ha<sup>-1</sup> yr<sup>-1</sup> in cereal, cash crop, livestock and land-limited villages, respectively (Figure 3.2). Most of the produced crop and livestock products were exported out of the villages. A large proportion of the households' food was imported, i.e., 37, 53, 40 and 99 % respectively, for the four village types.

The amount of N lost to the environment was 193, 243, 632 and 595 kg N ha<sup>-1</sup> yr<sup>-1</sup> in cereal, cash crop, livestock and land-limited villages, respectively (Figure 3.2). The high N losses in livestock and land-limited villages was mainly due to substantial losses from livestock manure (464 and 330 kg N ha<sup>-1</sup> yr<sup>-1</sup>, compared to 47 and 62 kg N ha<sup>-1</sup> yr<sup>-1</sup> in cereal and cash crop villages). N loss from human excreta was highest in land-limited villages. N loss through food waste and kitchen residue accounted for only a small percentage of the total N consumed by households (5.9-10%). Finally, the amount of N accumulating in cropland was 71, 135, 92 and 164 kg N ha<sup>-1</sup> yr<sup>-1</sup> for the four village types. At the county scale, this translated into an average N accumulation rate of 101 kg N ha<sup>-1</sup> yr<sup>-1</sup> across Quzhou.



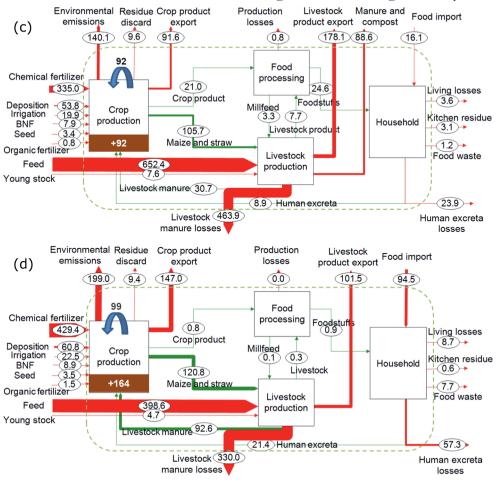


Figure 3.2. Nitrogen flows (kg N ha<sup>-1</sup> yr<sup>-1</sup>) at cereal (a), cash crop (b), livestock (c) and land-limited villages (d) in Quzhou. The numbers in the brown boxes indicate N accumulation (kg N ha<sup>-1</sup> yr<sup>-1</sup>) in cropland; the dashed green line represents the boundary of the villages; the green and red arrows indicate N flows within and across village type boundaries, respectively; the thickness of the arrows is proportional to their value; the blue arrows on the crop production compartment indicate straw returning.

When analyzing the KPIs of the different parts of the agrifood system at village level (Table 3.2), we observed that, in the crop production compartment, both N input and N loss (kg N ha-1 yr-1) were highest in the land-limited villages. The NUE of crop production ranged from 42% in cash crop and land-limited villages to 51% in cereal villages, while the NRR ranged from 88% (cash crop villages) to 95% (cereal villages). In the livestock production compartment, both N input and N loss were highest in the livestock villages. Compared to crop production, livestock production had a lower NUE

(18, 17, 24 and 19% in the four village types, respectively) and a much lower NRR (20% on average).

At the village type level, the KPI of total N import (N input minus N from deposition, biological N fixation and irrigation) clearly differed between the different village types, ranging from 365 kg N ha<sup>-1</sup> yr<sup>-1</sup> in cereal villages to 1015 kg N ha<sup>-1</sup> yr<sup>-1</sup> in livestock villages (Table 3.2). Total N loss was also highest in livestock villages, i.e., 646 kg N ha<sup>-1</sup> yr<sup>-1</sup>, which was almost three times the figure for cereal (200 kg N ha<sup>-1</sup> yr<sup>-1</sup>) and cash crop villages (257 kg N ha<sup>-1</sup> yr<sup>-1</sup>) (Table 3.2). The NRR in the different village types was inversely related to their N loss, with cereal villages showing the highest NRR and livestock villages the lowest. Cereal villages also showed the highest NUE of the agrifood system (41%), compared to 32% and 26% in cash crop and livestock villages, and as little as 17% in land-limited villages (Table 3.2). The villages rank in circularity performance as follows: cereal village > cash crop village > livestock village > land-limited village, as measured by the four KPIs.

Table 3.2. KPIs of N circularity, calculated for different village types of Quzhou, at the level of crop production, livestock production, and the agrifood system.

Compartme	Indicators	Cereal	Cash crop	Livestock	Land-limited
nt		village	village	village	village
Crop	N input (kg N ha <sup>-1</sup> yr <sup>-1</sup> )	404	523	460	641
production	N loss (kg N ha <sup>-1</sup> yr <sup>-1</sup> )	128	168	150	208
	NUE (%)	51	42	47	42
	NRR (%)	95	88	91	91
Livestock	N input (kg N ha <sup>-1</sup> yr <sup>-1</sup> )	75	97	769	524
production	N loss (kg N ha <sup>-1</sup> yr <sup>-1</sup> )	47	62	464	330
	NUE (%)	18	17	24	19
	NRR (%)	23	23	20	22
Agrifood system	N import (kg N ha <sup>-1</sup> yr <sup>-1</sup> )	365	494	1015	932
	N loss (kg N ha <sup>-1</sup> yr <sup>-1</sup> )	200	257	646	613
	NUE (%)	41	32	26	17
	NRR (%)	60	53	31	34

NUE and NRR indicate N use efficiency and N recycling rate, respectively.

#### 3.3.2. N flows between village types

All four village types in Quzhou were connected through N flows (Figure 3.3). The largest N flows were in the form of maize (25 kg N ha<sup>-1</sup> yr<sup>-1</sup> from cereal to livestock villages) and manure (11 kg N ha<sup>-1</sup> yr<sup>-1</sup> from livestock to cash crop villages). Cereal

villages were the largest supplier of N to the other villages (30 kg N ha<sup>-1</sup> yr<sup>-1</sup> of maize and cereal); in turn, they imported food from cash crop villages (0.3 kg N ha<sup>-1</sup> yr<sup>-1</sup> of fruits and vegetables) and livestock villages (1.3 kg N ha<sup>-1</sup> yr<sup>-1</sup> of livestock products). Livestock villages exported livestock manure to cash crop villages, while importing fruit and vegetables from them (0.4 kg N ha<sup>-1</sup> yr<sup>-1</sup>). Land-limited villages were the only village type that did not export any N to other villages; they imported 4.9 kg N ha<sup>-1</sup> yr<sup>-1</sup> of grains, fruits, vegetables, and livestock products for household consumption, in addition to 1.3 kg N ha<sup>-1</sup> yr<sup>-1</sup> of maize for livestock feeding.

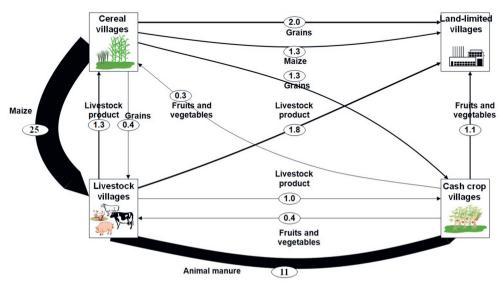


Figure 3.3. Nitrogen flows (kg N ha-1 yr-1) between different village types in Quzhou

# 3.3.3. County-level N flows

At the county level, total N input was 624 kg N ha<sup>-1</sup> yr<sup>-1</sup>, mostly in the form of chemical fertilizer (54%) and livestock feed (30%, Figure 3.4). This N was not efficiently used: the NUE of the county-level agrifood system was only 29%, due to large gaseous N losses (139 kg N ha<sup>-1</sup> yr<sup>-1</sup>), N losses through leaching, runoff and erosion (81 kg N ha<sup>-1</sup> yr<sup>-1</sup>) and direct discharge of livestock manure and human excreta (119 kg N ha<sup>-1</sup> yr<sup>-1</sup>). The average N accumulation in cropland was 101 kg N ha<sup>-1</sup> yr<sup>-1</sup>, with land-limited villages accumulating the most (164 kg N ha<sup>-1</sup> yr<sup>-1</sup>). The amount of N lost via kitchen residue and food waste was only 4 kg N ha<sup>-1</sup> yr<sup>-1</sup>. The sum of N loss and N accumulation (452 kg N ha<sup>-1</sup> yr<sup>-1</sup>) exceeded the N input by chemical fertilizer (339 kg N ha<sup>-1</sup> yr<sup>-1</sup>).

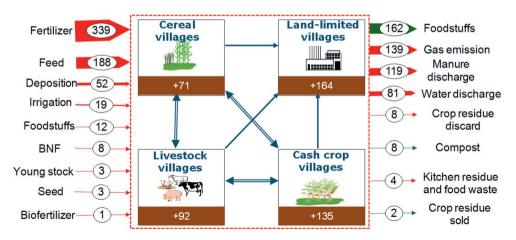


Figure 3.4. County-level N flows (kg N ha<sup>-1</sup> yr<sup>-1</sup>) in Quzhou. Values in the brown boxes indicate N accumulation in cropland. The dashed red line represents the boundary of the county. Arrows on the left and right side are input and output flows, respectively, with green arrows indicating N output in marketable products.

#### 3.3.4. Analysis of scenarios towards N circularity

To explore the extent to which county-level agrifood systems can achieve N circularity, we evaluated five scenarios, using N import, N loss, NUE and NRR as KPIs (Figure 3.5 and S3-S7). The first scenario (GMP), which included good management practices to reduce nutrient input in crop and livestock production (Tables S8-S10) and reduce household consumption of livestock protein (Table S4), decreased N import from 546 to 380 kg N ha<sup>-1</sup> yr<sup>-1</sup> and N losses from 350 to 217 kg N ha-1 yr-1. These gains translated into an increase in NUE from 29 to 47% but had limited impact on NRR. The second scenario (GMP+WR1), which included the above measures plus improved recycling of crop residues and livestock manure to cropland within and between villages, further decreased N import to 272 kg N ha<sup>-1</sup> yr<sup>-1</sup> and N loss to 131 kg N ha-1 yr-1. This not only resulted in a further increase of the system's NUE (to 61%), but also dramatically raised the NRR from 55 to 75%. When extending the recycling efforts to kitchen waste and human excreta (third scenario, GMP+WR2), the four KPIs of N import, N loss, NUE and NRR improved even further, to 242 kg N ha<sup>-1</sup> yr<sup>-1</sup>, 107 kg N ha<sup>-1</sup> yr<sup>-1</sup>, 66% and 87%, respectively. The fourth scenario (GMP+WR+STL), which combined previous measures with growing more legumes further improved NUE to 70% and further reduced N import to 214 kg N ha-1 yr-1, but had no additional effect on N loss and NRR. Involving legumes in the cropping system also reduced N accumulation in cropland by >70%, compared to GMP+WR2 (Figure S5 and S6). Compared to S0 (Figure 3.4), the GMP+WR+STL

scenario reduced N accumulation in cropland by as much as 98%, from 101 to 2.2 kg N ha<sup>-1</sup> yr<sup>-1</sup> (compare Figure 3.4 and Figure S6). Finally, the fifth scenario (GMP+WR+STL+RL), which combined all previous measures with reducing livestock improved all the four KPIs, i.e. N import (-37 kg N ha<sup>-1</sup> yr<sup>-1</sup>); N loss (-27 kg N ha<sup>-1</sup> yr<sup>-1</sup>); NUE (+9%); NRR (+5%). Compared to S0, the GMP+WR+STL+RL scenario roughly doubled the agrifood system's NUE and NRR while reducing N import and N loss by 68% and 77% (compare Figure 3.4 and Figure S7).

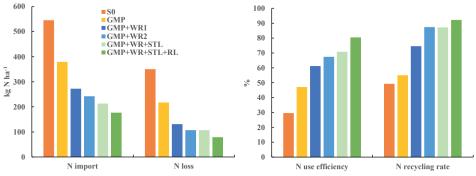


Figure 3.5. KPIs of N circularity in Quzhou under different scenarios. S0: baseline; GMP: good management practices; GMP+WR1: same as previous, plus improved recycling of waste from crop and livestock production; GMP+WR2: same as previous, plus improved recycling of household waste; GMP+WR+STL: same as previous, plus switching to legumes; GMP+WR+STL+RL: same as previous, plus reducing livestock breeding.

#### 3.4. Discussion

The results of our case study in Quzhou, China, show that the current agrifood system of Quzhou is generally linear, as measured by the four selected KPIs (N import, N loss, NUE and NRR). Our scenario analysis shows that integrated measures towards circular N use at village and county level could roughly double the county-level agrifood system's NUE and NRR while reducing N import and N loss by more than 60%, and N accumulation in soil by 99%.

#### 3.4.1. N circularity in Quzhou's agrifood system

Our findings show that the high N losses from cropland and livestock manure management are key limitations for circular N use in Quzhou's agrifood system despite variation among village types (Figure 3.2 and 4), which is consistent with previous research (Luo et al., 2018; Wang et al., 2020). In cereal and cash crop villages, N loss was mainly through environmental emissions from crop production, while N loss in livestock and land-limited villages was mainly through losses from the manure management chain (Figure 3.2). The substantial differences observed among village types prove the necessity to analyze N flows not only at county level but also at village

level. However, although villages are easy to govern and they can readily adopt technical recommendations, they are too small to achieve N circularity on their own. For example, Quzhou's land-limited villages accumulated 164 kg N ha<sup>-1</sup> yr<sup>-1</sup> in their cropland (Figure 3.2), which increases the risk of leaching and gaseous losses in the long term. These villages are unlikely to solve this problem without exporting N to areas needing N input, such as Quzhou's cereal and cash crop villages. In the latter two, the current share of organic fertilizer was low (6.8% and 16% of total fertilizer input) compared to the recommended rate of 50-75% (Xia *et al.*, 2017; Wei *et al.*, 2021). If all livestock manure and human excreta produced in Quhzou were recycled as organic fertilizer for cropland within and among Quzhou's villages, this would realize a rate of roughly 55%, showing the necessity to bridge the gap between village and county level N cycles.

Nitrogen use efficiencies differed between compartments of the agrifood system, affecting system performance. The NUE of the crop production compartment ranged from 42 to 51% across village types (Table 3.2), which is higher than values reported within China (28-44%) (Ma et al., 2010; Chen et al., 2016; Duan et al., 2021), but lower than the value in Europe (56%) (Billen et al., 2021). The higher values than other studies in China are most likely due to a difference in calculation method; in the ratio between crop product N output and total N input, we not only included the N output in grain, fruit and vegetables, but also in crop residues for feeding because of the latter's market value. At the system level, NUE differed strongly among village types, with land-limited villages achieving less than half the NUE of cereal villages (Table 3.2). This is mainly because land-limited villages have to import most of their feed and foodstuffs, and keep more livestock (which is N inefficient) than cereal villages (Table 3.1). Village-level differences in N loss and NRR were caused by similar reasons. At the county level, the NUE of the agrifood system (29%) was higher than the values reported for other areas in China and Europe (13-23%) (Ma et al., 2010; Billen et al., 2021). This is likely due to the fact that Quzhou is an export-oriented county, with most of its crop and livestock products marketed elsewhere. We did not consider N losses from feed production and food processing and consumption outside Quzhou. In other words, the food chain we considered was relatively short. A shorter food chain means less potential to loss and is generally characterized by a higher system NUE, but does not account for losses beyond the food chain (Quemada et al., 2020).

This study conducted scenario analysis by adding each scenario one more package of optimized practices. In this approach, we can explore the potential towards N circularity while identify the contribution of different practices and their interactions. Among our scenarios, the most complete package of measures (GMP+WR+STL+RL) increased the county-level system's NUE from 29 to 80%. Forty percent of this gain was achieved with improved recycling of organic waste (Figure 3.5), demonstrating that waste

recycling is an efficient strategy towards circular N use in Quzhou. This finding supports current Chinese policies to promote organic waste utilization (MOA, 2015, 2017), eliminate food waste (Anti-Food Waste Law, NPC, 2021), and encourages scientific research to recycle organic resources (Zhang et al., 2019). Among the different sources of organic waste in our study area, recycling of crop residues and livestock manure to local cropland had the greatest effect, increasing NUE from 47 to 61% (Figure 3.5). Unlike our expectation, recycling of human excreta and kitchen waste made a modest contribution (increasing NUE from 61 to 67%). A likely explanation is that Quzhou is an agricultural county, with cultivated land accounting for 78% of the total administrative area (NBSC, 2018), where the effects of households on system N flows are small compared to the effects of agricultural production. Previous studies also showed that recycling of human excreta improved N circularity only in regions with high population densities (Koppelmäki et al., 2021).

Scenario analysis further showed that the application of good management practices allowed the greatest reduction of N import and N loss (Figure 3.5). Among these practices, optimizing fertilizer application and substituting organic for chemical N fertilizer in crop production are well-studied (Cui et al., 2018). These practices synchronize N supply and crop N demand, which reduces excessive N fertilizer input and the subsequent N losses. Higher feed conversion ratio for livestock means less feed intake for the same amount of meat production. Improving feed conversion ratio is complicated, as it is not only related to feed composition but also to livestock genetics and environmental aspects, such as the livestock's thermal environment (Patience et al., 2015; Sell-Kubiak et al., 2017). Switching from keeping ruminants to monogastric livestock with high feed efficiency is also a potential strategy to improve the NUE of livestock production, but may exacerbate food-feed competition (Muscat et al., 2020) and therefore was not included in the work. Diet change to more plant and less livestock protein made a limited contribution to N circularity in Quzhou. The main reason is that livestock production was not reduced, to ensure food and nutrition security of foodscarce regions surrounding Quzhou. In addition, local meat consumption was already relatively low (Table S4). Finally, since legumes were not a common crop in Quzhou, a diet with more plant protein/legumes meant that these foods had to be imported (compare Figure 3.4 and Figure S3).

The need to import plant-based protein can be solved by introducing soybean and other legumes into Quzhou's plant production system, as investigated in the GMP+WR+STL scenario (Figure 3.5). For N circularity, including legumes in the cropping system indeed reduced N import and increased NUE, although the effect on N loss and NRR was minor (Figure 3.5). More importantly, substituting legumes for cotton in the cropping system significantly reduced N accumulation in cropland, because legume crops require

less N input and take up more N than the cotton crops they replaced (compare Figure S5 and S6). This finding supports that legumes should play an important role in strategies towards achieving circular agrifood systems (Vanlauwe *et al.*, 2019; Billen *et al.*, 2021), as they help to reduce chemical fertilizer import through biological N fixation and produce plant protein that can substitute emission-intensive meat protein.

The need to adapt livestock breeding to healthy diet can be achieved by reducing livestock density in Quzhou, as investigated in the GMP+WR+STL+RL scenario (Figure 3.5). Reducing livestock breeding means less feed imports and less livestock products exports. As inefficient livestock manure management is a key bottleneck for achieving N circularity, the GMP+WR+STL+RL scenario further reduced N loss while increasing the NUE (Figure 3.5). However, reducing livestock also means less available animal manure for crop production. Therefore, more chemical fertilizer needed to be imported to maintain the optimal fertilizer input as the GMP+WR+STL scenario, and it increased from 114 to 165 kg N ha<sup>-1</sup> yr<sup>-1</sup> (Figure S6 and Figure S7). Additionally, reducing livestock exports may threat food security of the food-scarce areas around the export-oriented county of Quzhou. Exporting regions such as Quzhou illustrate the delicate relationship between N circularity and food trade. Full circularity is infeasible and how to balance circularity and trade at appropriate scale requires further research.

The last two scenarios show that a reduction in livestock and increase in legumes to adapt to a healthy diet is a good lever to address circularity. However, there could be scenarios that perform even better as this study results in a discrete set of options. The use of optimization algorithms combined with scenario analysis may find the optimal crop-livestock configuration that adapts to healthy diet with best performance in KPIs (Seppelt *et al.*, 2013).

# 3.4.2. Incentivizing and accelerating the transition towards circular N use in agrifood systems

The results of our scenario analysis demonstrate the technical potential of integrated measures to transition towards N circularity. However, true change will not take place if the negative effects of these technologies are not solved (such as the health concerns of manure recycling), or if technologies are not diffused to and accepted by all stakeholders within the agrifood system. To accelerate the transition, we need widespread knowledge transfer and technology extension services, such as the Science and Technology Backyard (STB) platform in China (Zhang et al., 2016). This STB involves agronomists living in villages among farmers, connects the scientific community with the farming community to facilitate information exchange and innovation, and serves as a hub for government and agri-businesses to engage and improve their services.

# Circular nitrogen use in the agrifood system

Farmers will be more likely to adopt technologies supporting N circularity if there is compelling evidence that the new technologies are more profitable than the previous ones. Based on our literature review, agricultural production can indeed be improved by good management practices such as using enhanced efficiency fertilizer in crop production (Table S8) and improving feed conversion ratios in livestock production (Table S9). However, these practices generally require additional labor and/or capital input, which is a challenge for capital-starved farmers. Thus, to enable the transition towards circular agrifood systems, governments should provide financial incentives to shift profitability in favor of circular agricultural practices. Based on the experience that organic certification and labeling programs have promoted the development of the organic food industry (Kuchler *et al.*, 2017; Verburg *et al.*, 2022), policymakers could similarly establish certification standards and labelling for circular production practices, to promote investment in circular technologies and their adoption by farmers.

The transition towards N circularity can be further accelerated by creating synergies between N circularity and other policy goals, such as carbon sequestration, water management and biodiversity conservation. For example, adopting good management practices that reduce external imports can contribute to farm financial sustainability; recycling of organic resources to cropland can increase long-term carbon sequestration and enhance water retention and food production by improving soil quality (Maillard and Angers, 2014); advanced human excreta management can reduce the amount of water needed for toilet flushing (Wang *et al.*, 2019); and diversifying cropping systems can promote biodiversity conservation, particularly through inclusion of perennial legumes (Renard and Tilman, 2019; Zabel *et al.*, 2019). These synergies/potential tradeoffs between N circularity and other policy goals deserve further research.

# 3.4.3. Limitations of this study

In our study, analysis of N flows in the food processing compartment was limited to maize and wheat milling due to a lack of other food processing data. This may have resulted in an underestimation of N losses and overestimation of system NUE. In addition, we predicted considerable amounts of cropland N accumulation because not all the applied N can be absorbed by crops or lost to the environment in one year. This approach may underestimate the N losses on a multi-year basis. Furthermore, our selection of KPIs to evaluate system N circularity did not include any indicator related to agricultural yields and economic performance. Such indicators should be included in future research, to ensure that the proposed measures are realistic in terms of food security and feasible in terms of economic performance. Finally, in our scenario of switching to legumes, we assumed that legumes would substitute cotton. Such a change

will have an impact on fabric production and the clothing sector, which was beyond the scope of our research.

### 3.5. Conclusions

This study analyzed N flows in an agrifood system, considering not only crop and livestock production but also detailed household data. We selected four KPIs in line with the principles of circular agriculture to evaluate N circularity of the agrifood system and applied these KPIs to a case study bridging the gap between village and county level N cycle in Quzhou, China. The KPIs in this study enable transparency and comparability among different spatial regions and over time for measuring year-to-year progresses.

The Quzhou's agrifood system is representative of many systems worldwide, with N flows being far from circular due to the large external inputs and high environmental losses. Excessive inputs, inefficient agricultural production and livestock manure management are key bottlenecks for achieving circular N use. The villages rank in circularity performance as follows: cereal village > cash crop village > livestock village > land-limited village. Our scenario analysis demonstrates that recycling of organic waste is one of the most effective and feasible measures to move the agrifood system towards N circularity. This finding provides evidence to support further development of strategies and policy incentives to promote waste recycling. Accelerating the transition towards circular agrifood systems will require novel technology transfer services and institutional, behavioral, policy and market incentives to promote diffusion and acceptance of technologies by all stakeholders. Balancing the relationship between N circularity and agricultural trade to achieve sustainability at different scales will be the greatest challenge.

# **Acknowledgements**

We are grateful to Oene Oenema and Mart Ros for providing some research ideas, to Christien Ettema for her thorough editing of the manuscript, and to Lin Ma for authorizing our use of the NUFER model. This study was funded by the Sino-Dutch Agriculture Green Development Project of China Scholarship Council [No. 201913043] and Hainan University.

# **Supporting Information**

Supplementary information associated with this article can be found in the online version, at https://doi.org/10.1016/j.agsy.2023.103683

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Synergies in water security and nitrogen circularity: a case study in Quzhou, China

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#### **Abstract**

Water security, including both water quantity and water quality, is affected by nitrogen (N) cycles. Few studies, however, have integrated the water and N cycles to explore the potential impact of current water use and pollution on future water security. To fill this gap, we quantified the temporal dynamics of the water balance and N pollution in Quzhou, China, and evaluated the interactions between the water and N cycles. as well as their synergies. We found that despite a rise in water consumption, from 388 MCM in 2010 to 460 MCM in 2020, Quzhou has halted and even reversed its declining groundwater level through measures such as water transfer, improved use efficiency, and reduced groundwater extraction. A caveat, however, is that water transfer might externalize water scarcity to water-exporting regions. Regarding water quality, we found pollutants in groundwater to be generally below critical values, while NH<sub>4</sub>-N in surface water exceeded the safety threshold – although its concentration had decreased by 87% over the past two decades, according to our literature review. The downward trend of NH<sub>4</sub>-N concentrations in surface water could be attributed to reductions in drainage water with pollutants, thanks to more efficient agricultural water use. According to the scenario analysis, improved N management practices, such as optimized fertilization, could reduce N discharge from agrifood systems from 10.455 Mg N yr<sup>-1</sup> to 6.792 Mg N yr<sup>-1</sup>, decreasing the NH<sub>4</sub>-N concentration in surface water from 1.2 mg N l<sup>-1</sup> to 0.97 mg N l-1, which is below the critical threshold (1.0 mg N l-1). N could be further reduced, to 0.69 mg N l-1, by implementing other practices that promote circular N use, such as recycling organic waste and reducing livestock production. This study illustrates the possibility of achieving synergies between water security and N circularity.

#### 4.1. Introduction

Water scarcity threatens agricultural production, ecosystem services, and the sustainable development of human societies. The problem is made worse by climate change and increasing water consumption due to growing populations (FAO, 2021; Scanlon *et al.*, 2023). Already, two-thirds of the global population (4 billion people) suffers from severe water scarcity for at least one month every year (Mekonnen and Hoekstra, 2016). By 2050, global water consumption is expected to be 20–30% higher than the current level (WWAP, 2019; FAO, 2021). Climate change, too, brings increasing frequency of extreme weather events, such as droughts and floods. These exacerbate spatial and temporal mismatches between water resources and demand, making it more difficult for many societies to secure reliable water supplies (Pokhrel *et al.*, 2021; Scanlon *et al.*, 2023).

Water is a reoccurring theme in the United Nations' Agenda for Sustainable Development. Clean water and sanitation are integral to achieving 62 of the 163 targets, particularly for achieving Goal 3 on health and well-being, Goal 14 on life below water, and Goal 15 on life on land (Irannezhad et al., 2022). Water's central role in human development and sustainability has given rise to numerous global and regional initiatives, which are often presented in terms of water security (Qi et al., 2020; Hoek van Dijke et al., 2022; Irannezhad et al., 2022). Water security is defined as the capacity of humans to continuously access sufficient quantities of water of sufficient quality to maintain their livelihoods and development while sustaining freshwater resources (Irannezhad et al., 2022). The pursuit of water security thus considers both water quantity and quality aspects.

Strategies for securing adequate quantities of water encompass both surface water and groundwater and aim at improving water use efficiency, reducing water demand, and transferring water to where it is needed. Improving water use efficiency in agriculture, for example, can help save water and reconcile water competition between agriculture, industry, and the energy sector (Zhao et al., 2015; Flörke et al., 2018). However, nominal "savings" achieved through efficiency gains in one sector may overstate actual impacts on water supply, because water that is not consumed at one site is frequently discharged into surface water or goes to recharge groundwater, from where it is subsequently recovered and used elsewhere (Perry et al., 2009; Aeschbach-Hertig and Gleeson, 2012; Grafton et al., 2018). Therefore, to determine whether gains in water use efficiency really do conserve water at a larger scale requires a comprehensive accounting of water flows, including changes in surface water and groundwater storage.

Strategies to reduce water demand often involve irrigation, as irrigation accounts for 70% of global water withdrawals and 90% of water consumption (Poore and Nemecek,

2018; FAO, 2021). To reduce irrigation water consumption, various approaches have been found to be effective. Among these are switching to less water-intensive crops, meaning those that consume less water; leaving land fallow in drought seasons or years (Deng et al., 2021; Scanlon et al., 2023); relocating crop production from semiarid to more humid regions in order to leverage precipitation and reduce water extraction (Zhao et al., 2021; Wang et al., 2022; Scanlon et al., 2023); and minimizing nonbeneficial evaporation by implementing measures such as deficit irrigation, mulching, and targeted nutrient management (Chukalla et al., 2015; Grafton et al., 2018; Scanlon et al., 2023).

Water transfer is another strategy that can be applied to increase the quantity of local water available. Water transfer refers to either the physical redistribution of water through infrastructure projects or virtual water redistribution through traded products (Zhao et al., 2015). The world's largest water diversion project is the South–North Water Transfer Project, which China is implementing to divert water from the humid south to the semiarid north of the country. Nonetheless, transferring water brings risks, such as inducing water stress in the donor/exporting areas and increasing water pollution and salinization (Zhao et al., 2015; Zhuang, 2016; Long et al., 2020).

Beyond measures to increase supply quantities, water quality has become a global concern. Water pollution and deterioration are now widespread, with agrifood systems being the primary contributor (FAO, 2021; Van Vliet *et al.*, 2021). In China, Yu *et al.* (2019) found that anthropogenic nitrogen (N) discharge into freshwater was some 14.5 Tg per year (average 2010-2014). This is about 2.7 times the estimated "safe" discharge level. The European Union and United States are also experiencing degraded water quality, mainly due to agricultural runoff, and approximately half of their surface waters are rated poor due to high concentration of N and in some case phosphorus (P) (Scanlon *et al.*, 2023). A range of pollutants make water unfit for consumption and pose risks to human health (Irannezhad *et al.*, 2022). Worldwide, some 1.8 billion people must rely on drinking water that is contaminated with fecal matter, which puts them at risk of cholera and other diseases (Scanlon *et al.*, 2023). When water quality is included in water scarcity assessments, the share of global population considering to be suffering severe water scarcity increases from 30% to 40% (Van Vliet *et al.*, 2021).

Technologies have long been recognized for addressing water quality and quantity issues. Desalination systems, for example, remove salts from seawater and saline inland surface waters. However, the cost of these systems is still prohibitive, and disposing of the resultant brine remains difficult, limiting large-scale applicability (Van Vliet et al., 2021; Irannezhad et al., 2022). Wastewater reuse is another promising technological development. Wastewater treatment systems offer the potential to alleviate water scarcity while simultaneously reducing pollutant discharge into surface waters. Again,

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however, practical applications remain constrained by high energy requirements and the high cost of equipment and infrastructure (Liao et al., 2021; Van Vliet et al., 2021). Moreover, due to the complexity of the water cycle and interactions, implementation of new technologies must be carefully weighed. Shifting irrigation regimes from flood to sprinkler and drip systems, for example, can increase irrigation efficiency and reduce the risk of discharged nutrients compromising water quality. However, in saline regions these systems may exacerbate salinity, as sufficient recharge is needed to flush the salts out of saline soils (Grafton et al., 2018; Scanlon et al., 2023). Another innovation, circular agriculture, is being explored to better manage nutrient and waste flows in agrifood systems, thus mitigating the environmental impacts such as nutrient discharge into water (Kirchherr et al., 2017; Velasco-Muñoz et al., 2021). It is vital to seek synergies and minimize trade-offs among these technologies and innovations.

Considering the dual imperatives of water security – the need to secure sufficient quantities of water while also ensuring water quality – as well as the sometimes unforeseen consequences of technology implementations, the current study sought to quantify interactions between water security (i.e., water quantity and quality) and N circularity. Nitrogen enters water supplies mainly through agricultural runoff and wastewater discharge. High levels of N in water lead to eutrophication, diminishing water's capacity to sustain life. We first quantified changes in the surface water and groundwater balance over time, accounting for precipitation, evapotranspiration, runoff to surface water, recharge to groundwater, and outflow. We then assessed water quantity and quality and interactions between the two. Finally, we explored the potential to achieve water security by integrating water and N cycles. The study was conducted in the Quzhou county, China. China is a hotspot of water scarcity, due to water quantity and quality issues driven by excessive N use in agriculture (Rodell et al., 2018; Van Vliet et al., 2021; Schulte-Uebbing et al., 2022). Quzhou is located in the North China Plain, where water pollution (Strokal et al., 2016) and N losses (Wang et al., 2018) are especially acute. However, as a demonstration county for the Chinese "Agricultural Green Development" strategy (MOA, 2020), Quzhou seeks to be a forerunner in producing nutritious agricultural products, while minimizing environmental impacts.

#### 4.2. Materials and Methods

#### 4.2.1. Research area

Quzhou county lies in Hebei province, North China Plain. It has a sub-humid, warm, temperate, continental monsoon climate with four distinct seasons. The mean annual temperature is 13°C, and annual precipitation is 556 mm, of which approximately 60% occurs during summer. The county covers an area of 677 km² and includes 10 towns and 342 village-size administrative units. It had a registered population of 527,000 in

2017, with over 90% of inhabitants residing in rural areas (NBSC, 2018). Forest and arable land cover 89 km² and 525 km² of the county, respectively. Agricultural production here is high input, high output, and high nutrient surplus. The main crops are wheat, maize, cotton, and vegetables. Some 80% of the arable land is irrigated. Two rivers run through the county. During periods of irrigation when surface water is scarce, groundwater is the main water source, not only for agriculture, but also for industry and domestic use. Groundwater accounted for 70% of the water supplied in the county before the local government restricted groundwater withdrawal in 2014 (DWRH, 2021). According to Xu *et al.* (2018), this has caused groundwater overexploitation and induced groundwater overdraft. Additionally, Yang (2019) reported that Quzhou's water supply was affected by severe salinization. To promote water security, several pilot projects have been launched since 2014, including introduction of highly efficient, water-saving irrigation techniques, seasonal land fallowing, and comprehensive control of groundwater overdraft (Xu *et al.*, 2018; Deng *et al.*, 2021).

#### 4.2.2. Elaboration of water security

Based on the definition by Irannezhad et al. (2022), we elaborated water security as encompassing two aspects: (1) a sufficient quantity of water to meet requirements and (2) sufficient water quality to avoid environmental deterioration and human health problems. To evaluate water quantity, we determined changes in water level in the case study county, assuming that for a secure water supply the amount of water recharge should be equal to or larger than discharge. To evaluate water quality, we compared water quality characteristics against set quality standards. Our objective with this simplified elaboration was to measure water security and determine year-to-year changes in water security in the case study area. The elaboration differs from the concept of water circularity, in that sources of water are beyond the scope of consideration. For example, water transferred to Quzhou from other regions was regarded as locally sustainable, and its larger scale impacts were not evaluated.

#### 4.2.3. Calculation of the water balance

We analyzed the water balance and interaction between surface water and groundwater in Quzhou from 2010 to 2020. For the surface water balance, we considered water inflows, outflows, runoff and return flows to groundwater (see section 4.2.3.1 for detail). Return flows to groundwater included river water infiltration and surplus water recharge from agricultural, domestic, and industrial uses. Such recharge water may contain dissolved N, in which case it could potentially degrade groundwater quality (Figure 4.1). Calculations of the groundwater balance include inflows, outflows, return flows, extraction, and discharge (from groundwater to surface water). However, this study considered only the balance between return flows and groundwater extraction, as

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groundwater inflows from, and outflows to, surrounding aquifers were difficult to measure. Moreover, we assumed that no water discharge occurred, because the groundwater level in Quzhou was relatively deep (12 m), resulting in a disconnection between groundwater and surface water. All flow items are in million cubic meters (MCM) unless otherwise stated.

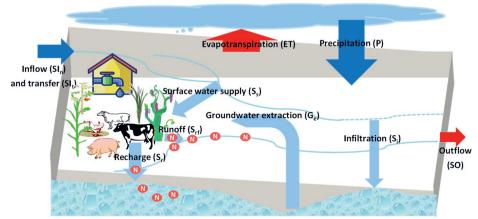


Figure 4.1. Schematic diagram of the water balance and nitrogen (N) discharge. Dark blue and red arrows indicate, respectively, input and output flows to and from the case study area. Light blue arrows are internal water flows within the case study area. Variables in parentheses represent the water flows elaborated in the balance equations in section 4.2.3.1.

#### 4.2.3.1. Surface water balance in Quzhou

The surface water balance reflects surface water supply and demand, including inflows, outflows and surplus. Surface water surplus ( $S_{SUP}$ ) was calculated as follows:

$$S_{sur} = S_s + G_e + P - ET \tag{1}$$

$$S_{\rm s} = SI - S_i - SO \tag{2}$$

$$SI = SI_n + SI_t \tag{3}$$

$$SO = SI * f_{io} \tag{4}$$

$$ET = \sum_{i=1}^{n} (ET_i[t] * A_i * 10 * T_i) * 10^{-6}$$
(5)

where  $S_s$  is the annual surface water supply for agricultural, domestic, and industrial uses;  $G_e$ , P, and ET represent annual groundwater extraction, precipitation, and evapotranspiration, respectively. SI is surface water inflow, made up of a natural flow component  $(SI_n)$ , specifically, river water, and water from transfer projects  $(SI_t)$ ;  $S_t$  is infiltrated river water; and SO is surface water outflow, calculated using the outflow fraction  $(f_{in})$ , which is a dimensionless factor estimated by taking the average value of all outflow fractions (0.72) in the Hebei Water Resources Bulletin for the years 2016-2020.

This means that 28% of the inflow is used in Quzhou. ET was calculated based on the Penman-Monteith method (Allen *et al.*, 1998). The subscript i is plant species in Quzhou, including various crops and forest cover, while n represents the number of plant species.  $ET_i[t]$  is the daily actual evapotranspiration (mm) for the specific species.  $A_i$  and  $T_i$  are, respectively, the planting area (ha) and crop-growing time (days). ET[t] is the actual evapotranspiration at time t, and is calculated as follows:

$$ET[t] = ET_0[t] * K_c[t] * K_s[t]$$
 (6)

where  $ET_0[t]$  is a daily reference evapotranspiration (mm), considering geographic location and climate, and K[I] is a crop coefficient that varies over different plant growth stages (initial, development, mid-season, and late season). Specifically, Kd/l remains constant during the initial growth stage (K[ini]) and the mid-season stage (K[mid]). During the development stage, it is assumed to linearly increase from  $K_{\epsilon}[mi]$  to  $K_{\epsilon}[mid]$ . For the late season stage, it is assumed to decrease linearly from  $K_0[mid]$  to  $K_0[mid]$ . Table S4.1 in the supplementary material presents specific crop planting dates and lengths of cropping seasons. The values of  $K_c$  for different plants and their growth stages were calculated and adjusted for the climate of Quzhou, following Allen et al. (1998) (Table S4.2). For non-growing periods (bare land),  $K_c$  was taken to be equal to  $K_c[ini]$ .  $K_s[t]$  is a dimensionless factor describing the effect of water stress on crop transpiration with a value between zero and one. As determining an accurate value for the actual rootzone water depletion requires many in situ measurements from different fields, this study used a simplified process: K<sub>s</sub> for irrigated crops was estimated using the values for surplus water  $(S_{sur})$  and infiltrated water  $(S_i)$  in a given year. When  $S_{sur}$  is positive, i.e., water supply was assumed to exceed demand, and water stress was considered not to occur ( $K_s = 1$ ). Slight stress ( $K_s = 0.9$ ) was expected when  $S_{sur}$  is negative while  $S_{sur} + S_i$ is positive, as in such a situation, river water infiltration could potentially counteract a water supply shortage. Moderate stress ( $K_s = 0.8$ ) is assumed when  $S_{sur} + S_i$  is negative. More severe water stress would not occur for irrigated crops due to timely irrigation, but it would occur for rainfed crops and forest and for croplands during non-growing periods. For these plants and periods, K, was assumed to be 0.5, as in Quzhou, plants may experience water scarcity in two of the four seasons (in spring and in autumn). Although water shortages are common in winter, most plants are dormant and water transpiration is extremely low. Our assumed values of  $K_s$  were indirectly justified by the fact that changes in water level (calculated using these values) compared well with measured data (see section 4.4.1 for detail).

#### 4.2.3.2. Groundwater balance

The groundwater balance ( $G_b$ ) reflects the change of groundwater level. A positive balance indicates increased groundwater level, and a negative balance indicates a decrease. It was calculated as follows:

$$G_h = S_i + S_r - G_e \tag{7}$$

where  $S_i$ , as above, is infiltrated river water;  $S_r$  is water recharge to groundwater from agricultural, domestic, and industrial uses.  $S_r$  was calculated based on  $S_{sur}$ , as  $S_{sur}$  in part recharges groundwater and in part discharges into surface water. The amount of groundwater recharge is low where artificial drainage is in use, as in that case, water flows directly to rivers through drainage canals. Following Döll et al. (2012), we calculated the recharge fraction of surplus water ( $f_{rg}$ ) based on the fraction of arable land that was artificially drained ( $f_{d,ir}$ ) as follows:

$$f_{rg} = 0.8 - 0.6 * f_{dir} ag{8}$$

Quzhou has an extremely small proportion of artificially drained land  $(f_{d,ir})$ , because the area is so dry that flooding seldom occurs. Thus, approximately three-quarters of surplus water went to recharge the groundwater.

 $G_b$  was converted to the change in groundwater level ( $GL_c$ ) as follows:

$$GL_C = (G_b/A_a)/0.1$$
 (9)

where  $A_a$  is the administrative area of Quzhou, which is 677 km<sup>2</sup>. The value 0.1 is the conversion fraction from  $CL_{\epsilon}$  (m) to groundwater storage (m) (Döll *et al.*, 2012), as groundwater can only be stored in soil pore spaces, which are limited in Quzhou's phreatic zone.

# 4.2.4. Water quality and potential for improvement

To acquire data on local water quality, we reviewed literature and consulted Quzhou's Water Conservancy Bureau. Table 4.1 presents the main pollutants that exceeded the set quality standards during the study period. The concentration of NO<sub>3</sub>-N was found to be within the defined safety boundaries in both surface water and groundwater during the entire study period (Yang, 2019), so it is not listed in the table and not analyzed further in this study. Quzhou's surface water did exhibit high NH<sub>4</sub>-N levels, despite some gradual improvement, and its groundwater exhibited high total hardness. Because the focus of our research was nutrient pollution, specifically N circularity, we continued our analysis focusing on the potential for reducing NH<sub>4</sub>-N in Quzhou's surface water.

Table 4.1. Main pollutants in surface water and groundwater in Ouzhou county, 2001-2017

	Surface	water	Groundwater					
	NH <sub>4</sub> -N (mg l <sup>-1</sup> )	COD (mg l <sup>-1</sup> )	NH <sub>4</sub> -N (mg l <sup>-1</sup> )	Total hardness (CaCO <sub>3</sub> mg I <sup>-1</sup> )	Reference			
Maximum limit value	1.0	20	0.5	450	PRC National Standard GB3838- 2002; GB/T 14848- 2017			
2001	9.5	*	*	*	Zhao (2013)			
2006	*	*	1.3	598	Zhao (2013)			
2010	7.5	*	0.8	763.5	Zhao (2013)			
2014	5.5	58	*	*	Jin (2017)			
2015	4.9	64	*	*	Jin (2017)			
2016	Above the limit	Below the limit	Below the limit	Above the limit	Yang (2019)			
2017	1.2	*	Below the limit	*	Water Conservancy Bureau of Quzhou County			

Note: Asterisks (\*) indicate unavailable data. COD means chemical oxygen demand, which indicates the capacity of water to consume oxygen during the decomposition of organic matter. Pollutants that never exceeded the limit any of the years are not shown.

As agriculture is the dominant consumer of water and also the main contributor of pollutants (Poore and Nemecek, 2018; FAO, 2021), water quality can be improved by reducing N losses in agrifood systems, including crop production, livestock production, food processing, and households in production areas. In a previous study (Wei *et al.*, 2023), we developed five scenarios and analyzed their effects in terms of achieving N circularity. These same scenarios proved applicable for the current study's investigation of water security, assessing the scenarios' effects on the availability of water and the impacts of water transfer, as well as regarding water quality, evaluating the potential of the scenarios to reduce NH<sub>4</sub>-N concentrations in surface water, compared to the baseline situation in 2017. For detailed scenario design and parameters, see Wei *et al.* (2023). The general scenario descriptions are as follows:

Baseline scenario (S0). N flows and discharges in the agrifood system reflect 2017 data from Quzhou county and our own 2019-2020 survey data on household consumption. As diets are slow to change, household consumption data in 2019-2020 were assumed to be representative of local diets in 2017 as well. Accordingly, scenario S0 can be regarded as the situation in the year 2017.

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Good management practices (GMP). This scenario explored the effects of better management practices in improving the efficiency of N flows in the agrifood system and reducing N discharge. Good crop production practices included optimal N fertilization, organic substitution of chemical N fertilizer, and enhanced-efficiency fertilizer. These could reduce total N fertilizer input by 14-50% and increase crop yield by 4-38%, depending on the crop species. Regarding livestock production, better feed conversion ratio could reduce feed input by 1-68%, depending on the livestock species. Additionally, this scenario includes good practices in household consumption, i.e. less animal and more plant protein as recommended by the EAT-Lancet Commission (Willett et al., 2019).

Combination of GMP plus crop and livestock waste recycling (GMP+WR1). In this scenario, crop and livestock waste (residues and manure) are recycled and treated as a resource. Residues are not sent to landfill, and all crop residues and livestock manure (except for unavoidable losses) are applied to cropland. Livestock manure is appropriately stored, avoiding direct discharge to the environment.

Combination of GMP+WR1 plus household waste recycling (GMP+WR2). In addition to recycling waste from crop and livestock production, household wastes are also recycled in this scenario. Human excreta discharge at the storage stage is avoided by replacing dry latrines with septic tanks or other clean toilets. All excreta are applied to cropland to substitute for chemical fertilizer, and all kitchen residues and food waste are used to feed livestock to substitute for concentrated feeds. Therefore, less external inputs are needed for crop and livestock production, and less nutrient losses occur in the household.

Combination of GMP+WR2 plus switching to legumes (GMP+WR+STL). In this scenario, soybean and other legumes are grown as a substitute for cotton or are planted as understory in orchards, to provide plant protein and reduce the need for food to be imported from outside Quzhou. Compared to cotton, soybean and other legumes require less N fertilizer (60 kg N ha<sup>-1</sup> yr<sup>-1</sup>, compared to 210 kg N ha<sup>-1</sup> yr<sup>-1</sup>) and less water (3,441 m³ ha<sup>-1</sup> yr<sup>-1</sup>, compared to 6,438 m³ ha<sup>-1</sup> yr<sup>-1</sup>, calculated based on the ET and planting area data in Figure 4.2) (Zhang et al., 2009; Li et al., 2023).

Combination of GMP+WR plus reducing livestock (GMP+WR+STL+RL). In this scenario, livestock production and consumption are reduced to adapt to the healthy diet proposed in the above scenarios. Resultant N losses are therefore also reduced.

The contribution of N discharge from the agrifood system ( $N_{dis}$ , Gg) to the NH<sub>4</sub>-N load ( $N_{load}$ , mg N l<sup>-1</sup>) in surface water was calculated as follows:

$$NH_{load} = (N_{dis} * 0.25 * 0.1 * 0.5/SI) * 10^{3}$$
 (10)

where the value 0.25 is the conversion fraction from the amount of N discharged to the amount of N entering surface water through runoff (Wei et al., 2021). The value 0.1 is the conversion fraction from the discharged total N to NH<sub>4</sub>-N, calculated based on a previously established database (Wei et al., 2021). The value 0.5 is the conversion fraction from discharged N to the N in surface water, considering that 50% of N is removed through retention and sedimentation before entering surface water (Schulte-Uebbing et al., 2022).

#### 4.2.5. Data sources

Table 4.2 summarizes the main input data and their sources. Main data sources were China's National Data Center for Meteorological Sciences, county statistical yearbooks, the Water Conservancy Bureau, and our own farm survey and literature reviews. Additionally, data on river flows were acquired from a web reference (LULUTONG, 2013), according to which the natural flow across Quzhou was 277 MCM in 2013. Annual river flows were assumed to be proportional to precipitation in each year. We thus estimated river flows in the other years based on this assumption and the reference value in 2013.

Table 4.2. Data sources

Data	Sources	Description			
Precipitation, temperature, relative humidity, and other meteorological data	National Data Center for Meteorological Sciences	Daily from 2010 to 2020			
Crop and forest planting structure, planting area of different plant species, administrative area	Quzhou Statistical Yearbook	Yearly from 2010 to 2020			
Groundwater and surface water supply, water quality, transported water	Water Conservancy Bureau of Quzhou County	Water supply and transfer from 2010 to 2020, water quality in 2017			
Crop planting dates, lengths of cropping seasons, crop management information	Farm survey	Survey conducted in 2019-2020			
Natural flows, crop coefficient, length of growth stages (initial, development, mid- season, and late season) for different plants, water quality	Literature review	Specific literature references are found throughout the main text			

### 4.3. Results

#### 4.3.1. Water quantity in Quzhou

Total water consumption, or ET, was 388 MCM in 2010, with wheat accounting for the largest proportion of consumption (34%), followed by cotton (25%), maize (18%), forest (8%), and vegetables (7%). Consumption increased to 460 MCM in 2020 (Figure 4.2a). This increase was attributable mainly to three vegetation types: wheat, maize, and forest. ET for these rose gradually from 130, 69, and 31 MCM, respectively, to 178, 94, and 50 MCM, with slight fluctuations. The higher ET for wheat and maize was related to climatic conditions rather than planting area, as planting area for these crops was relatively stable during the 11 years under study (Figure 4.2b and Figure S4.1). Changes in Quzhou's climate, particularly, higher wind speeds and greater relative humidity, increased the adjusted crop coefficient, contributing to the rise of ET (Figure S4.1; Eq. 6). Regarding forest, the increased ET could be partly explained by planting area, which increased by 34% from 2010 to 2020. Planting areas of cotton and vegetables decreased by 20% and 30%, respectively, which could account for their decreased ET (from 95 to 86 MCM for cotton and from 27 to 18 MCM for vegetables).

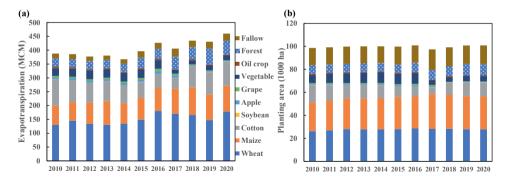


Figure 4.2. Evapotranspiration (a) and planting area (b) of various plants in Quzhou, 2010-2020

No clear pattern of change was found for precipitation in Quzhou from 2010 to 2020. The total water supply, consisting of both surface water and groundwater extraction, did gradually diminish. However, annual surface water supply increased slightly, from 49 to 57 MCM, while groundwater extraction decreased from 84 to 48 MCM annually. Surplus water tended to decrease (Figure 4.3), although it did increase in the first three years, due to high precipitation and low *ET*. The downward trend in surplus water could be indicative of less water pollution and less dissolved N losses, as these tend to parallel water discharge, according to our literature review (Figure S4.2). The surplus water values were negative in 2015, 2017, and 2019, pointing to water stress for crops in those years (see Figure 4.3).

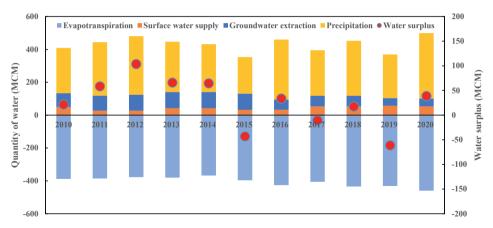


Figure 4.3. Surface water balance in Quzhou, 2010-2020

Infiltration from surface water to groundwater increased gradually from 12 MCM in 2010 to 99 MCM in 2020 (Figure 4.4). Surplus water recharge exhibited a decreasing trend, reflecting the decline in surplus water over the period (Figure 4.3). Also reflecting findings on surplus water, groundwater recharge was negative in the years 2015, 2017, and 2019. No discernible pattern of change was found in groundwater levels. The groundwater level fell by 0.74 m during the analyzed 11 years. However, most of this decrease took place in the first 6 years, during which the groundwater level fell by 2.09 m. In contrast, the groundwater level rose by 1.35 m over the last 5 study years.

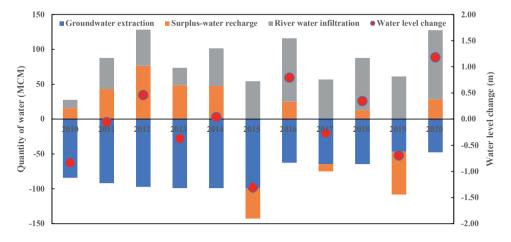
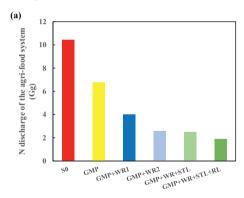


Figure 4.4. Groundwater balance and water level change in Quzhou, 2010-2020. Negative values for water level change indicate a decreasing water level.

#### 4.3.2. Water quality and potential for improvement

The primary water quality problem in Quzhou is the high concentration of NH<sub>4</sub>-N in surface water. To explore the potential of a circular agrifood system – including crop and livestock production, food processing, and household consumption – to reduce the NH<sub>4</sub>-N load in surface water, we evaluated the five scenarios introduced above, compared to the baseline in 2017 (Figure 4.5). In the baseline scenario (S0), total N discharge from the agrifood system was 10.4 Gg N yr<sup>-1</sup>, resulting in a NH<sub>4</sub>-N load of 0.59 mg N l<sup>-1</sup> in surface water (see Eq. 9). This value is half the actual NH<sub>4</sub>-N concentration (1.2 mg N l<sup>-1</sup>) in Quzhou in 2017. This suggests that the agrifood system accounted for half of the NH<sub>4</sub>-N load, with the other half attributable to other sources (such as industrial water use), though these were outside the scope of the current study (Figure 4.5b, portion below the blue line).

The first scenario, GMP, specified good management practices for crop and livestock production and healthy dietary habits. This reduced N discharge by 3.6 Gg N yr-1, corresponding to a decrease of NH<sub>4</sub>-N concentration by 0.21 mg N l-1. This translated to an NH<sub>4</sub>-N concentration in surface water of 0.97 mg N l<sup>-1</sup>, which is just below the set standard (1.0 mg N l-1). The second scenario, GMP+WR1, added to the GMP scenario improved recycling of crop and livestock residues and wastes and their application to croplands. This reduced N discharge further, from 4.0 Gg N vr-1 to 2.8 Gg N yr<sup>1</sup>, corresponding to a further decrease of NH<sub>4</sub>-N concentration in surface water by 0.16 mg N l-1 to 0.81 mg N l-1. In the third scenario, household wastes were also recycled or reused (GMP+WR2). This reduced the N discharge again, to 2.6 Gg N vr<sup>-1</sup>, corresponding to an NH<sub>4</sub>-N concentration of 0.73 mg N l-1. In the fourth scenario (GMP+WR+STL), combining previous measures with growing more legumes, N discharge decreased slightly to 2.5 Gg N vr-1. In the last scenario (GMP+WR+STL+RL), which combines all of the above measures with reduced livestock production, N discharge decreased to 1.9 Gg N yr<sup>-1</sup>, representing an NH<sub>4</sub>-N concentration of 0.69 mg N l-1 in surface water. This amounted to an 82% reduction in N discharge, compared to S0, with an NH<sub>4</sub>-N load of just 0.11 mg N l<sup>-1</sup> attributable to the agrifood system.



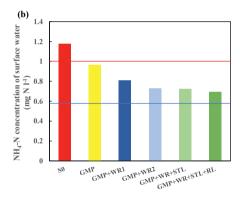


Figure 4.5. Nitrogen discharge (a) and corresponding NH<sub>4</sub>-N concentration in surface water (b) under the different scenarios. S0: baseline; GMP: good management practices; GMP+WR1: previous plus improved recycling of residues and waste from crop and livestock production; GMP+WR2: previous plus improved recycling of household waste; GMP+WR+STL: previous plus switching from cotton to legumes; GMP+WR+STL+RL: previous plus reducing livestock. The red line in (b) is the critical limit of NH<sub>4</sub>-N concentration in surface water; the blue line indicates NH<sub>4</sub>-N attributable to sources other than the agrifood system (such as industry).

#### 4.4. Discussion

#### 4.4.1. Water resources in Quzhou

To validate our findings, we compared our calculated groundwater level changes with measured or officially reported data. The calculations indicate that Quzhou's groundwater level fell by 1.25 m between 2010 and 2015, which is within the 0.5-1.7 m range reported elsewhere for the same period (Xu *et al.*, 2018; Wu and Wu, 2020). Moreover, the local government has maintained monitoring data on groundwater level since 2018 (DWRHP, 2019-2021). That data reports that the groundwater level increased by 0.92 m in 2018, decreased by 0.49 m in 2019, and then increased again by 1.68 m in 2020, which is comparable to the values calculated in this study; that is an increase of 0.34 m in 2018, a decrease of 0.70 m in 2019, and an increase of 1.18 m in 2020.

The downward trend of groundwater level was halted and even reversed in Quzhou starting in 2010 (Figure 4.4), thanks mainly to less groundwater extraction and relatively stable return flows from surface water to groundwater. In 2014 and 2015, Quzhou began to restrict groundwater extraction and implement projects to substitute surface water for groundwater (Xu et al., 2018). As a result, groundwater extraction has been halved since 2015, accompanied by increased return flows from surface water to groundwater (Figure 4.4). The latter can be attributed to water transfer projects, such as the South–North Water Transfer Project. These projects in Quzhou began operation

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in 2014. Since then, the quantity of imported water has gradually increased to approximately 65 MCM in 2020 (Bai, 2015; DWRH, 2021).

Despite water transfer projects' contributions to recovery of groundwater storage in water-receiving regions, they remain energy- and cost-intensive and may transfer water stress to water-exporting regions (Zhao *et al.*, 2015; Scanlon *et al.*, 2023). Therefore, their contributions to water security need to be evaluated connecting causes and effects on a larger scale. In addition, water transfer may lead to a false perception of unrestricted water availability, which may encourage water-receiving regions to expand water-intensive production and consumption (Grafton *et al.*, 2018). Demand-side strategies thus warrant concurrent attention.

Groundwater overexploitation can be mitigated by reducing water consumption. This was the aim of the Seasonal Land Fallowing Policy adopted in Quzhou in 2014 (Deng et al., 2021). That policy was designed to reduce the planting area of water-intensive winter wheat, which consumes the largest proportion of water among the different plant species in the county (Figure 4.2a). However, the results of this study indicate that the policy has not been put into effect, as the planting area of winter wheat did not in fact decrease. Indeed, water consumption of winter wheat has increased in the county since 2014, due to changing climatic conditions (Figure 4.2). Policy implementation could perhaps be stimulated with measures that take socioeconomic factors into account, such as by offering sufficient incentives for farmers to reduce winter wheat and by promoting development of local industry to shift surplus labor to off-farm jobs (Deng et al., 2021). Interestingly, afforestation has increased water consumption, resulting in reduced water availability in Quzhou (Figure 4.2). Previous research also found a net loss of water availability globally due to afforestation, although increased precipitation through evaporation recycling may offset this effect (Hoek van Dijke et al., 2022; Teo et al., 2022). Particularly in water-scarce areas, smart afforestation strategies are required that consider water resource endowments and ensure water availability.

Water resources were being used more efficiently in Quzhou, as indicated by the increased evapotranspiration, decreased water supply, and low surplus water (Figure 4.3). This may relate to the application of a high-efficiency, water-saving irrigation methods, as pilot projects have introduced sprinkler and drip systems in Quzhou (Xu et al., 2018). More efficient water use would decrease recharge from surface water to groundwater (Figure 4.4). Therefore, though more efficient water use in this case would not necessarily translate into water savings, it would reduce N discharge and thus help to alleviate water quality concerns. Studies published after 2010 show a positive linear relationship between the quantity of drainage water (run off to surface water or recharge to groundwater) and N discharge (Figure S4.2). Based on this relationship, we could

infer lower N pollution in Quzhou's surface waters from 2010 to 2020, attributable to the decrease in surplus water during this period. The gradual decrease in the load of NH<sub>4</sub>-N in surface water supports this inference (see Table 4.1).

#### 4.4.2. Options towards water security and N circularity

Considering the measures implemented in Quzhou, including water transfer, improved water use efficiency, and reduced groundwater extraction, water availability is no longer a problem in the county, as also suggested by other authors (Xu *et al.*, 2018; Zhao *et al.*, 2021). Thus, reducing water pollution remains key to achieve sustainable water security in Quzhou. That means reducing the NH<sub>4</sub>-N load in Quzhou's surface water (see Table 4.1). To that end, our scenario analysis provided insights into the mechanisms by which N circularity could mitigate water pollution.

According to the scenario analysis, the complete package of measures (GMP+WR+STL+RL) reduced the NH<sub>4</sub>-N load in surface water from 1.17 to 0.69 mg N l-1. This is far below the critical threshold of 1 mg N l-1 (see Figure 4.5b). Some 43% of this reduction was attributable to the application of good management practices (GMP). These practices encompassed a crop production component (optimizing fertilizer application and substituting organic for chemical N fertilizers to synchronize N supply and crop demand), a livestock production component (improving the feed conversion ratio to reduce feed intake and manure output), and a human consumption component (reducing intake of animal-sourced protein in the diet) (Wei et al., 2023). In addition, the improved recycling of crop residues and livestock manure (GMP+WR1) contributed some 32% of this reduction, mainly achieved by eliminating direct discharge from manure to surface water (Bai et al., 2016). Furthermore, the recycling of household wastes (GMP+WR2) contributed 17% of this reduction, emphasizing the need to consider households when seeking to combat water pollution. Including legumes and reducing livestock production made modest contributions towards reducing the NH4-N load in surface water (compare scenario GMP+WR+STL+RL and GMP+WR2 in Figure 4.5), as crop and livestock production would not induce excessive N discharge into water bodies if appropriately managed, as in the GMP+WR2 scenario.

In addition to addressing water quality concerns, the scenarios offer insights on water conservation and circularity. Switching from cotton to soybean (GMP+WR+STL) could reduce water requirements by 18 MCM, as cotton requires substantially more water than soybeans (calculated based on ET and planting area, see Figure 4.2). Additionally, the recycling of household waste to cropland (GMP+WR2) enables the reuse of household wastewater (8.4 MCM, data not shown). The water saved from these scenarios is equivalent to 41% of the water transferred to Quzhou (65 MCM). To eliminate the remaining 59% of the water transferred and enhance water circularity, the

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planting area of winter wheat could be reduced by 8,617 ha, or roughly 30% (calculated based on the data in Figure 4.2), as advocated under the Seasonal Land Fallowing Policy (Deng *et al.*, 2021). However, this may compromise local food security, as wheat and wheat-based products are a main food source in Quzhou and in northern China in general.

Our findings demonstrate the synergies between water security and N circularity. Additionally, promoting water security may be beneficial in pursuit of other objectives. For example, reducing groundwater extraction can also reduce energy consumption and the related greenhouse gas (GHG) emissions, as pumping is a significant unregulated source of GHG emissions in groundwater-fed agriculture (Wang *et al.*, 2012). Furthermore, potential tradeoffs between different objectives merit attention. Afforestation, for example, is widely regarded as an effective means of storing atmospheric carbon, mitigating climate change, and conserving biodiversity (Hoek van Dijke *et al.*, 2022; Stein-Bachinger *et al.*, 2022). However, as noted, it also increases water consumption and may reduce water availability in water-scarce regions. This suggests the need for future research to systematically consider co-benefits and tradeoffs between water security and other policy goals.

#### 4.4.3. Uncertainties and limitations

This study confronted a number of uncertainties and limitations, many related to data and assumptions. To estimate annual river flow, we assumed that river flow in different years was proportional to annual precipitation. Indeed, changes in groundwater levels calculated based on this assumption were found to be generally comparable to observed values, so this assumption is believed to be acceptable despite some uncertainty. In addition, the detailed mechanisms of discharged N being retained in soil and water were not considered in this study due to data limitations. Consistent with Schulte-Uebbing et al. (2022), we assumed that 50% of the discharged N was removed through retention and sedimentation before entering surface water. Furthermore, our elaboration of water security considered only environmental and biophysical aspects, though as other studies assert, water security also has economic and social welfare components, such as equitable access and distribution (Irannezhad et al., 2022). Moreover, water transferred from other regions was regarded as locally sustainable, whereas it carries the risk of externalizing water scarcity to water-exporting regions. Finally, this research focused on nutrient-related issues, specifically N pollution, in water bodies. However, other water resource issues, such as high total hardness, may also be important in the study area. Further research is needed to consider other possible issues and interactions.

#### 4.5. Conclusions

This study evaluated water security, elaborated in terms of water quantity and quality, for both surface water and groundwater. We quantified changes in the water balance over time by accounting for water flows in the case study area of Quzhou, China, from 2010 to 2020. We then conducted scenario analysis to understand synergies between N circularity and water security.

Our results confirm that Ouzhou has halted the drop of its groundwater level and in fact reversed the trend by introducing a set of water management strategies including restricting groundwater extraction, seasonal land fallowing, and water transfer. A caveat remains, however, that water transfer as a measure to increase local water availability might externalize the problem of water scarcity to water-exporting regions. The county's success in securing adequate quantities of water and improving water quality exemplifies the potential to increase water availability and qualities in water-scarce regions. Implementing such strategies on a large scale, however, requires weighing the environmental externalities of water transfer and groundwater overdraft. Water security in Quzhou was mainly threatened by high loads of NH<sub>4</sub>-N in surface water. The scenario analysis revealed the possibility of reducing the NH<sub>4</sub>-N load to a safe level, while simultaneously achieving both water security and N circularity. Less recharge flows, due to less and more efficient agricultural water use, would result in lower discharge of N pollutants. This is not a trade-off situation but a clear win-win. This study therefore suggests that while care must be taken in measures to promote water security, synergies can be found and tradeoffs minimized between different development goals.

# Acknowledgements

We are grateful to Zhan Xu for providing data on crop growth period. We also thank Gerardo van Halsema for providing some research ideas. This study was funded by the Sino-Dutch Agriculture Green Development Project of China Scholarship Council (No. 201913043), Hainan University, and the National Key Research and Development Program of China (2021YFD1700900).

# **Supporting information**

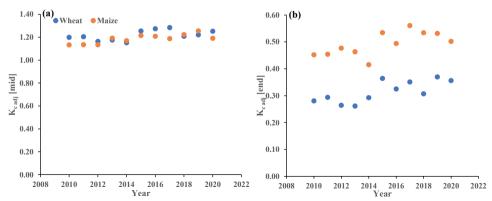


Figure S4.1. The adjusted crop coefficients for wheat and maize at mid-season (a) and late season (b) in Quzhou, 2010-2020. Kc,adj is adjusted crop coefficient based on the mean daily wind speeds, relative humidity of Quzhou during the growth stages.

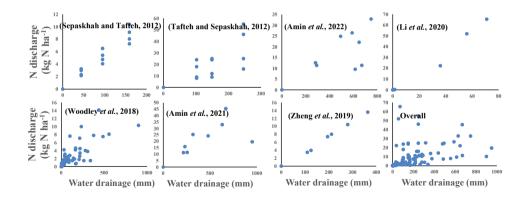


Figure S4.2. The reported relationship between the quantity of drainage water and N discharge. The data are from our literature review on the published articles after 2010.

Table S4.1. Planting dates and lengths of cropping seasons for different plants

Plant species	Sowing time	Harvest time	Growth period (day)		
Wheat	Oct I	Jun 10	252		
Maize	Jun 15	Sep 30	107		
Cotton	Apr 24	Nov 15	205		
Soybean	Jun 14	Sep 28	106		
Peanut	Apr 10	Aug 30	142		
Vegetables <sup>a</sup>	May 2	Jul 28	87		
Fruits	*	*	All the year		
Forests	*	*	All the year		

Note: the sowing and harvest days of vegetables were estimated by taking the average value of different vegetable species in Quzhou.

Table S4.2. Crop coefficient (K<sub>c</sub>) for different plants and their growth stages

Plant species	Initial		Development		Middle			Late				
	Period	Kc	$K_{c,adj}$	Period	Kc	$K_{c,adj}$	Period	Kc	$K_{c,adj}$	Period	Kc	$K_{c,adj}$
Wheat	30	0.30	0.30	161	0.30-1.15	0.30-1.20	30	1.15	1.20	31	0.25	0.28
Maize	28	0.30	0.30	29	0.30-1.20	0.30-1.13	30	1.20	1.13	20	0.50	0.45
Cotton	31	0.35	0.35	43	0.35-1.17	0.35-1.12	102	1.17	1.12	52	0.60	0.61
Soybean	15	0.40	0.40	26	0.40-1.15	0.40-1.10	45	1.15	1.10	20	0.50	0.45
Peanut	35	0.40	0.40	35	0.40-1.15	0.40-1.14	37	1.15	1.14	35	0.60	0.55
Vegetables	20	0.60	0.60	30	0.60-1.10	0.60-1.10	27	1.10	1.10	10	0.87	0.84
Apple	125	0.30	0.30	61	0.30-0.95	0.30-0.94	113	0.95	0.94	66	0.50	0.50
Grape	158	0.30	0.30	54	0.30-0.85	0.30-0.85	82	0.85	0.85	71	0.45	0.41
Forests	125	0.30	0.30	61	0.30-0.95	0.30-0.94	113	0.95	0.94	66	0.50	0.50

Note: Kc,adj was adjusted crop coefficient based on the mean daily wind speeds, relative humidity and crop heights during the growth stages, so the value would be different for different years with varying climate conditions. In this table, the climate conditions in 2010 were used to show a case. Information for forests was unavailable, and we assumed it was the same as the value of apple.

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Optimizing carbon and nitrogen cycles towards net-zero greenhouse gas emissions in agrifood systems: a case study in Quzhou, China

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#### **Abstract**

Agrifood systems emit substantial amounts of greenhouse gases (GHG) into the atmosphere, but simultaneously sequester carbon (C) originating from atmospheric CO<sub>2</sub> in soils. Their net effects on the GHG balance are rarely documented. This study aimed to quantify C cycles and GHG emissions in an agrifood system at both village and county levels and explore the potential to transition towards net-zero emissions. We integrated a modified material and nutrient flow model (NUFER) and a soil C cycle model (RothC) to calculate C cycles in the case area of Quzhou, China. Results showed that net photosynthesis predominantly contributed to C input to the agrifood system, while soil respiration and microbial respiration during manure storage accounted for most of the C output at all village types. Net CO<sub>2</sub> emissions from the agrifood system in Quzhou were positive because the amount of C sequestration in soils (819 kg C ha-1 yr<sup>-1</sup>) could not offset CO<sub>2</sub> emissions (847 kg C ha<sup>-1</sup> yr<sup>-1</sup>) from fossil fuels used in the agrifood system, despite village-level variations. Scenario analysis demonstrated that the system could be transformed to net-zero CO<sub>2</sub> emissions by adopting good management practices and recycling organic wastes. However, achieving net-zero GHG emissions may necessitate the substitution of fossil energy by clean energy. Policies and incentives to promote net-zero emissions and circular agriculture can be mutually reinforcing. The methods and options explored in this study provide insights on managing C cycles and GHG emissions in the agrifood system, facilitating the transition towards net-zero emissions and climate neutrality.

Optimizing carbon and nitrogen cycles towards net-zero emissions

### 5.1. Introduction

Agrifood systems are key to food security and achieving the goal of limiting warming at the end of this century to below 1.5 °C compared to the pre-industrial level (Crippa et al., 2021; FAO, 2022; Matthews and Wynes, 2022). They are responsible for the provision of adequate nutritious food in the face of a growing population and consumption level worldwide (Chaudhary et al., 2018; FAO, 2022). Meanwhile, they emit a third of global anthropogenic greenhouse gases, and induce about 22% of global mortality due to air pollution (Poore and Nemecek, 2018; Crippa et al., 2021; Crippa et al., 2022). The future of the agrifood system needs to nourish humanity in an environment- and climate-friendly way.

Quantification of carbon (C) cycles in agrifood systems is necessary to understand their net CO<sub>2</sub> emissions (Wolf *et al.*, 2015). These cycles include both C flows and storage in the compartments of crop production, livestock production, food processing, household consumption and waste management (Wolf *et al.*, 2015; Wei *et al.*, 2023). The C storage in agrifood systems is mainly in soils which constitute an important C pool on earth (Sykes *et al.*, 2019). Promoting soil C sequestration is often listed as a potential solution to combat climate change (Sykes *et al.*, 2019; Walker *et al.*, 2022). However, the direct measurement of soil C storage and changes is costly and time-consuming for large areas (Paul *et al.*, 2023). In addition, the change of soil C storage is a slow process requiring long-term observations, and the annual change rate depends on the initial soil C content which affects sequestration potential due to C saturation (Lugato *et al.*, 2014; Moinet *et al.*, 2023). Modelling is regarded as a feasible option to predict soil C storage and changes, but proper validation before use is vital to ensure prediction accuracy. Unfortunately, more than 70% of the models simulating soil C storage are not validated (Garsia *et al.*, 2023).

Carbon sequestration in soils and agrifood systems should be considered in light of CO<sub>2</sub> emissions caused by the energy and power consumption required for system functioning (Lorenz and Lal, 2018; Adetona and Layzell, 2019). The energy and power consumption is primarily induced by groundwater pumping for irrigation (Wang *et al.*, 2012; McDermid *et al.*, 2023), machinery operation for seeding and harvesting, the use of plastic mulch for cash crops (Guo *et al.*, 2022), and livestock production (Xing *et al.*, 2022). The consequent CO<sub>2</sub> emissions could counteract or even outweigh C sequestration in soils (Wolf *et al.*, 2015; Poore and Nemecek, 2018), i.e. the agrifood system itself might be a CO<sub>2</sub> source rather than a sink. When considering net climatic effects, the situation is even worse due to non-CO<sub>2</sub> greenhouse gas (GHG) emissions, namely CH<sub>4</sub> and N<sub>2</sub>O which have higher global warming potential (GWP) than CO<sub>2</sub> (Feral, 2015). Even though more than 80 countries, representing approximately 70% of

global GHG emissions, have pledged net-zero emissions mostly by the current mid-century (Matthews and Wynes, 2022; Zhao *et al.*, 2022), the potential and options to achieve net-zero CO<sub>2</sub> (C neutrality) or even net-zero GHG (climate neutrality) emissions in the agrifood system are still unclear.

The net-zero CO<sub>2</sub> and net-zero GHG emissions in the agrifood system may be promoted by strategies that increase C sequestration and reduce GHG emissions (Wang et al., 2021; Wolf et al., 2021). For crop production, better management practices reduce fertilizer inputs, mitigate nutrient and N<sub>2</sub>O losses, increase crop biomass and thus C inputs to cropland (Janzen et al., 2022; Minasny et al., 2022); for livestock production, feed management and rumen manipulation increase feed conversion ratio and reduce CH<sub>4</sub> emissions by more than 20% (Arndt et al., 2022; Fang et al., 2023); the recycling of animal and human manure to cropland contributes to C sequestration in soils, aiding in reducing C and N emissions to the environment (Rose et al., 2015; Li et al., 2021); adjusting plant structure could reduce N and C emissions while ensure food security (Wang et al., 2022); reducing livestock breeding also yields climate benefits (Resare Sahlin and Trewern, 2022). All these strategies influence both CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O emissions (Poore and Nemecek, 2018), and their true effect on GHG emissions can't be accurately evaluated without considering both C and N cycles in the agrifood system.

The practical implementation of management strategies is affected by spatial scale (Yan et al., 2020; Wei et al., 2023). The expansion of management measures requires policy actions, while policy and governance are generally constrained by administrative boundaries (Brownlie et al., 2022; FAO, 2022). China is the largest emitter of GHGs, accounting for 26% of the global CO<sub>2</sub> emissions during 2010-2019 (UNEP, 2020). In China, agricultural extension services are organized at county level, and farm recommendations and agricultural policies are often implemented by a county agricultural bureau. County-specific support and policy is necessary for implementing optimization strategies (Wang et al., 2022). In addition, county/district was reported to be the optimal scale to restore circularity and reduce emissions (Van der Wiel et al., 2020; Wei et al., 2023), because it is not only sufficiently small to capture local circumstances, facilitating product transport and exchange, but also large enough to include all compartments of agrifood systems.

To explore the options to achieve net-zero emissions in the agrifood system at an optimal scale, considering both C sequestration and emissions of all GHGs, we selected Quzhou county (China) as a case study. China pledged to peak CO<sub>2</sub> emissions by 2030 and achieve C neutrality by 2060 (Feng and Fang, 2022). Quzhou is a demonstration county for China's "Agricultural Green Development" strategy (MOA, 2020), seeking to produce nutritious agricultural products while minimizing environmental impacts.

We first quantified C flows in different compartments of the agrifood system. We then calculated CO<sub>2</sub> emissions induced by energy and power consumption required for the agrifood system functioning. Thereafter, we evaluated the net GHG emissions of Quzhou's agrifood system. Finally, we explored the scenarios to achieve a C- or climateneutral system.

### 5.2. Materials and methods

#### 5.2.1. Study area

Quzhou county is in the center of North China Plain. It has a sub-humid, temperate, continental monsoon climate. The county covers an area of 677 km² and includes 10 towns and 342 village-size administrative units. The registered population is 527,000 in 2017, with over 90% of the inhabitants residing in rural areas (NBSC, 2018). Agricultural production here is in great transition. Grain yield has increased from 1.5 to 7.1 Mg ha¹ while meat production has increased fifty times from 1145 to 57,440 Mg yr¹ in the past four decades. The main crops are winter wheat, summer maize, cotton, and vegetables and fruits. Eight percent of the arable land is irrigated. The main livestock includes layer, broiler, sheep and goat, and cattle.

To account for the heterogeneity of agrifood systems within in Quzhou, the 342 villages was grouped into four types according to the method we used before (Wei et al., 2023): 1) cereal villages, 2) cash crop villages, 3) livestock villages, and 4) land-limited villages. Villages in the same type are considered to be similar in production and consumption patterns, and C flows from one village type to the same would not take place. Carbon fluxes of each village type are average values of all villages within the type. These fluxes at village levels were then scaled up to county levels based on C flows per arable land, the average arable land area of each village type, the number of villages in each type, and inter-village C flows of crop and livestock products and manure. The interflows were collected in a year-round survey and a detailed explanation of the survey is provided in Wei et al. (2023). We only considered the arable land, and the land for housing and infrastructure was excluded because they rarely involve in C cycles.

# 5.2.2. Carbon cycles in the agrifood system

The time scale of this research is one year, and C cycles in the agrifood system based on the 2017 county statistical data (the latest available data when conducting this work) and our 2019–2020 survey data was regarded as the baseline scenario (S0).

#### 5.2.2.1. Soil C sequestration

Soil C sequestration was calculated based on the changes in annual content of soil organic carbon (SOC) modelled using the Rothamsted Carbon (RothC) model, which has been widely used to predict soil C stocks depending on input in Europe and elsewhere (Coleman and Jenkinson, 2014; Garsia *et al.*, 2023). We validated the model against data from an 8-year field experiment in Quzhou, and it performed well in predicting soil C stocks and changes for all fertilizer treatments (Figure S5.1). For this study, we used the average value of the predicted soil C sequestration during 2020-2060, considering that the rate of soil C sequestration is temporal dynamic due to C saturation, and China plans to reach C neutrality by 2060. Figure 5.1 presents the annual rate of soil C sequestration at different village types in Quzhou.

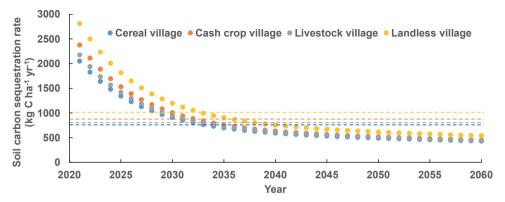


Figure 5.1. The predicted annal rate of soil carbon sequestration in different village types in Quzhou based on current soil C content, land use and agricultural management practices (circles). The horizontal dashed lines indicate the average values for individual village types from 2020-2060.

### 5.2.2.2. Carbon flows in the agrifood system

Carbon flows in different compartments of the agrifood systems were quantified using a model adapted from the Nutrient Flows in food chain, Environment and Resource use (NUFER) model. The NUFER model has been used to assess nitrogen (N) and phosphorus (P) flows in agrifood systems, including the compartments of crop production, livestock production, food processing, and household consumption (Ma et al., 2010; Bai et al., 2018; Wei et al., 2023). According to its framework, we modified and expanded the model to quantify biomass and C flows in the agrifood system. Carbon flows were calculated for the crop production, livestock production, food processing, household consumption, and waste management compartments. For the food processing compartment, we only considered milling of wheat and maize because of limited data and claimed minor effects of this compartment in this area (Wei et al., 2023).

Carbon flows were quantified based on material flows in the agrifood system and the C content of specific materials. Data on detailed material flows are presented in the following section. Data on C content of specific materials is presented in Table S5.1.

For the crop production compartment, C inputs include net photosynthesis, seeds, and organic fertilizers; outputs include crop products, crop resides, soil respiration, and soil C discharge. We used specific input and output data for individual crops. Data on crop-specific application rates of organic fertilizers, the amount of seeds, crop products and residues were collected in a survey of 1308 crop farmers throughout Quzhou (Wei *et al.*, 2023); the quantity of soil C discharge was estimated by organic N discharge and the ratio of organic C and N (C/N) in discharged water. The amount of crop-specific organic N discharge was acquired from our previous calculation (Wei *et al.*, 2021; Wei *et al.*, 2023), and the C/N ratio in discharged water is assumed to be 10 (Ros *et al.*, 2010); net photosynthesis was estimated based on mass balance of all the input and output flows as well as C sequestration for individual crops. It is assumed to equal the sum of C outputs plus C sequestration, minus the C inputs through flows other than net photosynthesis.

For livestock production, C inputs include feeds and imported young stock; outputs include animal products, enteric CH<sub>4</sub> emissions, animal manure, and animal respiration. Data on the young stock, feed composition, and feeding rates for different livestock species, animal products were collected from a survey of 94 livestock farmers throughout Quzhou (Wei *et al.*, 2023); the amount of animal manure was calculated based on animal-specific ratio between manure and feed intake as suggested by Wolf et al. (2015); the enteric CH<sub>4</sub> emissions were estimated by emission factors as suggested by Xing et al. (2022), and detailed data are presented in Table S5.2; data on animal respiration were estimated based on mass balance of all the inputs and output flows for individual animal species.

For households, C inputs are foodstuffs, and outputs include food waste, kitchen residues, human excreta, and respiration. The amount and types of food consumed, food waste, kitchen residues were collected in a year-round survey (Wei *et al.*, 2023); the amount of human excreta was calculated using the factors provided in (Wen *et al.*, 2021), i.e. 14.5 kg C yr<sup>-1</sup> per person; data on human respiration was estimated based on mass balance of all the inputs and output flows.

Waste management includes plant residues, animal manure and human excreta, connecting livestock production and household consumption with crop production. The produced plant residues are recycled to cropland, sold as feed, or discarded to landfill. The amount of each portion was collected from the survey of 1308 crop farmers (Wei *et al.*, 2023); the produced animal manure was recycled to cropland or sold as

fertilizer after the process of storage and/or treatment. The amount of recycled and sold manure was collected from the survey of 94 livestock farmers (Wei et al., 2023). During the storage, some 50% of the manure is discharged to the environment (Bai et al., 2016); CH<sub>4</sub> and CO<sub>2</sub> are also emitted during storage. Data on CH<sub>4</sub> emissions were from Xing et al. (2022), and detailed data are presented in Table S5.3. Data on CO<sub>2</sub> emissions were calculated based on mass balance of all produced manure and the mentioned flows; for human excreta, the proportions of different flows were assumed to be the same as animal manure due to limited data.

#### 5.2.3. Energy and power emissions

The functioning of agrifood systems entails irrigation, machinery operation, agrochemicals. The CO<sub>2</sub> emissions induced by these processes were calculated as cropand animal-specific. For every crop, irrigation-induced CO<sub>2</sub> emissions were calculated by multiplying an emission factor with the amount of electricity consumed. The same method was used to quantify CO<sub>2</sub> emissions induced by machine (diesel-powered) and plastic mulch produced within Quzhou. Detailed data and parameters are listed in Table S5.4 and S5.5. Emissions from the production of electricity and plastic mulch were incorporated in this study because there are thermal power stations and plastic plants within Quzhou, while emissions induced by the production of pesticides and chemical fertilizers were not considered because they are produced outside the boundary of Quzhou. For every animal species, the emissions were calculated by multiplying emission factors with the amount of consumed electricity, coal and diesel. These fossil fuels are utilized for ventilation, heating (in winter), and animal feeding in livestock production. Detailed data and parameters are listed in Table S5.5 and S5.6.

### 5.2.4. Net emissions of CO<sub>2</sub> and GHGs

Net emissions in this study refer to the gap between C sequestration in the agrifood system and CO<sub>2</sub>-C emissions required for system functioning. They are calculated as follows.

$$NC = EC_{E\&P} - SC_S \tag{1}$$

where NC means net  $CO_2$  emissions (kg  $CO_2$ -C ha<sup>-1</sup>);  $EC_{E\&P}$  indicates emitted  $CO_2$ -C induced by energy and power consumption as stated in section 5.2.3;  $SC_S$  is sequestered  $CO_2$ -C in soils, calculated with the RothC model. A negative value for NC represents a net  $CO_2$  sink of the agrifood system, while a positive value indicates a net  $CO_2$  source. The  $CO_2$ -C emissions from animal and human respiration stem from feeds and foods while C in these biomasses is assimilated from  $CO_2$  in the atmosphere. Therefore, these emissions were not taken into account here. The  $CO_2$  emissions from manure management and crop residue disposal were not considered either for the same reason.

$$NGHG = (EC_{E\&P} - SC_S) * 44/12 + EC_{CH_4-C} * 16/12 * 28 + (E_{NH_3-N} * 0.01 + E_{NO_3^--N} * 0.0075 + E_{N_2O-N}) * 44/28 * 265$$
(2)

where NGHG is net GHG emission (kg CO<sub>2</sub> eq ha<sup>-1</sup>);  $EC_{CH_4-C}$  is emitted CH<sub>4</sub>-C from the agrifood system as stated in section 5.2.2.2.  $E_{NH_3-N}$ ,  $E_{NO_3^--N}$ , and  $E_{N_2O-N}$  are emitted N as NH<sub>3</sub>, NO<sub>3</sub>- and N<sub>2</sub>O from the agrifood system; their values are from our previous calculations (Wei *et al.*, 2023); the values of 44/12, 16/12 and 44/28 are mass conversion factors of C to CO<sub>2</sub>, CH<sub>4</sub>, and N to N<sub>2</sub>O; the values of 28 and 265 represent the global warming potential of CH<sub>4</sub> and N<sub>2</sub>O in units of CO<sub>2</sub> equivalents over a 100-year period based on the fifth IPCC assessment report (Feral, 2015); The values of 0.01 and 0.0075 are the indirect N<sub>2</sub>O-N emissions from emitted NH<sub>3</sub>-N and leached NO<sub>3</sub>-N base on IPCC methodology (Paustian *et al.*, 2006). Negative values for *NGHG* represent a net CHG sink, while a positive value indicates a net source.

#### 5.2.5. Scenarios towards net-zero emissions

The agrifood system in Quzhou induces substantial environmental emissions, and such emissions can be mitigated by better management practices, organic waste recycling from livestock production and households to crop production, adjusting the structure of crop planting and animal feeding (Wei *et al.*, 2023). According to these mitigation practices, five advanced scenarios were designed, as done in our previous study (Wei *et al.*, 2023). In that study, we quantified N emissions affected by these scenarios; here we explored options toward net-zero emissions in the agrifood system, consider both C and N cycles.

Good management practices (GMP). This scenario includes better management practices for both crop production, livestock production and household consumption. Good crop production practices in this study are optimal fertilization, the combined use of organic and inorganic fertilizers, and enhanced-efficiency fertilizers. These could reduce fertilizer input and N emissions, increase crop biomass and thus C inputs to cropland, depending on the crop species. Detailed parameters were from Wei et al. (2023). Good livestock production practices, such as rumen manipulation, could reduce enteric CH<sub>4</sub> emissions by 21% (Arndt *et al.*, 2022). Good household consumption means less animal and more plant protein as recommended by the EAT-Lancet Commission (Willett *et al.*, 2019).

Combination of GMP and recycling of crop and livestock waste (GMP+WR1). Crop and livestock waste (residues and manure) are recycled and treated as a resource. Residues are not sent to landfill, and all crop residues and livestock manure (except for unavoidable losses) are well managed and applied to cropland, resulting in a further reduction of chemical fertilizer use. Livestock manure is appropriately stored. Direct discharge of

manure to the environment is avoided and CH<sub>4</sub> emissions during manure management are reduced by 50%, 36% and 26%, respectively, for pig, cattle and sheep/goat (Xing et al., 2022).

Combination of GMP+WR1 and recycling of household waste (GMP+WR2). In addition to recycling waste from crop and livestock production, household wastes are also recycled in this scenario. Human excreta discharge is avoided by replacing dry latrines with septic tanks or other clean toilets. All excreta are applied to cropland to replace chemical fertilizers, and all kitchen residues and food waste are used to feed livestock as substitute for concentrated feeds. CH<sub>4</sub> emissions during human excreta management are reduced by 50%, just as pig manure in the last scenario.

Combination of GMP+WR2 and switching to legumes (GMP+WR+STL). Soybean and other legumes are grown as a substitute for cotton or are planted as understory in orchards in this scenario, to provide plant protein and reduce the need for food to be imported from outside Quzhou. Compared to cotton, soybean and other legumes require less agrichemical inputs therefore produce less C emissions (Zhang et al., 2009; Guo et al., 2022).

Combination of GMP+WR and reducing livestock (GMP+WR+STL+RL). Livestock production is reduced by 87% to adapt to the healthy diet proposed in the above scenarios. This proportionately reduces CH<sub>4</sub> emissions from enteric fermentation and manure management, CO<sub>2</sub> emissions from energy and power consumption due to livestock production.

### 5.3. Results

### 5.3.1. Carbon cycles in agrifood systems

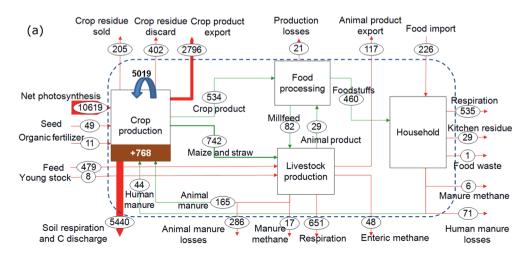
### 5.3.3.1. Village-level C cycles in agrifood systems

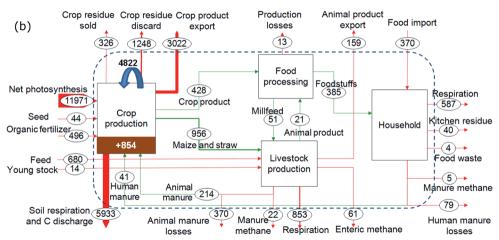
At village level, net photosynthesis was the dominant C input into the crop compartment, which reached 10.6, 12.0, 11.6 and 14.1 Mg C ha<sup>-1</sup> yr<sup>-1</sup> at cereal, cash crop, livestock and land-limited villages, respectively (Figure 5.2). Land-limited villages exhibited the highest net photosynthesis because their croplands received more nutrient and organic matter inputs compared to those in other types of villages. Soil respiration and C discharge accounted for approximately half of the C outputs from the crop production compartment. A moderate amount of C went to landfill sites through crop residue discards which were 402, 1248, 710 and 1168 kg C ha<sup>-1</sup> yr<sup>-1</sup>, respectively, at cereal, cash crop, livestock and land-limited villages; most of crop residues were recycled to croplands. The values of C sequestration in soils were 768, 854, 805 and 1012 kg C

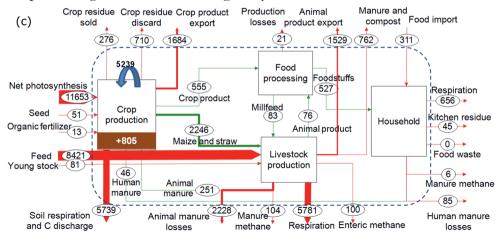
ha-1 yr-1 C at cereal, cash crop, livestock and land-limited villages, respectively. The sequestration rates ranked identical to net photosynthesis in the different villages.

The values of C inputs through feed were 8421 and 4991 kg C ha<sup>-1</sup> yr<sup>-1</sup> for the livestock and land-limited village, respectively, while the rates were <1000 kg C ha<sup>-1</sup> yr<sup>-1</sup> for cereal and cash crop villages (Figure 5.2). The majority of the C input was emitted to the atmosphere through animal respiration. In addition, a considerable amount of C was lost to the environment due to impropriate manure management and the values were 286, 370, 2228 and 1554 kg C ha<sup>-1</sup> yr<sup>-1</sup>, respectively, for cereal, cash crop, livestock and land-limited villages. A small amount of C was emitted as CH<sub>4</sub>-C through enteric fermentation of animals and anaerobic fermentation of manure. The total amount was 65, 83, 204 and 198 kg C ha<sup>-1</sup> yr<sup>-1</sup> for cereal, cash crop, livestock and land-limited villages, respectively.

The inputs through food were <500 kg C ha<sup>-1</sup> yr<sup>-1</sup>, except for land-limited villages which imported 1774 kg C ha<sup>-1</sup> yr<sup>-1</sup> in food (Figure 5.2), because they did not have enough land to be self-sufficient. Human respiration consumed some 80% of the total C input to the household, and human manure accounted for another non-trivial portion, although most of it was lost to the environment (71, 79, 85, and 204 kg C ha<sup>-1</sup> yr<sup>-1</sup> for the four village types, respectively). A small amount of C was also emitted as CH<sub>4</sub>-C through anaerobic fermentation of human manure. Amounts of C in food waste and kitchen residue were not large; their values were <150 C ha<sup>-1</sup> yr<sup>-1</sup> for all four village types.







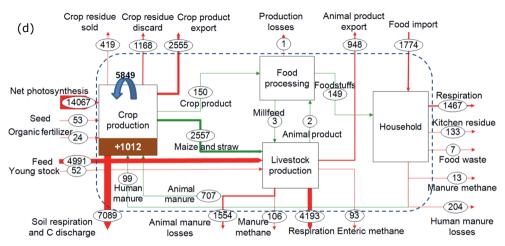


Figure 5.2. Carbon flows (kg C ha<sup>-1</sup> yr<sup>-1</sup>) for cereal (a), cash crop (b), livestock (c) and land-limited villages (d) in Quzhou. The numbers in the brown boxes indicate carbon sequestration (kg C ha<sup>-1</sup> yr<sup>-1</sup>) in soil; the dashed blue lines represent the boundaries of the villages; the green and red arrows indicate carbon flows within and across village boundaries, respectively; the thickness of the arrows is proportional to their value; the blue arrows on the crop production compartment indicate straw returning.

### 5.3.1.2. C flows between village types

Carbon flows connected the four village types (Figure 5.3). Livestock villages imported a large amount of maize from cereal (575 kg C ha<sup>-1</sup> yr <sup>-1</sup>) and cash crop villages (269 kg C ha<sup>-1</sup> yr <sup>-1</sup>) as feed for livestock production. In turn, they exported animal manure to cash crop villages because they did not have enough cropland to apply the manure while cash crops normally need substantial amounts of manure and other organic fertilizers. Land-limited villages imported a small amount of maize (17 kg C ha<sup>-1</sup> yr <sup>-1</sup>) from cash

crop villages. In addition, they imported grains (52 kg C ha<sup>-1</sup> yr <sup>-1</sup>), animal products (19 kg C ha<sup>-1</sup> yr <sup>-1</sup>) as well as fruits and vegetables (19 kg C ha<sup>-1</sup> yr <sup>-1</sup>) from cereal, livestock and cash crop villages, respectively, because they did not have enough arable land for production. Cereal villages exported grains to other villages while imported fruits and vegetables (8.8 kg C ha<sup>-1</sup> yr <sup>-1</sup>) from cash crop villages and animal products (12.8 kg C ha<sup>-1</sup> yr <sup>-1</sup>) from livestock villages.

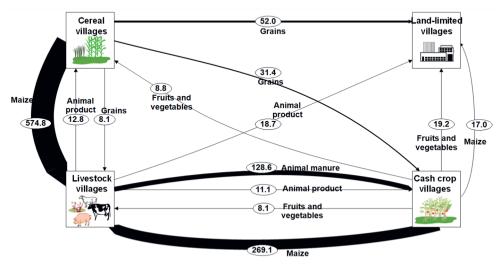


Figure 5.3. Carbon flows (kg C ha-1 yr-1) between the village types in Quzhou.

### 5.3.1.3. County-level C cycles in agrifood systems

County-level C cycles were calculated based on village-level cycles upscaled by arable land and flows between the village types (Figure 5.4). Like the village-level data, net photosynthesis contributed the most to C input in the agrifood system at county level, with a value of 11.5 Mg C ha<sup>-1</sup> yr <sup>-1</sup>, followed by feed (1843 kg C ha<sup>-1</sup> yr <sup>-1</sup>) and foodstuffs (239 kg C ha<sup>-1</sup> yr <sup>-1</sup>). The inputs from seed, young stock and biofertilizer were minor. The average value of C sequestration in cropland was 819 kg C ha<sup>-1</sup> yr <sup>-1</sup>, with land-limited villages accumulating most per arable land. However, the total amount of C sequestration in land-limited villages is the lowest (data not shown) due to their limited arable land at the county level. The CO<sub>2</sub>-C emissions from the respiration of livestock and humans accounted the majority of the C output from Quzhou's agrifood system. Water discharge and crop residue discard induced C losses by 860 and 761 kg C ha<sup>-1</sup> yr <sup>-1</sup>, respectively. A considerable amount of C was exported outside Quzhou as marketable foodstuffs (2070 C ha<sup>-1</sup> yr <sup>-1</sup>), crop resides (271 C ha<sup>-1</sup> yr <sup>-1</sup>) and composting (45 C ha<sup>-1</sup> yr <sup>-1</sup>), at the cost of CH<sub>4</sub> emissions by 118 C ha<sup>-1</sup> yr <sup>-1</sup>.

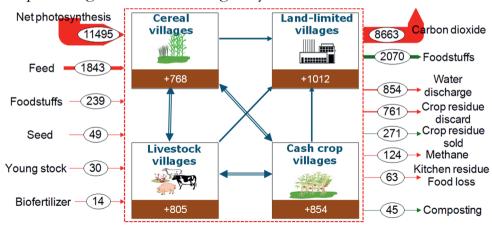


Figure 5.4. Carbon cycles (kg C ha<sup>-1</sup> yr<sup>-1</sup>) in Quzhou. Numbers in the brown boxes represent carbon sequestration in cropland within the type of village. The dashed red line indicates the boundary of Quzhou. Blue arrows within the boundary are flows between the village types within Quzhou. Arrows on the left and right sides are input and output flows, respectively, with green arrows representing C outputs in marketable products.

# 5.3.2. CO<sub>2</sub> emissions induced by fossil fuel use in the agrifood system of Ouzhou

The CO<sub>2</sub> emissions induced by fossil fuel use in the agrifood system were 654, 834, 1112 and 1155 kg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup> at cereal, cash crop, livestock and land-limited villages, respectively (Table 5.1). The total value for Quzhou would be 819 kg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup> when upscaling village-level data using arable land at different village types. The production and consumption of electricity for irrigation and animal production contributed the most (approximately 70%) to CO<sub>2</sub> emissions at all villages in Quzhou. Diesel consumption for machinery operation and animal production induced emissions of 138, 148, 149 and 174 kg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup> at the four village types, respectively. The use of coal for animal production caused emissions of 12 kg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup> at cereal village and 94 kg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup> at livestock village, and this disparity reflected the differences in stock density of animals. In addition, the production of plastic mulch for crop production resulted in approximately 10% of the total CO<sub>2</sub> emissions.

Table 5.1. CO<sub>2</sub> emissions (kg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup>) caused by the functioning of agrifood systems

	Electricity	Diesel	Coal	Plastic mulch	Sum
Cereal village	461	138	12	43	654
Cash crop village	548	148	21	117	834
Livestock village	800	149	94	68	1112
Land-limited village	799	174	62	120	1155

#### 5.3.3. Scenarios towards net-zero CO<sub>2</sub> and net-zero GHG emissions

At current conditions (S0), the net CO<sub>2</sub> emission from the agrifood system was positive for Quzhou, because the value of CO<sub>2</sub> emissions caused by energy and power consumption for system functioning exceeded that of C sequestration in soils (Figure 5.5a). The net GHG emissions would be 7.5 Mg CO<sub>2</sub> eq ha<sup>-1</sup> yr<sup>-1</sup>, when considering the global warming effects of CH<sub>4</sub> as well as direct and indirect N<sub>2</sub>O emissions (Figure 5.5b). Although the total amount of CH<sub>4</sub>-C emission from the agrifood system was significantly lower than that of C sequestration in soils (Figure 5.4), its global warming effect outweighed that of C sequestration due to the higher global warming potential per amount of C for CH<sub>4</sub> compared to CO<sub>2</sub>.

Good management practices (GMP) could increase crop biomass production and thus C sequestration in soils, transform the agrifood system from a C source into a C sink (Figure 5.5a). The net CO<sub>2</sub> emission would be -0.10 Mg CO<sub>2</sub>-C ha<sup>-1</sup> vr<sup>-1</sup>, although the net GHG emissions remained high (5.6 Mg CO<sub>2</sub> eq ha-1 yr-1) due to global warming effects caused by CH<sub>4</sub> as well as direct and indirect N<sub>2</sub>O emissions. The second advanced scenario (GMP + WR1), including the above measures plus the recycling of crop residues and animal manure to substitute chemical fertilizer use on cropland, further reduced the net CO<sub>2</sub> emission to -0.23 Mg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup> and net GHG emissions to 4.5 Mg CO<sub>2</sub> eq ha<sup>-1</sup> yr<sup>-1</sup>. The net CO<sub>2</sub> and GHG emissions didn't change significantly when extending the recycling effort to household wastes (compare GMP + WR1 and CMP + WR2) due to their limited quantitative contribution to the county flows and emissions. The fourth advanced scenario (GMP + WR + STL), including the above measures plus growing more legumes, slightly reduced CO<sub>2</sub> emissions, but it had a limited contribution to the changes in net CO<sub>2</sub> and GHG emissions. The last advanced scenario (GMP + WR + STL + RL) included all the previous measures plus reduced stock rate of animals by 87%. It substantially reduced the net CO<sub>2</sub> emissions to -0.39 Mg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup> by reducing energy and power consumption caused by livestock production. Meanwhile, it further reduced the net CHG emissions by 90% from 4.2 to 0.49 Mg CO<sub>2</sub> eq ha<sup>-1</sup> yr<sup>-1</sup> (compared to the previous scenario), because the global warming effects of CH<sub>4</sub> as well as direct and indirect N<sub>2</sub>O emissions were significantly reduced in this scenario as a result of less livestock and manure production.

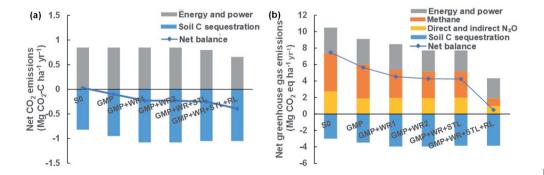


Figure 5.5. Net CO<sub>2</sub> (a) and greenhouse gas (b) emissions in different scenarios in Quzhou. S0: baseline (current situation); GMP: good management practices; GMP + WR1: same as previous and recycling of crop and livestock waste; GMP + WR2: same as previous and recycling of household waste; GMP + WR + STL: same as previous and switching to legumes; GMP + WR + STL + RL: same as previous and reducing livestock breeding.

#### 5.4. Discussion

This study evaluated both C flows and storage in an agrifood system by combing an adapted NUFER model and a validated RothC model. Net emissions from the agrifood system were quantified using C sequestration against CO<sub>2</sub> emissions required for system functioning. Our results demonstrate a net-positive CO<sub>2</sub> emission in the agrifood system of Quzhou. The system can transition to net-zero or even net-negative CO<sub>2</sub> emissions by adopting advanced practices, but the modelled scenarios do not lead to net-zero GHG emissions. As far as we are aware, this is the first study quantifying C cycles in an entire agrifood system as well as its net CO<sub>2</sub> and GHG emissions.

## 5.4.1. C cycles in the agrifood system of Quzhou

Carbon flows and storage comprise C cycles in the agrifood system as shown in Figure 5.1-5.3. This study assumed that C storage/sequestration occurred only in soils. Carbon in foodstuffs or other biomass was not considered to be "storage" because it would be re-emitted into the atmosphere after being used, unless it is not consumed but preserved (Lorenz and Lal, 2018; Xing *et al.*, 2022). The CO<sub>2</sub> emissions from the respiration of livestock and humans are not counted when evaluating the net emissions of agrifood system, because they stem from C in biomasses assimilated from CO<sub>2</sub> in the atmosphere.

The amount of C sequestration in soils ranged from 768 to 1012 kg C ha<sup>-1</sup> yr-1 for different village types, with an average value of 819 kg C ha<sup>-1</sup> yr-1 (Figure 5.1 and Figure 5.3), which is higher than the reported values of 140-700 kg C ha<sup>-1</sup> yr<sup>-1</sup> in China (Fang

et al., 2018; Tao et al., 2019; Hübner et al., 2021). The potential reason could relate to initial C content in soils. Soils with lower initial C content tend to retain more C than SOC-rich soils (Hübner et al., 2021; Oldfield et al., 2022). The initial C content is on average approximately 0.8% in the soil of Quzhou (data not shown), while it exceeds 2% in previous reports across China (Fang et al., 2018; Hübner et al., 2021).

#### 5.4.2. Net GHG emissions of the agrifood system

The net CO<sub>2</sub> emissions by cereal and cash crop villages were negative, but they were positive from livestock and land-limited villages (Compare Figure 5.1 and Table 5.1). Such disparities prove the necessity to consider village-level heterogeneity of agrifood systems when analyzing county-level cycles in Quzhou. The variations in net emissions across villages were primarily attributed to different livestock densities. Livestock production contributes significantly to CO<sub>2</sub> emissions through electricity and diesel consumption (Table S5.5 and S5.6), even though the recycling of animal manure could enhance C sequestration in arable soils (Figure 5.1).

When analyzing CO<sub>2</sub> emissions, we only considered processes occurring within Quzhou, including the production and utilization of electricity and plastic mulch, as well as the utilization of diesel and coal (NBSC, 2018). Emissions induced by the production of diesel, coal, pesticides and chemical fertilizers were excluded as they occur outside Quzhou. This externalizes emissions to agrichemical-producing regions. Meanwhile, Quzhou exports massive amounts of food, and this internalizes emissions caused by production for consumption in other regions. The net effects of food exchange and trade on emissions in Quzhou are beyond the scope of this study, but agricultural trade generally contributes to reducing CO<sub>2</sub> emissions from agrifood systems (OECD and FAO, 2022; WTO, 2022). Further research is needed to quantify and minimize the production-, consumption- and trade-induced CO<sub>2</sub> emissions.

The current agrifood system in Quzhou is a net CO<sub>2</sub> source (Figure 5.5). It may be problematic to rely on soil C sequestration in combating climate change. Soil C sequestration could increase by 28%, as indicated by our scenario analysis based on the average value of C sequestration from 2020-2060, with the recycling of crop residues and animal manure making the biggest contribution (Figure 5.5). However, soil C sequestration will decrease over time (Figure 5.5) and will thus be lower after 2060. To address long-term climate change, additional negative emission technologies, such as direct air C capture and storage, are required. In the optimal scenario (GMP + WR + STL + RL), net CO<sub>2</sub> emissions were negative, but net GHG emissions remained positive, although reduced by 93%, with the most important contribution from reducing livestock. Nevertheless, achieving global climate targets necessitates achieving net-zero GHG emissions (Rogelj *et al.*, 2018). In addition to the practices included in our

Optimizing carbon and nitrogen cycles towards net-zero emissions scenarios, clean energy may contribute to further reducing GHG emissions from the agrifood system (Lorenz and Lal, 2018; Shindell and Smith, 2019; Xing *et al.*, 2022). Greenhouse gas emissions from the energy and power consumption were 2.4 Mg CO<sub>2</sub> eq ha<sup>-1</sup> yr<sup>-1</sup> in the optimal scenario (Figure 5.5b). If these emissions could be reduced by 20% through the substitution of fossil energy with clean sources such as bioenergy, solar and wind power, the agrifood system in Quzhou would achieve net-zero GHG emissions, ensuring climate neutrality.

### 5.4.3. Implications of this study

This study demonstrates the technical potential of integrated practices to transition towards net-zero emissions in the agrifood system in Quzhou, China. In general, achieving net-zero GHG emissions in the agrifood system itself is challenging, and we cannot expect agrifood systems to offset GHG emissions in industry and other sectors. Agricultural production in Quzhou is representative for China, which is in transition from extensive low-efficient to intensive high-efficient and high-emission production (Chen et al., 2016; Bai et al., 2018; Wei et al., 2023). Such transformations are common in middle-income, rapidly-developing countries such as India, Indonesia, Brazil and Pakistan (FAO, 2022; OECD and FAO, 2022; Kang et al., 2023). These countries contribute to more than one-third of the global increase in food demand as well as relevant resource inputs and environmental emissions due to ongoing urbanization and changing diets with more animal products (UNEP, 2020; FAO, 2022; Kang et al., 2023). In addition, low-income regions (such as sub-Saharan Africa) are projected to experience similar transformations in the future (FAO, 2022; OECD and FAO, 2022), which may aggravate future climate change if not appropriately managed. The development paths of "emit first and treat later" followed by most high-income countries are not replicable for low- and middle-income countries in combating global warming. The options explored in this study provide insights for the global community on managing C cycles in the agrifood system and transitioning towards net-zero emissions.

The net-zero transition is far from easy and must be institutionally, economically and socially viable. Widespread knowledge transfer and technology extension services are necessary to diffuse low-emission technologies to all stakeholders within agrifood systems, such as the Science and Technology Backyard platform in China, which facilitates information exchange and innovation by incorporating agronomists, farmers, government and agri-businesses together (Zhang et al., 2016; Kang et al., 2023). Financial support is needed to credit agricultural C sequestration and incentivize low-emission management practices (Oldfield et al., 2022), and the credit requires accurate quantification of the amount of C sequestered using a widely accepted approach (Garsia

et al., 2023). Creating synergies between net-zero emissions and other policy goals could also promote the transition. For example, the advanced practices included in the scenarios of this study are also essential for N circularity in the agrifood system and N related environmental impact (Wei et al., 2023). Therefore, policies and incentives to promote circular agriculture and net-zero emissions are mutually reinforcing.

#### 5.4.4. Limitations of this study

In this study, crop production data were obtained from the latest available data (around 2020), while soil C content was evaluated from 2020 to 2060 using the Roth C model. When modelling the annual SOC change, crop production and relevant C input into soil were assumed to be sable, but they may actually increase when the soil becomes more fertile due to an increase in SOC. This aspect was not considered because of limited data. Population size was also assumed to be stable in our scenario analysis, which might be unrealistic between now and 2060, but the effect should also be minor because the household compartment contributed to the overall C flows and emissions in a limited manner (Figure 5.3) In addition, C flows in the food processing compartment were limited to maize and wheat milling due a lack of data. This underestimates the amount of C losses from the agrifood system, but the effect may be minor in our case area according to previous research (Wei et al., 2023). Furthermore, the proportions of different flows for human excreta were assumed to be the same as animal manure due to limited data, introducing some uncertainty. However, the impact is also minimal since the management of human excreta made a trivial contribution to net emissions, as indicated by the scenario analysis (compare the scenario GMP + WR1 and GMP + WR2). Finally, CO<sub>2</sub> and GHG emissions caused by biomass transport were not included in our analysis. When accounting for these emissions, achieving net-zero emissions in the agrifood system would be even more challenging.

### 5.5. Conclusions

This study quantified C cycles in an agrifood system by integrating a material and nutrient flow model with a soil C cycle model. Net emissions from the agrifood system were subsequently quantified using C sequestration against CO<sub>2</sub> emissions required for system functioning in Quzhou, China. Scenario analysis demonstrated the potential and options to achieve net-zero CO<sub>2</sub> and GHG emissions in the agrifood system.

The agrifood system in Quzhou is representative of China and many other developing countries. The amount of C sequestration in the system was 819 kg C ha<sup>-1</sup> yr<sup>-1</sup>, while CO<sub>2</sub> emissions from energy and power consumption required for system functioning were 847 kg C ha<sup>-1</sup> yr<sup>-1</sup>, indicating a net-positive CO<sub>2</sub> emission despite village-level variations. Although the system can generally transition to net-zero or even net-negative CO<sub>2</sub> emissions by adopting good management practices and recycling organic wastes, it can hardly transition to net-zero GHG emissions. Therefore, the agrifood system itself is a GHG source and could not offset emissions from industry and other sectors. In addition to promoting C sequestration in soils and reducing emissions, clean energy may be necessary to achieve net-zero GHG emissions and climate neutrality. The transition to net-zero GHG emissions warrants institutional, economic and social changes and incentives.

# Acknowledgements

We are grateful to Marjoleine Hanegraaf for assisting in using the RothC model, to Xiaoxi Xia for assisting in validating the RothC model, and to Lin Ma for granting permission to use and modify the NUFER model. This work was funded by the Sino-Dutch Agriculture Green Development Project of China Scholarship Council [No. 201913043] and Hainan University.

# **Supporting information**

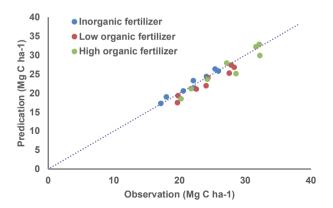


Figure S5.1. The observed and predicted soil carbon storage in Quzhou. The observed values are from a long-term experiment in Quzhou (Li et al., 2020); the predicated values are from the prediction of the RothC model. Different colors refer to fertilizer treatments, and dots in the same color indicate years from 2009 to 2016.

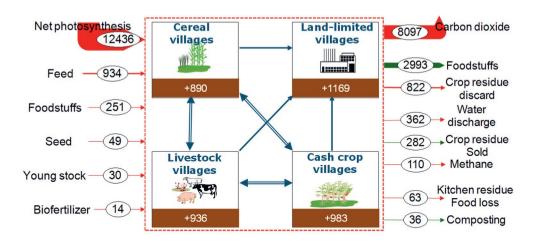


Figure S5.2. The C flows (kg C ha<sup>-1</sup> yr<sup>-1</sup>) in Quzhou under the scenario of good management practices (GMP). Values in the brown boxes indicate C accumulation in cropland. The dotted red line represents the boundary of the county. Arrows on the left and right side are input and output flows, respectively, with green arrows indicating C output in marketable products.

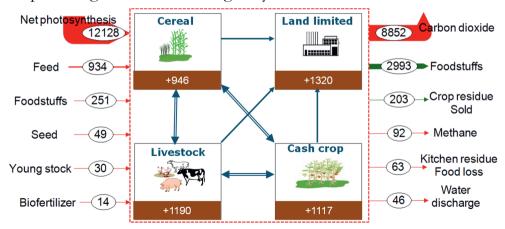


Figure S5.3. The C flows (kg C ha<sup>-1</sup> yr<sup>-1</sup>) in Quzhou under the scenario of GMP+WR1: same as the GMP, plus improved recycling of waste from crop and livestock production. Values in the brown boxes indicate C accumulation in cropland.

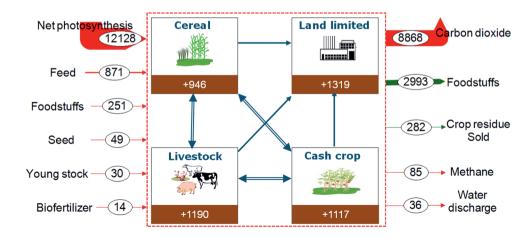


Figure S5.4. The C flows (kg C ha<sup>-1</sup> yr<sup>-1</sup>) in Quzhou under the scenario of GMP+WR2: same as the GMP+WR1, plus improved recycling of household waste. Values in the brown boxes indicate C accumulation in cropland.

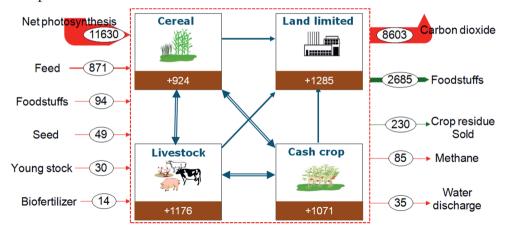


Figure S5.5. The C flows (kg C ha<sup>-1</sup> yr<sup>-1</sup>) in Quzhou under the scenario of GMP+WR+STL: same as the GMP+WR2, plus switching to legumes. Values in the brown boxes indicate C accumulation in cropland.

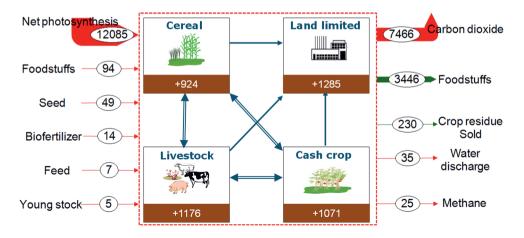


Figure S5.6. The C flows (kg C ha<sup>-1</sup> yr<sup>-1</sup>) in Quzhou under the scenario of GMP+WR+STL+RL: same as the GMP+WR+STL, plus livestock reduction. Values in the brown boxes indicate C accumulation in cropland.

Table S5.1. Carbon content of different materials in the agrifood system

ŀ	tems	C content (%)		ltems	C content (%)		Items	C content (%)
	rice	41.9		pig	33.0		Premix	38.3
	wheat	40.0		cow	27.6		Concentrates	38.3
	maize	39.6		poultry	17.4		Complete feed	38.3
	millet	40.9		Sheep/goat	27.6		maize	39.6
	sorghum	39.6		rabbit	27.6		Bran	38.3
Plant food	Other cereal	41.4	Animal food	egg	16.6	Feed	Soybean meal	40.9
	beans	46.0		milk	5.5		Plant residues	44.0
	peanut	54.6		fishery	27.6			
	vegetable	6.2		Ham	20.6			
	fruit	6.2		By product of livestock	26.3			

Note: the data are from Le Noe et al. (2017); Liang et al. (2019); Wen et al. (2021); Wolf et al. (2021).

Table S5.2. Parameters to evaluate the amount of animal manure and enteric methane emissions

Animal species	The ratio between manure and feed	Enteric methane emissions (kg CH <sub>4</sub> -C head <sup>-1</sup> yr <sup>-1</sup> )
Pig	0.29	0.75
Cattle	0.41	55.58
Sheep/goat	0.37	6.38
Broiler	0.34	0.00
Laying	0.30	0.00

Table S5.3. Parameters on CH<sub>4</sub> emissions during manure storage

Animal species	Pig	Cattle	Sheep/goat	Poultry	Laying
Value (kg CH₄-C head <sup>-1</sup> year <sup>-1</sup> )	4.91	11.25	1.67	0.01	0.01

Table S5.4. Annual agricultural inputs for crop production

Cropping system	Electricity (kWh ha <sup>-1</sup> )	Diesel (kg ha <sup>-1</sup> )	Plastic mulch (kg ha <sup>-1</sup> )	Coal (kg ha <sup>-1</sup> )
Wheat-maize	2078	241	0	0
Cotton	933	144	38	0
Vegetables	2100	74	88	0
Fruits	2100	74	88	0

Note: the data are from Guo et al. (2022)

Table S5.5. The emission factors of different agricultural inputs

Inputs	Electricity	Diesel	Plastic mulch	Coal
Unit	kg CO <sub>2</sub> -eq kWh <sup>-1</sup>	kg CO <sub>2</sub> -eq kg <sup>-1</sup>	kg CO <sub>2</sub> -eq kg <sup>-1</sup>	kg CO₂-eq kg <sup>-1</sup>
Value	1.08	3.19	19.04	2.65

Note: the data are from Guo et al. (2022); Xing et al. (2022)

Table S5.6. Annual inputs for livestock production

Animal species	Electricity (kWh head <sup>-1</sup> )	Diesel (kg head <sup>-1</sup> )	Plastic mulch (kg head <sup>-1</sup> )	Coal (kg head <sup>-1</sup> )
Pig	11.1	1.25	0	0.000
Cattle	255.68	27.43	0	3.085
Sheep/goat	5.21	0.61	0	0.000
Broiler	0.33	0.32	0	0.001
Laying	1.58	0.13	0	0.001

Note: the data are from Guo et al. (2022); Xing et al. (2022)

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# General discussion

Zhibiao Wei

### 6.1. Overview

The main aim of this thesis was to explore circularity options for N, C, and water in agrifood systems at the county scale to provide insight into the overall impact on circularity. We selected Quzhou county (China) as a case study. China is the largest emitter of reactive N (Chen et al., 2016) and greenhouse gases (UNEP, 2020), and it is also a hotspot of water scarcity (Rodell et al., 2018; van Vliet et al., 2021). Quzhou is a demonstration county for China's "Agricultural Green Development" strategy seeking to be a demonstration area for producing nutritious agricultural products while minimizing environmental impacts (MOA, 2020). This thesis quantified the current flows of N (Chapters 2 and 3), water (Chapter 4) and C (Chapter 5) within Quzhou. The current flows contributed to identifying key issues that limited N, water and C circularity in the agrifood system. Based on these key issues, this thesis explored options to transition towards a sustainable agrifood system by incorporating N circularity (Chapter 3), water security (Chapters 3 and 4) and climate neutrality (Chapters 3 and 5).

# 6.2. Main findings

The novel aspects of this thesis are

- An comprehensive analysis of nutrient flows in an entire agri-food system, considering not only crop and livestock production but also households and waste management
- An evaluation of promoting circularity at an optimal research scale
- An assessment of synergies and trade-offs in water security and N circularity in an agrifood system
- An exploration of advanced scenarios to achieve climate neutrality in an agrifood system, considering both N and C cycles

These aspects are further explained and discussed in the subsequent sections.

### 6.2.1. Nitrogen circularity in an entire agrifood system

Restoring N circularity requires quantifying nutrient flows at an optimal scale to describe the current state (Van der Wiel et al., 2020). To quantify N flows in the agrifood system, we firstly assessed the loss ratios of N leaching and runoff in response to various types of fertilizers. Compared with chemical fertilizers, organic fertilizers overall reduced the among of N leaching and runoff by 15% and 29%. The loss ratios of N leaching and runoff were 14% and 4.5%, respectively, from chemical fertilizer, and 9.2%

and 2.6%, respectively, from organic fertilizer (Chapter 2). Combining these loss ratios, farm surveys, statistical data, a nutrient flow model (NUFER) and parameters from literature review, we quantified N flows in the entire agrifood system, including households, in Quzhou. Household consumption data were collected in a year-round survey across the county. Nitrogen flows were generally linear in Quzhou's agrifood systems, as measured by the four selected KPIs, i.e. N import, N loss, N use efficiency (NUE) and N recycling rate (NRR). The KPIs are in line with the principles of circular agriculture. They enable transparency and comparability among different spatial regions and over time for measuring year-to-year progresses. The total N import was 546 kg ha<sup>-1</sup> yr<sup>-1</sup>, of which 54% was lost to the environment. The NUE of the agrifood system and NRR of excreta were both <30%. Excessive inputs, inefficient agricultural production and livestock manure management were key issues that limited circular N use (Chapter 3). Other research confirmed that agricultural production was inefficient, and N losses were too high to ensure safe air and groundwater quality in the same area (Meng *et al.*, 2022; Meng *et al.*, 2024).

To account for heterogeneities in N flows at the county level, this thesis grouped the 342 villages in Quzhou into four types: cereal villages, cash crop villages, livestock villages and land-limited villages. Each type of village was characterized by diverse production and consumption patterns as well as population densities. This approach reflects the differences in farming systems and management, and it proved to work well for villages participating in the Science and Technology Backyard platform in Quzhou (Zhang et al., 2016). At the village type level, total N import ranged from 365 kg N ha<sup>-1</sup> yr<sup>-1</sup> in cereal villages to 1015 kg N ha<sup>-1</sup> yr<sup>-1</sup> in livestock villages. Total N loss was also highest in livestock villages, i.e. 646 kg N ha<sup>-1</sup> yr<sup>-1</sup>, which was approximately three times the figure for cereal and cash crop villages. The NRR in the different village types was inversely related to their N loss, with cereal villages showing the highest NRR and livestock villages the lowest. Cereal villages also showed the highest NUE of the agrifood system (41%), compared to 32%, 26% and 17% in cash crop, livestock and land-limited villages, respectively (Chapter 3). The villages ranked in circularity performance as follows: cereal village > cash crop village > livestock village > landlimited village, as measured by the four KPIs.

This thesis investigated options to increase N circularity, including the application of good management practices in crop and animal production; household dietary change to more plant protein; recycling of organic waste; growing legumes instead of cotton; and reducing livestock breeding. All measures combined could roughly double the county-level N use efficiency and N recycling rate while reducing N loss by >60%. Among these measures, the recycling of organic waste was one of the most effective and feasible measures to move towards circularity. Forty percent of the increased NUE

was achieved with improved recycling of organic waste. Among the different sources of organic waste, the recycling of crop residues and livestock manure to local cropland had the greatest effect, increasing NUE from 47 to 61%. Unexpectedly, the recycling of human excreta and kitchen waste made a modest contribution (Chapter 3). A likely explanation is that Quzhou is an agricultural county, with cultivated land accounting for 78% of the total administrative area, where the effects of households on system N flows are small compared to the effects of agricultural production.

The application of good management practices resulted in significant reduction of both N import and N loss. Among these practices, optimizing fertilizer application and substituting organic for chemical N fertilizer in crop production synchronized N supply and crop N demand, thus reducing excessive N fertilizer input and the subsequent N losses (Cui et al., 2018; He et al., 2023). Dietary change to more plant and less livestock protein made a limited contribution to N circularity in Quzhou (Chapter 3). The main reason was that local meat consumption was already relatively low. In addition, the excessive livestock production in Quzhou was not reduced to ensure food and nutrition security of megacities surrounding Quzhou, such as Beijing which could not produce adequate food for self-sufficiency. When decreasing livestock production to adapt to the healthy diet, N circularity could be further improved because of fewer feed imports, fewer livestock product exports, and lower N emissions (Chapter 3). However, the reductions result in less available animal manure for crop production, necessitating increased import of chemical fertilizers to sustain optimal fertilizer levels. Furthermore, reducing livestock exports could jeopardize food security in neighbouring fooddeficient areas reliant on Quzhou's export-oriented produce. Quzhou exemplified the delicate relationship between N circularity and food trade.

# 6.2.2. Synergies in water security and N circularity in the agrifood system

This thesis evaluated water security elaborated in terms of water quantity and quality, for both surface water and groundwater, along with their potential synergies and trade-offs with N circularity in the case area of Quzhou, China. (Chapters 2, 3 and 4). Despite a rising water consumption, from 388 to 460 MCM between 2010 and 2020, Quzhou has halted and even reversed its declining groundwater level through measures such as water transfer, restricting groundwater extraction and seasonal land fallowing (Chapter 4). However, water transfer as a measure to increase local water availability might externalize water scarcity to water-exporting regions. Its contributions to water security need to be evaluated connecting causes and effects on a larger scale. Regarding water quality, the concentrations of most pollutants in groundwater were below critical values, while NH<sub>4</sub>-N concentration in surface water exceeded the safety threshold – although

its concentration had decreased by 87% over the past two decades. The downward trend of NH<sub>4</sub>-N concentrations in surface water could be attributed to reductions in drainage water with pollutants, thanks to more efficient agricultural water use (Chapter 4). This is inconsistent with other research in this area, which claimed that the NO<sub>3</sub>-N concentration in groundwater was also beyond the critical value according to their modelling (You *et al.*, 2023; Meng *et al.*, 2024). This discrepancy could be explained by the fact that they model the concentration of NO<sub>3</sub>-N in the recharge flows, whereas this study uses data from the direct measurement of water bodies. We believe that the NO<sub>3</sub>-N concentration in water bodies is more direct and accurate reflection of water quality compared to that in recharge flows.

Improved N management practices, such as optimized fertilization, could reduce N discharge from agrifood systems from 10,455 Mg N yr<sup>-1</sup> to 6,792 Mg N yr<sup>-1</sup> in Quzhou, decreasing the NH<sub>4</sub>-N concentration in surface water from 1.2 mg N l<sup>-1</sup> to 0.97 mg N l<sup>-1</sup>, which is below the critical threshold (1.0 mg N l<sup>-1</sup>). It could be further reduced to 0.69 mg N l<sup>-1</sup>, by implementing other practices that promote circular N use, such as recycling organic waste and reducing livestock production (Chapters 3 and 4). Synergies between achieving water security and N circularity were found in the agrifood system. Fewer recharge flows, due to less and more efficient agricultural water use, would result in lower discharge of N pollutants, bringing improved water quality.

#### 6.2.3. Climate neutrality and circularity in the agrifood system

This thesis quantified C cycles and net GHG emissions in the agrifood system of Quzhou, by integrating a modified material and nutrient flow model (NUFER) and a soil C cycle model (RothC). Net photosynthesis predominantly contributed to C input to the agrifood system, while soil respiration and microbial respiration during manure storage accounted for most of the C output at all village types. Net CO<sub>2</sub> emissions from the agrifood system in Quzhou were positive because the amount of C sequestration in soils (819 kg C ha<sup>-1</sup> yr<sup>-1</sup>) could not offset C emissions (847 kg C ha<sup>-1</sup> yr<sup>-1</sup>) from energy and power consumption required for system functioning (Chapter 5). The system could be transformed to net-zero CO<sub>2</sub> emissions by adopting good management practices, recycling organic wastes and reducing livestock production. Good management practices increased C sequestration in soils and reduced GHG emissions, including direct and indirect N<sub>2</sub>O emissions. Adopting these practices would reduce the net CO<sub>2</sub> emissions to -0.10 Mg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup>, although the net GHG emissions remained high (5.6 Mg CO<sub>2</sub> eq ha-1 yr-1) due to the global warming effects caused by CH<sub>4</sub> emissions and energy consumption. All measures combined reduced the net CO<sub>2</sub> emissions to -0.39 Mg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup>, but the net CHG emissions were still positive (0.49 Mg CO<sub>2</sub> eq ha<sup>-1</sup> yr<sup>-1</sup>). Overall, although the system can transition to net-zero or

even net-negative CO<sub>2</sub> emissions by adopting advanced management practices, the modelled scenarios do not lead to net-zero GHG emissions.

The agrifood system itself is a GHG source and could not offset emissions from industry and other sectors. In addition to promoting C sequestration in soils and reducing emissions, clean energy might be necessary to achieve net-zero GHG emissions and climate neutrality (Chapters 3 and 5). As far as we are aware, this is the first study quantifying C cycles in an entire agrifood system as well as its net CO<sub>2</sub> and GHG emissions.

# 6.3. Limitations of this study and future research

#### 6.3.1. Research scale and externalities

This work was conducted at the county scale to facilitate the implementation of circularity, because this scale encompasses the full agrifood system, and its various compartments of the agrifood system are spatially close enough to form a network in which agricultural products and nutrients can be easily exchanged. In addition, public agricultural extension services are generally organized at the county level. Therefore, this thesis only considered processes within the county. Emissions stemming from the production of energy and agrichemicals were excluded if they occurred outside the county. This externalizes emissions to agrichemical-producing regions. When incorporating the externalization, the emissions from the agrifood system would be higher, and the advanced scenarios to reduce external inputs would contribute more to mitigating emissions. Water transferred to the county from other regions was regarded as locally sustainable, whereas it carries the risk of externalizing water scarcity to water-exporting regions. Emissions caused by exported foodstuffs were included, which internalized emissions caused by consumption in other regions. The net effects of food-related material and nutrient trade on emissions were beyond the scope of this thesis.

Upscaling the identified optimal scale is vital to mitigate externalities and support policymaking. Achieving full circularity is impractical because local food production can only meet less than one-third of the global population's needs (Kinnunen *et al.*, 2020). Research conducted at the optimal scale will inevitably externalize emissions to agrichemical-producing regions while internalizing emissions resulting from consumption in other regions. Scaling up the research scope allows for encompassing these externalities and internalities, thereby balancing circularity at the optimal scale and agricultural trade on a broader scale. Research conducted at a larger scale can consequently provide insights for policymaking to advance sustainable circular agrifood systems, bridging the gap between large-scale policy formulation and fine-scale implementation.

While circularity is vital, local food production often falls short of meeting regional demands in various areas, necessitating food flows that balance surplus and deficit regions. Schulte-Uebbing et al. (2022) showed that without causing N pollution, America and Australia could produce over double their estimated minimum regional demand, Europe generally meets its needs, while Africa and Asia cannot meet regional demands. This implicates that food trade is essential to ensure every region staying within the N boundary. International food trade contributes to nutrient access and enables some poorer countries/less productive areas to nourish more people (Wood et al., 2018). However, this trade also shifts environmental and social impacts between production and consumption regions. An in-depth question is where crops and livestock should be produced, based on environmental boundaries or based on self-sufficiency. Further research is needed to quantify and minimize the production-, consumption- and trade-induced emissions. Striking a balance between circularity and agricultural trade at an appropriate scale is crucial for transitioning towards a sustainable future.

#### 6.3.2. C sequestration in soils

This thesis quantified net C emissions from the agrifood system by assessing C sequestration in soils against C emissions required for system functioning. Soil C storage and changes were modelled using a validated RothC model because the change of soil C storage is a slow process requiring long-term observations. The annual rate of change in SOC is affected by the soil's C sequestration potential due to C saturation (Lugato et al., 2014; Moinet et al., 2023). Carbon saturation in the RothC model is predicted based on soil physiochemical properties, including clay content (Coleman and Jenkinson, 2014; Garsia et al., 2023). However, Craig et al. (2021) have highlighted that biological mechanisms may also regulate C saturation patterns. Ecological constraints on microbial biomass, such as competition or predation, have been observed to potentially reduce the rate of mineral-associated SOC formation as soil carbon inputs increase (Craig et al., 2021; Islam et al., 2022). When incorporating such biological constraints, the soil would reach C saturation at a lower SOC content. Consequently, the amount of annual soil C sequestration would decrease compared with the value in this study. This will make it even harder to achieve climate neutrality in the agrifood system. An improved understanding of how both biotic and abiotic factors contribute to C saturation will enhance our ability to predict and manage the time-dynamic C sequestration in soils.

In addition, the model we utilized could only predict C sequestration in topsoil (0-23 cm), a limitation prevalent in most soil C cycle models, as subsoil measurements are difficult, time-consuming and labour-intensive. However, subsoil stores the majority of soil C (Chen *et al.*, 2023). The dynamics of SOC stock changes and their underlying

mechanisms may differ between the topsoil and the subsoil (Chen et al., 2022; Henneron et al., 2022). Plant C input predominantly drives topsoil C destabilization, whereas mineral protection by iron-aluminum oxides and cations plays a more critical role in preserving SOC in the subsoil (Chen et al., 2021). Therefore, processes influencing soil C sequestration markedly differ with soil depth. For example, no-tillage management increases surface SOC storage compared to conventional tillage but reduces it in deeper layers (Cai et al., 2022); more C inputs via plant roots elevate SOC in the topsoil, yet they accelerate subsoil C decomposition because subsoil C persistence results from its low energy quality coupled with inadequate energy supply from roots, and sufficient root energy supply alleviates the substantial energy limitations on decomposition (Henneron et al., 2022). Further research is imperative to delineate how management practices impact C sequestration in the subsoil, offering valuable insights for refining models to accurately predict C dynamics across the entire soil profile.

# 6.3.3. Interactions between N, water and C cycles, as well as interactions with phosphorus cycles.

This thesis intended to identify synergies and trade-offs between N, water and C cycles. However, certain nuanced relationships among these elements were not incorporated because of limited data. For example, waste recycling to cropland increased soil C sequestration (Chapter 5), while it may also improve soil water holding capacity (WHC). This improvement may increase water retention and reduce water infiltration and N discharge from soils (Bagnall et al., 2022). Notably, this specific process was not factored into the water balance analysis in Chapter 4 owing to conflicting views on its effects. Some studies asserted that management-induced changes to SOC substantially affected soil WHC (Bagnall et al., 2022), but others argued for a negligible effect (Minasny and McBratney, 2018a, b). It is therefore not possible to evaluate the potential effect of this interaction in this study. The discrepancy in these findings might stem from variations in how soil bulk density data were obtained. Soil bulk density depends on SOC content and soil management, and it affects soil WHC. Further research is imperative to elucidate the intricate connections between management-induced SOC changes, bulk density, and WHC. Such investigations will empower future models to illuminate scenarios in which changes in soil management, resulting in increased SOC, are likely to impact the water available to plants. In addition, they enable better estimation of hydrologic and nutrient flows, i.e., how management-induced SOC changes will affect water and nutrient discharges as well as water quality.

The following mechanisms were not considered in this thesis either due to limited data. In our scenarios, optimized fertilization includes reducing the amount of N fertilizer, which promotes the transfer of assimilated C from aboveground to belowground plant

biomass (Pausch and Kuzyakov, 2018). More belowground biomass means higher SOC formation efficiency (Villarino *et al.*, 2021), which will again affect water infiltration and discharge. Clarifying these mechanisms warrants more research.

The phosphorus (P) cycle was not incorporated in this thesis because it did not induce key environmental problems in the case area. Even in developed areas previously suffering from eutrophication caused by excessive nutrients in water bodies, the P inputs to water bodies have decreased because of the introduction of wastewater-treatment plants and P-free detergents (Tong et al., 2017; Ibáñez and Peñuelas, 2019). In contrast to P cycles, the imbalanced N/P ratios in water and atmosphere deserve more attention in the future. On one hand, the improvement in water quality due to P decline results in high N/P ratios, which triggers undesirable changes in the water ecosystems, such as the collapse of phytoplankton populations and the subsequent spread of macrophytes (Ibáñez and Peñuelas, 2019). On the other hand, the imbalanced atmospheric N and P deposition will change nutrient availability to plants, which may further influence the structure and function of terrestrial ecosystems (Zhu et al., 2016; Penuelas et al., 2020).

Although the P cycle was not analysed in this thesis, our scenario design indirectly reflected the importance of managing N/P ratios in agrifood systems. For instance, in the scenario outlining good management practices in crop production for N cycles (Chapter 3), we integrated the partial (rather than full) substitution of chemical fertilizers with organic ones based on the N application rate. This decision was made partly due to the higher N/P ratio in organic fertilizers compared to the demand for crops. A full substitution based on the N rate would lead to an excessive amount of P for crop production.

### 6.3.4. The scenario design on diet

This thesis regarded the EAT-Lancet diet (Willett et al., 2019) as a healthy and sustainable dietary choice, which is a flexitarian diet, rather than vegetarian or vegan. Vegetarian or vegan diets are often praised for their environmental friendliness because plant-based foods have lower environmental and social costs than livestock-based ones (Errickson et al., 2021; Xu et al., 2021; Herzon et al., 2023). However, plant-based foods lack many essential micronutrients such as calcium, vitamin B6 and B12 as well as omega-3 long-chain polyunsaturated fatty acids (Anonymous, 2019; Damerau et al., 2019; Mazac et al., 2022). People on a vegan diet typically require food supplements to obtain these nutrients. The production and transportation of these supplements may result in substantial environmental emissions. Consequently, when accounting life cycle

costs, the claim that vegetarian and vegan diets are more environmentally friendly might not hold true

Livestock serves multiple functions. Firstly, in many parts of the developing world, it functions as a form of savings or 'bank accounts' (Moyo and Swanepoel, 2010). Farmers sell livestock during the dry season when crop production is impossible, generating income to purchase daily necessities. Second, livestock acts as 'biological fermenters' that accelerate the degradation of crop residues into manure, making it more readily available for use in crop cultivation, particularly in areas where chemical fertilizers are insufficient (Moyo and Swanepoel, 2010; Bai *et al.*, 2018). Moreover, livestock plays a crucial role in converting biomass unsuitable for direct human consumption into food (Muscat et al., 2020; Herzon et al., 2023).

Therefore, a vegetarian or vegan diet may not be the panacea for the current environmental and social problems, although livestock-based foods in current quantities and qualities compromise the state of the environment, health and society (Errickson et al., 2021; Resare Sahlin and Trewern, 2022; Herzon et al., 2023). The key issue will be to accurately quantify the "less but better" meat consumption within the diet. A European study suggested modifying the EAT-Lancet diet to promote circularity in livestock production (van Selm et al., 2022). This is due to the discrepancy between the recommended consumption of livestock-based foods in the EAT-Lancet diet and the available quantities of by-products within the agrifood system and grassland resources. Recent studies are exploring ways to mitigate the environmental impact of food production by incorporating future foods into human diet, including earthworms, insects and algae (Mazac et al., 2022; Bai et al., 2023). However, the practical adoption of these future foods is limited by social and cultural acceptance barriers (Chaudhary and Krishna, 2019; Mazac et al., 2022). Further efforts are necessary to educate individuals on the similarities between these future foods and familiar ones, creating incentives for procurement by institutional and corporate food businesses. In addition, future research should delve into the factors driving changes in consumer behaviour and guide dietary shifts.

# 6.4. Implications for society and science

### 6.4.1. The optimal research scale for circularity

This project illustrated circularity options for N, C, and water in agrifood systems at a relevant scale. We propose that the optimal scale in China is at the county level, acknowledging potential variations in other countries. For example, in Germany, a district might be a suitable scale (van der Wiel *et al.*, 2021). In the US, a potential scale may be the proposed "manureshed" which refers to the lands surrounding livestock

production where manure nutrients can be recycled to meet environmental, production, and economic goals (Spiegal et al., 2020). The manureshed could be managed among livestock farms and crop farms within a county or even among adjacent counties. In this study, the county scale worked well. It encompasses the entire agrifood system, and all livestock manure and human excreta produced in Quhzou can be recycled as organic fertilizer for cropland within and among Quzhou's villages, which will result in an organic substitution rate of approximately 55% of total fertilizer inputs into cropland. Additionally, the scale incorporates public agricultural extension services and farm recommendations. Moreover, this scale serves as the fundamental unit for implementing agricultural policies in China. These criteria could assist other countries in identifying the ideal research scale for promoting circularity.

Downscaling the identified optimal scale is crucial to recognize the underlying heterogeneity in nutrient flows. To accurately quantify nutrient flows at the county level, it's imperative to comprehend the underlying diversity of villages with different types of production and consumption patterns as well as population densities. Attempting to quantify nutrient flows within and among each village is unfeasible. In this thesis, the 342 villages in Quzhou were grouped into four types: cereal villages, cash crop villages, livestock villages and land-limited villages (Chapters 3 and 5). This division aims to provide a snapshot of villages with shared characteristics, aiding in understanding the diverse production and consumption patterns influencing nutrient flows within the agrifood system. This approach worked well in identifying key limitations and exploring options for N circularity and C neutrality. While not universally prescriptive, this classification sheds light on characterizing various production and consumption patterns for further research.

### 6.4.2. The role of agrifood systems in combating climate change

According to this research, even in an optimized scenario (Chapter 5), the agrifood system remains a source of GHG emissions. This implicates that the agrifood system should not be expected to compensate for emissions originating from industry and other sectors. Alongside promoting C sequestration in soils and emission reduction, adopting clean energy sources can further contribute to curbing GHG emissions within the agrifood system. Bioenergy with Carbon Capture and Storage (BECCS) stands out as a critical clean energy solution (Xu et al., 2022). However, widespread implementation of BECCS relies on an abundant biomass supply, which could exert considerable pressure on terrestrial biospheres, freshwater, and land use (Cobo et al., 2022). In contrast, Direct Air Carbon Capture and Storage emerges as an environmentally favourable technology due to its lower environmental impacts and its ability to prevent adverse effects on terrestrial biospheres (Cobo et al., 2022), although the current high

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cost impedes the widespread use of this technology. In general, achieving climate neutrality demands both transformation in the food system, the reduction of fossil fuel use and the integration of negative emissions technologies. This transition necessitates institutional, economic, and social changes, as well as incentives.

### 6.4.3. Creating synergies among policy goals in promoting circularity

The advanced measures explored in our scenarios could simultaneously promote N circularity, water security and climate neutrality (Chapters 3, 4 and 5). For example, recycling waste for agricultural use aids in restoring circularity, concurrently enhancing soil C sequestration and mitigating water pollution by reducing N discharge into water bodies. Furthermore, effectively managing waste, particularly human faeces, plays a crucial role in sustainable development through improved sanitation. It's important to note that untreated faecal matter contributed to over 870,000 sanitation-related deaths in 2016 (McNicol *et al.*, 2020). These interdependencies highlight how policies and incentives aimed at fostering circularity, climate neutrality, clean water and sanitation can mutually reinforce each other. Facilitating synergies among policies requires collaborative efforts across multiple sectors and actors in the policymaking process.

### 6.4.4. Fostering dietary changes

The world grapples with a triple burden of malnutrition: overnutrition (leading to overweight), undernutrition (resulting in underweight), and micronutrient deficiencies (often termed 'hidden hunger') (Anonymous, 2023; Wang et al., 2023). These nutrition crises are symptomatic of the systemic issues within our global food system, posing significant public health challenges. Specifically, the diet in the case area includes too many refined grains (carbohydrates) and few beans (Chapter 3), potentially contributing to conditions like diabetes (Sheng et al., 2021). A western diet high in red meat correlates with cardiovascular diseases and strokes due to the elevated ratio of saturated to polyunsaturated fats and the presence of heat-induced carcinogens in red meat, in contrast to plant-based protein sources (Willett et al., 2019). Fostering dietary changes is crucial in addressing environmental and healthy burdens.

The EAT-Lancet diet serves as a model for a sustainable and healthy diet. However, it is not universally prescriptive, and individual countries may need to tailor their recommended diets based on local cultural and social conditions. Research confirms that the necessary adjustments in the intake amounts of food items are highly country-specific in achieving sustainable diets (Chaudhary and Krishna, 2019). Encouraging changes in consumer behaviour and dietary patterns requires concerted efforts.

### 6.5. Concluding remarks

This thesis aimed to explore circularity options for N, C, and water in agrifood systems at an optimal scale, with Quzhou county (China) selected as the case study. Nitrogen leaching and runoff from cropland are important pathways for N losses, and they also influence water security in agrifood systems. Calibrating the loss ratios of leaching and runoff for various types of fertilizers is essential before quantifying nutrient flows.

This thesis identifies synergies in N circularity, water security and C neutrality. Fewer recharge flows, due to less and more efficient agricultural water use, would result in lower discharge of N pollutants. The agrifood system itself is a GHG source and could not offset emissions from industry and other sectors. It is possible to transition towards a N-circular, water-secure and C-neutral agrifood system, but achieving climate neutrality requires both a transformation in the food system and the integration of negative emissions technologies, alongside clean energy sources. Such transition warrants technological, institutional, economic and social changes and incentives.

While circularity is crucial, achieving full circularity is impractical because local food production falls short of meeting regional demands in various areas. Striking a balance between circularity and agricultural trade at an optimal scale is pivotal for transitioning towards a sustainable future. Multi-scale research plays a key role in advancing sustainable circular agrifood systems by bridging the gap between large-scale policy formulation and fine-scale implementation. Encouraging dietary changes is essential in tackling environmental and health challenges. Future research should delve deeper into the factors that drive changes in consumer behaviour and guide dietary shifts.

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### **Summary**

Linear and inefficient agrifood systems result in substantial losses of nitrogen (N), carbon (C) and water to the environment, causing severe environmental and social challenges. Addressing these challenges requires closing the loops of N, C, and water within the agrifood system, encompassing the compartments of crop production, livestock production, food processing, household consumption and waste management. However, the interplay between N, C, and water introduces complexities. How to close N, C and water cycles simultaneously is unclear, particularly at a system level involving households. Furthermore, promoting circularity necessitates selecting the most relevant spatial scale. The area should be sufficiently small to capture the local circumstances, enable transport and thereby facilitate nutrient exchange, but also large enough to include all components of the agrifood system. Field and farm scales are too small as they exclude practices to promote nutrient cycling among various farming systems. National scale is generally too large, obscuring underlying heterogeneity and hindering the optimization of nutrient cycles on smaller scales. A district or county meets these requirements and may be a suitable scale.

This thesis aimed to explore circularity options for N, C, and water in agrifood systems at the county scale. We selected Quzhou county (China) as a case study. China is the largest emitter of greenhouse gases and pledged to achieve carbon neutrality by 2060. Quzhou is a demonstration county for China's "Agricultural Green Development" strategy seeking to be a demonstration area for producing nutritious agricultural products while minimizing environmental impacts. The specific research objectives were: 1) to quantify the loss ratios of N leaching and runoff in croplands and assess N flows in an entire agrifood system; 2) to evaluate the impact of available options to promote N circularity and their synergistic effects; 3) to assess water security in an agrifood system, elaborated in terms of water quantity and quality, along with their potential synergies and trade-offs with N circularity; 4) to explore potential for achieving net-zero GHG emissions (climate neutrality) in an agrifood system. These objectives were addressed in Chapter 2-5, in addition to a general introduction (Chapter 1) and a general discussion (Chapter 6).

Chapter 2 quantified the loss ratios of N leaching and runoff from cropland. The types of fertilizer affected N leaching and runoff. Compared with chemical fertilizers, organic fertilizers overall reduce the among of N leaching and runoff by 15% and 29%. The loss ratios of N leaching and runoff were 14% and 4.5%, respectively, from chemical fertilizer, and 9.2% and 2.6%, respectively, from organic fertilizer. Application approaches of organic inputs significantly affected N leaching and runoff. Partial

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substitution of chemical fertilizers by organic fertilizers with the same amount of total N generally reduces leaching and runoff without compromising crop yield. The optimal substitution rates differed between leaching (40–60%) and runoff (60–100%) when substitution was based on equal amounts of total N. This work emphasizes the importance to calibrate the loss ratios of leaching and runoff for different types of fertilizers.

Combining the above loss ratios, farm surveys, statistical data, a nutrient flow model (NUFER) and parameters from literature review, Chapter 3 quantified N flows in a complete agrifood system, including households, at both the village and county level. Nitrogen flows were generally linear in Quzhou's agrifood systems, as measured by the four selected key performance indicators (KPIs), i.e. N import, N loss, N use efficiency (NUE) and N recycling rate (NRR). The total N import was 546 kg ha<sup>-1</sup> yr<sup>-1</sup>, of which 54% was lost to the environment. The NUE of the agrifood system and NRR of excreta were both <30%. Excessive inputs, inefficient agricultural production and livestock manure management were key issues that limited circular N use. To account for heterogeneities in N flows at the county level, this chapter grouped the 342 villages in Quzhou into four types: cereal villages, cash crop villages, livestock villages and land-limited villages. The villages ranked in circularity performance as follows: cereal village > cash crop village > livestock village > land-limited village, as measured by the four KPIs.

Based on the N cycles, Chapter 3 further investigated options to increase N circularity, including application of good management practices in crop and animal production; household dietary change to more plant protein; recycling of organic waste; growing legumes instead of cotton; and reducing livestock breeding. All measures combined increased the system's NUE by 172% and NRR by 87%, while reducing N import by 68% and N loss by 77%. Among these measures, the recycling of organic waste contributed most to N circularity. Among the different sources of organic waste, recycling of crop residues and livestock manure to local cropland had the greatest effect, increasing NUE from 47 to 61%. Unlike our expectation, recycling of human excreta made a modest contribution (increasing NUE from 61 to 67%). A likely explanation is that Quzhou is an agricultural county, with cultivated land accounting for 78% of the total administrative area, where the effects of households on system N flows are small compared to the effects of agricultural production.

The application of good management practices resulted in the significant reduction of both N import and N loss. Among these practices, optimizing fertilizer application and substituting organic for chemical N fertilizer in crop production synchronized N supply and crop N demand, thus reducing excessive N fertilizer input and the subsequent N

losses. Dietary change to more plant and less livestock protein made a limited contribution to N circularity in Quzhou. The main reason was that local meat consumption was already relatively low. In addition, the excessive livestock production in Quzhou was not reduced to ensure food and nutrition security of megacities surrounding Quzhou, such as Beijing where could not produce adequate food for self-sufficiency. When decreasing livestock production to adapt to the healthy diet, N circularity could be further improved because of less feed imports, less livestock products exports, and lower N emissions. However, this reduction results in less available animal manure for crop production, necessitating increased import of chemical fertilizers to sustain optimal fertilizer levels.

Chapter 4 evaluated the temporal dynamics of water quantity and quality, for both surface water and groundwater, as well as interactions between water security and N circularity in the case area of Quzhou, China. Despite a rise in water consumption, from 388 MCM in 2010 to 460 MCM in 2020, Quzhou has halted and even reversed its declining groundwater level through measures such as water transfer, restricting groundwater extraction and seasonal land fallowing. However, water transfer as a measure to increase local water availability might externalize water scarcity to water-exporting regions. Its contributions to water security need to be evaluated connecting causes and effects on a larger scale. Regarding water quality, pollutants in groundwater was generally below critical values, while NH<sub>4</sub>-N in surface water exceeded the safety threshold – although its concentration had decreased by 87% over the past two decades. The downward trend of NH<sub>4</sub>-N concentrations in surface water could be attributed to reductions in drainage water with pollutants, thanks to more efficient agricultural water use.

Improved N management practices, such as optimized fertilization, could reduce N discharge from agrifood systems from 10,455 Mg N yr<sup>-1</sup> to 6,792 Mg N yr<sup>-1</sup>, decreasing the NH<sub>4</sub>-N concentration in surface water from 1.2 mg N l<sup>-1</sup> to 0.97 mg N l<sup>-1</sup>, which is below the critical threshold (1.0 mg N l<sup>-1</sup>). It could be further reduced to 0.69 mg N l<sup>-1</sup>, by implementing other practices that promote circular N use, such as recycling organic waste and reducing livestock production. Synergies between achieving water security and N circularity were found in the agrifood system. Less recharge flows, due to less and more efficient agricultural water use, would result in lower discharge of N pollutants, bringing improved water quality.

Chapter 5 explore potential for achieving net-zero GHG emissions (climate neutrality) in an agrifood system. It quantified C cycles and GHG emissions in the agrifood system of Quzhou, by integrating a modified material and nutrient flow model (NUFER) and a soil C cycle model (RothC). Net photosynthesis predominantly contributed to C input

### Summary

to the agrifood system, while soil respiration and microbial respiration during manure storage accounted for most of the C output at all village types. Net CO<sub>2</sub> emissions from the agrifood system in Quzhou were positive because the amount of C sequestration in soils (819 kg C ha<sup>-1</sup> yr<sup>-1</sup>) could not offset CO<sub>2</sub> emissions (847 kg C ha<sup>-1</sup> yr<sup>-1</sup>) from energy and power, despite village-level variations with net-negative emissions at cereal and cash crop villages but net-positive emissions at livestock and land-limited villages.

The system could be transformed to net-zero CO<sub>2</sub> emissions by adopting good management practices, recycling organic wastes and reducing livestock production. Good management practices increased C sequestration in soils and reduced GHG emissions, including direct and indirect N<sub>2</sub>O emissions. Adopting these practices would reduce the net CO<sub>2</sub> emissions to -0.10 Mg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup>, although the net GHG emissions remained high (5.6 Mg CO<sub>2</sub> eq ha<sup>-1</sup> yr<sup>-1</sup>) due to the global warming effects caused by CH<sub>4</sub> emissions and energy consumption. All measures combined reduced the net CO<sub>2</sub> emissions to -0.39 Mg CO<sub>2</sub>-C ha<sup>-1</sup> yr<sup>-1</sup>, but the net GHG emissions were still positive (0.49 Mg CO<sub>2</sub> eq ha<sup>-1</sup> yr<sup>-1</sup>). Thus, the agrifood system itself is a GHG source and could not offset emissions from industry and other sectors. In addition to promoting C sequestration in soils and reducing emissions, clean energy might be necessary to achieve net-zero GHG emissions.

The results demonstrate the possibility of transitioning towards a N-circular, water-secure, and C-neutral agrifood system. The recycling of organic waste is one of the most effective and feasible measures to move towards circularity. Less recharge flows, due to less and more efficient agricultural water use, would result in lower discharge of N pollutants. This is not a trade-off situation between N and water cycles but a clear win-win. In addition, synergies in N circularity and C neutrality are also identified. Although the system can transition to net-zero or even net-negative CO<sub>2</sub> emissions by adopting advanced management practices, the modelled scenarios do not lead to net-zero GHG emissions. Thus, the agrifood system itself is a GHG source and could not offset emissions from industry and other sectors. Balancing circularity and agricultural trade to achieve sustainability at different scales will be the greatest scientific challenge.

### **Acknowledgements**

My PhD journey has come to an end after four and a half years. I sometimes felt the journey was so long, but now have the same old sentiment: how time flies. When looking back on the journey, I want to express my deep gratitude to the many people who have brought me to this point.

Ellis. You have always been there behind me when I steer the wheel of my PhD research. During the first year and a half of my PhD study, I had to remain in China due to project requirements and Covid regulations. At that time, we had to communicate and discuss online, which was not very efficient due to unclear training goals for PhD study and different working habits between Chinese and Dutch. After a period of struggling, I got used to the goal and way of being an independent researcher. My PhD project have gone smoothly (I personal think) since then. We supervised an MSc study together despite spending longer time than usual. This experience prompts me to reflect on the meaning of teaching and supervising. It helps pave the way for my future career in teaching and educating. I am happy and satisfied with who I am now and many thanks for your supervision and help.

Zhenling. Thank you so much for your support, supervision and subsidy when I was in China. My personal situation was bad during that period owing to emotional problems, miscommunication with supervisors and Covid regulations. You supported me to go to Quzhou and do my own research (partly because there was no Covid regulations there at that time). In Quzhou, I was able to live together with many friends and colleagues, which made me feel better thereafter. I published my first paper in the first year of my PhD study under your supervision. Through this accomplishment, I experienced a sense of fulfilment, which meant a lot to me considering my struggling situation at that time.

Petra. I appreciate having you as one of my promoters. You introduced me to your research group (Water Resources Management). I had very pleasant experiences with the group drinking and writing days. Before embarking on this PhD project, I didn't have much experience with water cycles. You answered many of my "stupid" questions. For example, why water scarcity is aggravated despite water not disappearing at the global level. Whenever I encountered problems with water management, you were available to help me directly or to indicate whom I could consult for assistance. I have learned the most in the chapter on water, and I hope to continue learning and growing throughout my future career, as we have done together.

Minghao. Thank you for being a part of the supervision team for my PhD project. Appreciate that you are always available for me as daily supervisor. Your careful explanations and modifications have greatly assisted me in completing all the chapters

### Acknowledgements

of my thesis. Your wealth of interesting ideas is inspiring. Hope to have the opportunity to collaborate on more innovative projects with you in the future.

I would like to thank the assistance from Marnella, Susy, Jingmeng, Ying Zhang, Claudius, Peter and Yiyan. Marnella, you picked me up on that sunny morning and introduced the WUR and SBL to me on April 6th 2021. That's where my career at WUR started. Thank you so much for helping me settling down and get used to the life in Wageningen. Susy, your work as a secretary enables my project to go well and makes my life in Wageningen easier. Jingmeng and Ying, I am grateful for your assistance with my PhD project from the Chinese side. Claudius and Peter, you introduced how a PhD student take leadership of their own project. Thanks to Yiyan's assistance, my work went well at China Agricultural University.

Special thanks to Tao/Benz, Yifei, Guichao/Dazhanglang. Yifei, we came together to the Netherlands three years ago and lived quite near in Wageningen. It was so good to have you around me in such an unfamiliar environment and culture. Benz, I would not forget that you picked me up from Haarweg to your home and served me my first meal in the Netherlands. We had excellent travels to Switzerland, Germany, Czechia, Slovakia, Hungary, Austria, Belgium, Luxembourg. Dazhanglang, my leisure time in Wageningen became more enriched after you arrived – playing poker and majiang, picking "gems" alongside the Rine river, gathering wild cherries, chives, walnuts, chestnuts and blackberries. Most of my free time in Wageningen revolved around you three during the first year and later until you gradually returned to China. The experiences shared with you are indelible to me.

I want to show my deep gratitude to Yizan, Laiquan, Jinghan, Muying, Zhengyuan. It's great to have you Yizan in the same research group for work and as a neighbour for life. Laiquan, you have helped me a lot with daily life and have always been a reliable hand for mechanical-related problems. The time we spent as roommates is memorable. Next is "the trio at Bornsesteeg" (although only Zhengyuan is still living at Bornsesteeg now ahaha). Jinghan, our pastry master, thank you so much for your treats. Muying, our tour guide, you are a 100% reliable traveling companion. Zhengyuan, our talk show host, it is wonderful that you have been living nearby the campus, and I know whom to ask for help whenever I need assistance at the campus.

I also want to thank the help and care from my "classmates" and "bosses" in the Agricultural Green Development project: Jinghan, Yu, Zhengyuan, Muying, Jie, Yanan, Qi, Zhan, Hongyu, Jiali, Shiyi, Mingzhao. We have been at the same pace and always have a common language to share and talk. Wish you all have a bright and happy future.

Many thanks to Ling, Siwen, Haixing and Juju (my cruise team to Mediterranean). Juju, although you actually were not together with us in that trip, I felt you were there because you are a binding companion with them in my view:). The marvellous trip together with you has been caved in my mind. Siwen, a (former) social queen in Wageningen, the legend about you continues although you have returned to China. Haixing, have fun with disco dancing. Ling, you are always considerate and ready for help. Special thanks for offering me the quilt this time when I came back to the Netherlands, helping me avoid using the sky as a blanket~. You guys have offered indispensable company for me for the past year and a half.

Thank you to my officemates, Mirjam, Meixiu, Hannah, Anna-Reetta, Jiyu, Margot, Shevani, Erne, and Zhongchen. It is your companionship that makes my life and work in Wageningen colorful and abundant. Mirjam, especially, you are so accommodating, teaching me Dutch, playing pingpang together and helping me formatting my thesis.

My gratitude to people in the SBL family, including but not limited to Laura, Gidena, Omotola, Mubonda, Angel, Erne, Tullia, Karen, Luuk, Enno, Chindi, Doina, Bobby, Felix, Leila, Frank, Selene, Howard, Maria, Michiel, Rachel, Gerlinde, Ron, Jan-willem, Marie, Gabriel, Paolo, Giulia, Carmen, Alix, Nikos.

Thanks to our badminton group: Laiquan, Yuxiang, Yizan, Zhenpeng, Zhaoqi, Jiyu, Dongdong, Junhan, Jiawen, Siwen, Ruolin, Bowen, Yujuan, Yalin Zulin, Maric-Chaclolle and many others.

Thanks to people in the AGD programme: Zhilong, Fanlei, Yaowen, Donghao, Weitong, Xiaoxi Guo, Luncheng, Lu Liu, Dongfang, Mingyu, Xueyuan, Jianan and others.

Thanks to Hao Yu, Xu Han, Yue Wang, Meijun, Xiaoxi Xia, Xuenan, Ke Peng, Dailing Jing Zhang, Shan Jiang, Xingjuan, Chen Wang, Yingcheng, Ye Liu, Yalan, Zhixiang, Oene and many others for enriching my life in Wageningen and CAU.

My gratitude extends to the people in the WRM group: Xiulu, Hui, Muying and Jiawen, Matthijs, Roze, Jidapa and others.

Thanks to my nice roommates: Noortje, Tim, Sophia, Maaike, Leonoor, Bleike and Marieke. I appreciate sharing the pleasant living space with you.

I also want to thank the students working in STB who assisted me in conducting my survey in Quzhou, especially Chen Yang and Junna. You two offered me a room to live at the apple village (Xianggongzhuang). This was a great help to my work because I needed to visit farmers every day at that time. I miss the time living and eating together with you.

### Acknowledgements

Last, my parents and brother. 感谢父母最无私的关爱。出国三年,去年回家才知道你们竟然自认为没能在人生道路上给我足够的帮助,甚至为此自责。我深感不解与愧疚,常年在外未能及时沟通感情,你们无私的爱和支持已是最大的帮助。希望以后常伴左右,我也能成为你们的骄傲和依靠。感谢弟弟付出,万幸有你常在爸妈身边。

My PhD journey is also a process of learning and growth. I have reached this point relying on the collective contributions of the individuals mentioned as well as many others who remain unnamed. I would like to show my sincerest thanks to all of you once again. Hope that flowers bloom along the way in the future.

Zhibiao Wei Wageningen, the Netherlands March, 2024

### About the author

Zhibiao Wei was born in Shangqiu City, Henan Province, China, on the 19th of November 1993. As the son of a traditional Chinese farmer, Zhibiao has always cared about food and the land. He began his bachelor's degree majoring in Agricultural Resources Environment Huazhong Agricultural University in 2011. After a four-year study programme, he obtained the BSc degree and started pursuing his MSc degree at China Agricultural University. He modelled nutrient flows and losses in Chinese forage production system at the provincial level during his master's studies under the supervision of Prof. Fusuo Zhang and Prof. Lin Ma. After graduation in



2017, he decided to do more practical works apart from modelling. He worked (as research assistant) on fields and assisted in managing a long-term experiment on grassgrain rotations, exploring ways to abate NH<sub>3</sub> and GHG emissions. In 2019, he obtained an opportunity to do PhD research in Wageningen University & Research, funded by the Sino-Dutch Agriculture Green Development Project of China Scholarship Council. Since then, he has been doing an interdisciplinary project focusing on a sustainable circular agrifood system. This involves exploring circularity options for nitrogen, carbon, and water in agrifood systems at an optimal scale. Zhibiao likes to play badminton, cook and try new things in his free time.

### List of publication

- Wei Z. B., Ying H., Guo X.W., Zhuang M.H., Cui Z.L., Zhang F.S., Substitution of mineral fertilizer with organic fertilizer in maize systems: A meta-analysis of reduced nitrogen and carbon emissions. Agronomy, 2020.
- Wei Z. B., Hoffland E., Zhuang M.H., Hellegers P., Cui Z.L., Organic inputs to reduce nitrogen export via leaching and runoff: A global meta-analysis. Environmental Pollution. 2021
- Wei Z. B., Zhuang M.H., Hellegers P., Cui Z.L., Hoffland E., Towards circular nitrogen use in the agri-food system at village and county level in China. Agricultural Systems. 2023
- Wei Z. B., Xu W., Zhuang M.H., Hoffland E., Cui Z.L., Hellegers P., Synergies in water security and nitrogen circularity: a case study in Quzhou, China. To be resubmitted
- Shao H., Wu X.B., Chi H.H., Zhu F.B., Liu J.H., Duan J.H., Shi W.J., Xu Y., **Wei Z. B.\***, Mi G.H.\*, How does increasing planting density regulate biomass production, allocation and remobilization of maize temporally and spatially: A global meta-analysis. Under review
- Wang C., Miao Q., **Wei Z. B.**, Guo Y. X., Li J. Y., Fan Z. Y., Hu Y. X., Zhang H., Sun J. W., Cui Z. L., Nutrient runoff and leaching under various fertilizer treatments and pedogeographic conditions: A case study in tobacco (Nicotiana tabacum L.) fields of the Erhai Lake basin, China. European Journal of Agronomy. 2024
- **Wei Z. B.**, Zhuang M.H., Hellegers P., Cui Z.L., Hoffland E., Optimizing carbon and nitrogen cycles towards net-zero greenhouse gas emissions in agrifood systems: a case study in Quzhou, China. To be submitted.

# PE&RC Training and Education Statement

With the training and education activities listed below the PhD candidate has complied with the requirements set by the C.T. de Wit Graduate School for Production Ecology and Resource Conservation (PE&RC) which comprises of a minimum total of 32 ECTS (= 22 weeks of activities)



### Review/project proposal (4.5 ECTS)

- From waste to crop: closing N, C and water cycles in Quzhou County, Hebei province, China

#### Post-graduate courses (14.9 ECTS)

- Agricultural green development; CAU & WUR (2019)
- Theories of system analysis; CAU (2019)
- Introduction to R & Rstudio; PE&RC (2021)
- Life cycle assessment modelling of solid waste systems-application of the EASETECH model: Technical University of Demark (2021)
- Grasping sustainability; SENSE (2022)
- Basic statistics; PE&RC (2022)
- Statistical uncertainty analysis of dynamic models; PE&RC and WIMEK (2022)
- Farming systems and rural livelihoods analysis: PE&RC and WASS (2023)

#### Deficiency, refresh, brush-up courses (1.5 ECTS)

- Grammar & vocabulary; Wageningen in to Languages (2021)

#### Invited review of journal manuscripts (2 ECTS)

- Plant and soil: enhancing soil nitrogen supply and maintaining rice yield through partial replacement of chemical nitrogen with food waste-derived organic fertilizer (2023)
- Scientific reports: nitrogen, phosphorus and potassium budget in crop production in South-Asia: regional and country trends during the last five decades (2023)

#### Competence, skills and career-oriented activities (3.9 ECTS)

- Personal leadership the basis of effective and efficient academic development; PE&RC (2019)
- Scientific writing: Wageningen in to Languages (2020)
- How to supervise a MSc student; SBL, PE&RC (2021)

#### Scientific integrity/ethics in science activities (0.6 ECTS)

### PE&RC Training and Education Statement

Scientific integrity: WGS (2021)

# PE&RC Annual meetings, seminars and PE&RC weekend/retreat (1.5 ECTS)

- PE&RC Workshop carousel (2021)
- PE&RC Midterm retreat (2021)
- PE&RC Last year retreat (2023)

### Discussion groups/local seminars or scientific meetings (8.65 ECTS)

- The annual AGD meeting (2019-2023)
- Workshop on modelling N and water flows (2021)
- Soil on 1 meeting (2022)
- Discussion group of sustainable intensification of agricultural systems (2022-2023)
- Discussion group of emissions and nutrient management (2022-2023)

### International symposia, workshops and conferences (4.1 ECTS)

- CIRCULAR@WUR conference; Wageningen (2022)
- Wageningen soil conference; Wageningen (2023)

### BSc/MSc thesis supervision (3 ECTS)

- Validation of the RothC model to the results of long-term experiments on the dryland of China

### Colofon

# **Funding**

The research described in this thesis was financially supported by the Sino-Dutch Agriculture Green Development Project of China Scholarship Council (No. 201913043) and Hainan University.

Cover design: Sen Li and Zhibiao Wei Printed by ProefschriftMaken