THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Assessing reactive nitrogen flows in European agricultural systems

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ABSTRACT

In most ecosystems, nitrogen is a scarce and growth-limiting nutrient. Natural and anthropogenic processes convert unreactive atmospheric dinitrogen (N_2) into reactive, bioavailable, nitrogen. This conversion occurs mainly through biological nitrogen fixation (BNF), industrial ammonia (NH_3) synthesis, and oxidation of N_2 into nitrogen oxides (NO_x) during combustion of fossil fuels. The rate of anthropogenic conversion of N_2 into reactive nitrogen has increased dramatically in the last century and today roughly equals the rate of all natural processes combined. The most important benefit is that NH_3-derived fertilizer and BNF improve crop yields in agriculture. However, anthropogenic activities have also increased the turnover of reactive nitrogen in the environment, with unintended consequences for human well-being and ecosystem functioning. NO_x and NH_3 in the atmosphere contribute to smog and related health problems. Emissions of nitrous oxide (N_2O) contribute to global warming. Nitrogen fertilizes and acidifies terrestrial and aquatic ecosystems, sometimes with dramatic effects on biodiversity.

This thesis makes two types of contributions to the understanding of reactive nitrogen flows in European agricultural systems. First, it estimates and analyzes nitrogen flows with a special focus on livestock production and on uncertainties in agricultural nitrogen budgets, with results that can be used to assess the effects of potential changes to the food system. A comparison of organic and conventional milk production systems shows that organic milk is likely associated with less nitrogen losses to the environment, but this conclusion may change with improved BNF estimates. Second, the thesis analyzes and discusses three quantitative indicators to assess nitrogen losses associated with food products or production systems, with focus on system boundaries, uncertainties and environmental relevance. The indicators have limited environmental relevance since they aggregate nitrogen flows over time, space, and of different chemical forms. This analysis contributes to the understanding of how nitrogen indicators can be used for research and communication with the public and policy-makers.

Keywords: nitrogen, agriculture, food, nitrogen budget, nitrogen footprint, nitrogen indicator, uncertainties

SAMMANFATTNING

I de flesta ekosystem är kväve ett tillväxtbegränsande näringsämne. Både naturliga och antropogena processer omvandlar inert dikväve (N_2) från atmosfären till reaktiva, biologiskt användbara former. Denna omvandling sker främst genom biologisk kvävefixering (BNF), industriell ammoniakproduktion (NH_3) och oxidation av N_2 till kväveoxider (NO_x) vid förbränning av fossila bränslen. Den antropogena omvandlingen av N_2 till reaktivt kväve har ökat dramatiskt under det senaste århundradet och motsvarar idag alla naturliga processer sammantaget. Den främsta nyttan är att ammoniakbaserade kvävegödsel och BNF ökar skördarna i jordbruket. Dock har antropogena processer också ökat omsättningen av reaktivt kväve i miljön med en rad oavsiktliga konsekvenser för mänskligt välbefinnande och ekosystemens funktion. NO_x och NH_3 i atmosfären bidrar till luftföroreningar och därmed till hälsoproblem. Utsläpp av lustgas (N_2O) bidrar till global uppvärmning. Kväve göder och försurar terrestra och akvatiska ekosystem, ibland med dramatiska följder för den biologiska mångfalden.

Den här uppsatsen bidrar på två sätt med kunskap om flöden av reaktivt kväve i europeiska jordbrukssystem. För det första uppskattar den och analyserar kväveflöden med särskilt fokus på animalieproduktion och på osäkerheter i jordbrukets kvävebudgetar. Resultaten kan användas för att bedöma effekter av möjliga förändringar i matsystemet. En jämförelse mellan ekologisk och konventionell mjölkproduktion visar att ekologisk mjölk sannolikt medför mindre kväveutsläpp till miljön, men också att denna slutsats kan komma att ändras med förbättrade uppskattningar av BNF. För det andra analyserar och diskuterar uppsatsen tre kvantitativa indikatorer för att bedöma kväveförluster från livsmedelsproduktion, med fokus på systemgränser, osäkerheter och miljömässig relevans. Indikatorerna har begränsad miljömässig relevans eftersom de summerar kväveflöden över tid, rum och olika kemiska former. Analysen bidrar med kunskap om hur kväveindikatorer kan användas för forskning och kommunikation med allmänheten och beslutsfattare.

APPENDED PUBLICATIONS

This thesis consists of an extended summary and the following appended papers:

Paper A	R. Einarsson and U. M. Persson (2017). Analyzing key constraints to biogas production from crop residues and manure in the EU—A spatially explicit model. *PLOS ONE* **12** (1), e0171001. DOI: `10.1371/journal.pone.0171001`.
Paper B	R. Einarsson, C. Cederberg and J. Kallus (2017). Nitrogen flows on organic and conventional dairy farms: a comparison of three indicators. *Nutrient Cycling in Agroecosystems*, pp. 1–14. DOI: `10.1007/s10705-017-9861-y`.
Paper C	R. Einarsson (2017). *The nitrogen footprint — environmentally relevant?* Working Paper.

Author contributions

Paper A: RE conceived the idea. RE collected data, designed the model, and made the calculations. RE and UMP interpreted the results and wrote the paper.

Paper B: CC and RE conceived the idea. RE selected and cleaned data and made the calculations with input from CC. RE and JK designed the statistical analysis. RE wrote the paper with input from CC and JK.

Paper C: RE conceived the idea and wrote the paper.

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CONTENTS

Abstract **i**

Sammanfattning **ii**

Appended publications **iii**

Acknowledgements **v**

1 Introduction **1**

2 Background **5**
2.1 The nitrogen cycle 5
2.2 Nitrogen fixation rates and stocks 11
2.3 Unintended consequences 13
2.4 Closing the nitrogen budget 17

3 Present work **21**
3.1 Paper A 22
3.2 Paper B 28
3.3 Paper C 35

4 Discussion and outlook **39**

References **43**

Paper A **57**

Paper B **81**

Paper C **103**

CHAPTER 1

Introduction

Nitrogen is essential for life as we know it. It is a building block of several important biomolecules, for example proteins, which have a vast range of functions in organisms including catalysis, signaling, and transport of substances; nucleic acids, which encode the genetic information of all known life forms; and adenosine triphosphate (ATP), which transports energy inside cells. Reproduction and metabolism implies a constant turnover of nitrogen in various forms.

In most ecosystems, nitrogen is in short supply [1–3]. Among animals, limited protein intake is often a constraint to growth and reproduction: herbivores compete for limited supplies of nitrogen-rich plant tissues such as fresh grass, seeds, pollen, and young leaves; and carnivores compete for a limited amount of other animals to prey on [4]. In soils, plants and microorganisms compete for limited nitrogen resources [5]. Nitrogen is essential and it is scarce.

Ironically, though, when nitrogen had just been discovered in the 1770s, the French chemist Lavoisier called it "azote", meaning "without life" [6]. Lavoisier was referring to dinitrogen (N_2) which makes up 78% of the volume of the air and is lifeless in the sense that it is relatively inert, resistant to chemical conversion into biologically useful forms, due to the strong triple bond between its two atoms, $N{\equiv}N$. It is therefore common to distinguish N_2 from all other forms of nitrogen, which are collectively called reactive nitrogen.

But the abundant atmospheric N_2 is not completely unavailable: the triple bond can be broken by a class of microorganisms called diazotrophs ("dinitrogen eaters"; remember Lavoisier's name "azote"). The conversion of N_2 into reactive nitrogen is also known as nitrogen fixation, and therefore diazotrophs may also be called biological nitrogen fixers. Their special ability is a great advantage, but it is not a free lunch: nitrogen fixation is costly in terms of energy and this partly explains why nitrogen fixers have not outcompeted every other form of life [7, 8]. Nevertheless, biological nitrogen fixation (BNF) occurs in a wide range of ecosystems and is by far the most important source of new reactive nitrogen in natural ecosystems [9].

In agriculture it is important to avoid nitrogen scarcity. To promote growth of certain plants, humans have altered nitrogen flows for thousands of years, both by applying nitrogen-rich animal manures to soil and by cultivating legumes, such as beans and clover. Legumes contribute reactive nitrogen through a symbiotic relationship with a class of nitrogen-fixing bacteria called rhizobia. The legume plants provide the rhizobia with energy-rich carbohydrates in exchange for the service of nitrogen fixation. Legumes have been cultivated since long before BNF was scientifically described, probably for more than 6,500 years [10].

Until less than a century ago, BNF in agriculture was the largest anthropogenic source of new reactive nitrogen. Today the largest source is the Haber-Bosch process, invented by Fritz Haber in 1909 and engineered to industrial scale by Carl Bosch [10]. The Haber-Bosch process forces N_2 to react with hydrogen gas (H_2) into ammonia (NH_3) which is mainly used to produce fertilizer. The last large-scale mechanism for nitrogen fixation is the oxidation of N_2 into nitrogen oxides (NO_x) that mainly happens as an undesired side effect when fossil fuels are combusted in air. Nitrogen oxides are thus emitted into the atmosphere from power plants, cars, and other combustion processes.

Taken together, these anthropogenic activities now fix more nitrogen than all natural terrestrial processes combined. This has far-reaching implications for humanity and for planet Earth. The primary intended effect is that agriculture has been able to support an impressive increase in food production as the world population has quadrupled, from less than two billion people when Fritz Haber invented industrial nitrogen fixation, to more than 7.5 billion today. But there are also many unintended consequences as much of the anthropogenically fixed nitrogen is eventually lost to the environment, where it damages human well-being and disturbs the nitrogen scarcity that is common in natural ecosystems.

Motivation, aim and contributions

Humanity is facing a dilemma as the agriculture and food systems that feed the world also emit reactive nitrogen to the environment with consequences that are widely seen as severe problems. Anthropogenic nitrogen in the environment has a range of adverse effects on human health and well-being and furthermore changes ecosystem functioning and reduces biodiversity, possibly with serious long-term effects for human well-being [11–14]. The EU and its member states have many policies in place to control these problems, but there is a long way to go if the EU policy targets are to be met [13].

Much research has been invested into understanding how nitrogen flows in the European food system, what changes would be possible to make, and what combinations of changes would be most efficient to reduce the environmental problems. An overarching aim of my research is to build on these efforts and contribute to an assessment of the biophysical limits to what can be done. This thesis works towards that aim by investigating the relationship between the intended and the unintended nitrogen flows in agriculture, i.e., between the nitrogen in food products and the nitrogen lost to the environment.

The thesis makes two types of contributions. First, it estimates and analyzes nitrogen flows in European agricultural systems, with results that can be used to assess the effects of potential changes to the food system. Second, it analyzes and discusses quantitative indicators to describe nitrogen losses associated with food products or production systems, and thereby improves our understanding of how these indicators can be used for research purposes and for communication with the public and policy-makers.

Disposition of this thesis

The thesis consists of four chapters introducing and summarizing my research, followed by three appended papers. Chapter 2 gives further background on reactive nitrogen: how is nitrogen transformed and transported in the environment, and what are the unintended consequences for humans and ecosystems? Chapter 3 summarizes and discusses the three appended research papers. Chapter 4 ends with general reflections on my research so far and an outlook into challenges that I might take on next.

CHAPTER 2

Background

How is nitrogen transformed and transported in the environment and in agriculture? What are the major stocks of reactive nitrogen, and what is the global rate of fixation? What are the unintended consequences as anthropogenic nitrogen is lost to the environment? These are some key questions I will address here.

This chapter is necessarily very selective in its scope and depth, as all these questions lead into complex research fields. If after reading this chapter the reader feels inspired to find out more, then my work is successful.

Section 2.1 gives an overview of how nitrogen is used and transformed in agriculture and the environment. Section 2.2 presents estimates of the global rate of nitrogen fixation and of the major stocks of reactive nitrogen. Section 2.3 describes some important unintended consequences of nitrogen losses to the environment and how they affect the environment and human health. Section 2.4 introduces a few key reasons as to why it is hard to close the nitrogen budget, i.e., to understand where nitrogen inputs to the environment end up.

2.1 The nitrogen cycle

To understand *how* nitrogen is transformed in the environment, I think it is useful to first ask *why*. In this overview of the nitrogen cycle I will refer to two driving forces. The first one is familiar from the previous chapter, namely that nitrogen occurs in essential biomolecules: nucleic acids, proteins, ATP, and many more. All life forms use nitrogen to build and maintain their tissues. Fixing N_2 from the atmosphere or harvesting nitrogen from other sources is a necessary part of life.

The other driving force has to do with energy. Life cannot exist without a source of energy. Humans must eat to replace the energy that is lost working, breathing, reproducing, destroying old tissues, and building new ones; and all other life forms must eat too, in one way or another. Photosynthetic life forms are a special case as they "eat" sunlight. Most other life forms hunt and eat each other, at least figuratively speaking. In both cases it is useful to think about eating in terms of the oxidation and reduction of organic compounds, for example sugar ($C_6H_{12}O_6$). Many life forms make use of the chemical energy stored in organic compounds by oxidizing, or "burning", the carbon:

$$C_6H_{12}O_6 + 6\,O_2 \longrightarrow 6\,CO_2 + 6\,H_2O.$$

Remember that oxidation is to lose electrons and reduction is to gain electrons. Oxidation and reduction must always balance out, so we can conclude that oxygen is reduced when it oxidizes sugar. Photosynthetic plants work the opposite way: they use solar energy to produce sugar by reducing the carbon in CO_2. Again, since oxidation and reduction must balance out, we conclude that oxygen is oxidized—sounds stupid, but it is correct—when it goes from H_2O to O_2. This reminder of redox chemistry is all we will need later to understand why some life forms can use certain nitrogen compounds to harvest energy from energy-rich organic compounds.

Nature is full of bacteria and other organisms that actively promote chemical reactions. These reactions can often be understood in terms of the above-mentioned driving forces: obtaining building materials for biomolecules, or energy to live on. Bacteria promote reactions by producing enzymes tailored to catalyze the given reaction. Enzymes are really quite remarkable: remember that nitrogen-fixing bacteria do their work at ambient pressure and temperature (around 1 bar and 290 K), while the Haber-Bosch process operates at 200 bar and over 700 K. In the following I will mostly skim over these details and write that microorganisms "perform" or "mediate" a reaction.

Another note on terminology is that I will often refer to bacteria,

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[1] Another reason to collectively refer to microbes is the constant discovery of new species. For example, it was discovered only about twelve years ago that archaea, in addition to bacteria, contribute to nitrification [15, 16]; and the notion that eukaryotes contribute to reduction of nitrate has also started accumulating evidence only in the last couple of decades [17]. Thus, remembering the less specific name "microbe" is less likely to be outdated knowledge in a few years.

archaea, fungi, amoebas, etc., as microorganisms or microbes. To understand the nitrogen cycle in full detail it is important to know the difference, but for this overview it will suffice to think of them as a very diverse group of tiny organisms.[1]

The nitrogen cycle and its effects on the environment are documented in many textbooks and review articles. This section draws on Refs [17–20], but I have chosen to not interrupt the text with specific references except in a few spots which are less well known.

The many forms of nitrogen

Nitrogen occurs in many chemical forms with widely varying properties, from inert N_2 to proteins to the explosive nitroglycerin ($C_3H_5N_3O_9$). In this thesis I avoid going into details and instead mostly discuss four classes of nitrogen compounds: (1) oxidized inorganic forms (positive oxidation state); (2) N_2, which has neutral oxidation state; (3) reduced inorganic forms (negative oxidation state); and (4) organic nitrogen forms, which are usually reduced. All the forms that are discussed in this thesis are listed in Table 2.1. The following sections will introduce the main biological transformations between these chemical forms, as illustrated in Figure 2.1.

Table 2.1: Examples of nitrogen compounds, ordered by the oxidation state of the nitrogen.

Classification	Name or formula	Oxidation state
Oxidized inorganic	NO_3^- (nitrate)	+5
	NO_2 (nitrogen dioxide)	+4
	NO_2^- (nitrite)	+3
	NO (nitrogen monoxide)	+2
	N_2O (nitrous oxide)	+1
Neutral oxidation	N_2 (dinitrogen)	0
Reduced inorganic	NH_3 (ammonia)	-3
	NH_4^+ (ammonium)	-3
Organic	proteins, amino acids, nucleic acids, etc	-3

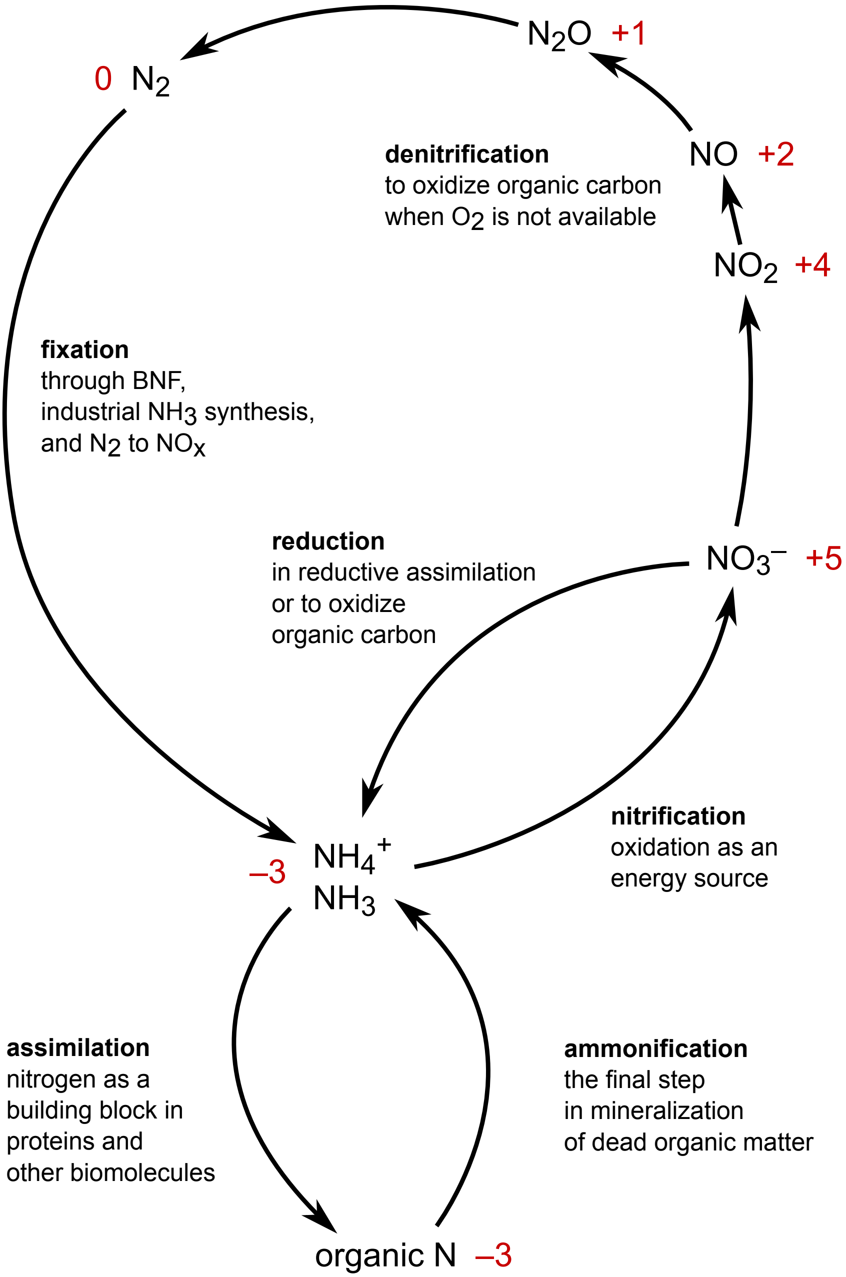

Figure 2.1: Key transformations between chemical forms in the nitrogen cycle, corresponding to the main parts of this text. The numbers in red are oxidation states of the nitrogen.

Nitrogen fixation

Nitrogen fixation is any process where N_2 is converted to reactive nitrogen. In nature, this happens in several ways. First and most important is BNF, which is performed by a range of diazotrophs (Chapter 1). Important examples of diazotrophs are free-living cyanobacteria (sometimes called blue-green algae) which are found both in soils and water, as well as the symbiotic rhizobia that live inside legume roots. BNF is the reduction of N_2 into ammonia (NH_3) or ammonium (NH_4^+). The other and much smaller natural nitrogen-fixing process is lightning which oxidizes N_2 into nitrogen oxides (NO and NO_2, collectively called NO_x).

Reduction and assimilation

Plants and microbes can take up and assimilate inorganic nitrogen, i.e., transform it into amino acids and other useful organic forms. The most important inorganic forms for plants and microbes are ammonium and nitrate (NO_3^-). Nitrate must first be reduced to ammonia before it can be assimilated, and this requires energy input. Hence, plants and microbes typically prefer ammonium since it is energetically cheaper. The combined reduction and assimilation of nitrate is sometimes called assimilatory reduction, but in Figure 2.1 the two steps are illustrated separately.

Nitrification

Oxidation of ammonia to nitrate releases energy. Some microbes live on energy that they harvest from this oxidation. The oxidation is microbially mediated in multiple steps. There are microbes specialized in each of the steps, but the whole process is collectively called nitrification. Nitrification partly explains why inorganic nitrogen in soil is more often oxidized than reduced. Another important explanation is the preferential uptake of reduced nitrogen by plants and microbes. Reduced inorganic nitrogen is really popular among plants and microbes, and is usually assimilated or oxidized as soon as it is available.

The fully oxidized end result of nitrification is nitrate, but as an intermediate step some microbes produce nitrous oxide (N_2O). Thus, some N_2O gas can seep out from the soil as a by-product of nitrification. This flow is omitted from Figure 2.1 since it is comparatively small. However, N_2O is a potent greenhouse gas, which I will get back to in Section 2.3.

Mineralization: depolymerization and ammonification

As soon as someone or something dies, or when a plant drops a leaf, decomposition starts. Decomposition means that a whole array of different microbes, fungi and animals start eating dead tissues, extracting energy and nutrients from them, and in the process breaking them into smaller and smaller parts. As an example, consider decomposition of a protein molecule. A protein is a polymer of amino acids. The first decomposition step is depolymerization, which means to break the polymer into individual amino acids. Depolymerization, like most other reactions mentioned here, is catalyzed by microbes producing specialized enzymes. Then one of two things happen: either the amino acid is directly taken up and used by a plant or a microbe, or it is further broken down by other microbes that produce other specialized enzymes. The final decomposition step for an organic nitrogen compound is called ammonification, which means that the organic nitrogen is transformed into ammonium or ammonia.

The combined process of depolymerization and ammonification is called mineralization. Figure 2.1 shows only the ammonification step since the illustration only distinguishes nitrogen forms by oxidation state and by organic/inorganic. In that sense the illustration is deceptively simple, because it does not show that some organisms assimilate organic nitrogen directly, or in other words, that many interesting things happen within the organic pool. Examples of processes within the organic pool include uptake of organic nitrogen compounds in plants and microbes, but importantly also all the protein that humans eat: in contrast to plants and microbes humans can only build new proteins from amino acids, not from ammonium.

Denitrification and other reduction processes

Denitrification is the stepwise reduction of NO_3^- to NO_2^-, NO, N_2O, and finally N_2. It mainly occurs in oxygen-poor or oxygen-free conditions where denitrifying microbes use nitrogen compounds to “burn” organic carbon, or more precisely, they oxidize organic carbon by reducing inorganic nitrogen. Carbon oxidation releases more energy when it is done with O_2, but in environments where O_2 is not available there is a niche for microbes that can use the less efficient nitrogen-based oxidation. Both nitrification and denitrification are performed by microbes to gain energy. In the case of nitrification, the energy is harvested directly from the oxidation of ammonia. In contrast, denitrification in itself uses energy and is only done to harvest even more energy from oxidation of carbon compounds.

Denitrification often leads to gaseous nitrogen losses to the atmosphere since the final product is usually one of the gases N_2 or N_2O. In some ecosystems, especially wetlands where there is little oxygen, denitrification can lead to substantial removal of reactive nitrogen.

I finally mention two other processes that reduce inorganic nitrogen. The first is anaerobic ammonia oxidation, or anammox for short. The reaction occurs only in oxygen-free environments and produces dinitrogen from ammonium and nitrite: $NO_2^- + NH_4^+ \longrightarrow N_2 + 2\,H_2O$. Anammox bacteria perform this process to gain energy that they use to reduce CO_2 to useful organic carbon. Anammox was discovered in 1995 and has so far not been identified as an important process in most soils.

The last process in this overview of the nitrogen cycle is called dissimilatory nitrogen reduction to ammonium (DNRA). As the name implies, DNRA reduces NO_3^- all the way to NH_4^+. This seems to happen primarily when there are plenty of carbon sources and very few electron acceptors, which makes sense since the reduction to ammonia is energy-intensive: it can only be motivated if energy-rich carbon compounds are available and electron acceptors are lacking. DNRA is a significant process at least in wetlands and marine environments, but its importance for nitrogen cycling in soils is yet unclear [17, 19].

2.2 Nitrogen fixation rates and stocks

The global rate of anthropogenic fixation is about 200 $\mathrm{Tg\,N\,y^{-1}}$ (million metric tonnes of nitrogen per year[2]). By comparison, BNF in natural ecosystems is estimated around 128 $\mathrm{Tg\,N\,y^{-1}}$ in terrestrial ecosystems

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[2] The unit $\mathrm{Tg\,N\,y^{-1}}$ is read as teragrams of nitrogen per year. $1\,\mathrm{Tg} = 10^{12}\,\mathrm{g} = 10^{6}\,\mathrm{kg}$, or in plain English one million metric tonnes. Teragrams is a good word because it avoids any confusion with the UK ton of 2,240 pounds (also known as a “long ton”) or the US ton of 2,000 pounds (“short ton”).

and 120 Tg N y^{-1} in marine systems, and lightning around 4 Tg N y^{-1}. Although estimates are quite uncertain (see Table 2.2), it seems likely that the anthropogenic fixation roughly equals all natural fixation globally, or twice the natural terrestrial fixation.

Table 2.2: Global estimates of nitrogen fixation rates. Based on a compilation by Fowler et al. [21], except for NO_x emissions from Jaeglé et al. [22]. The rates in the middle column, except for NO_x, are best estimates according to Fowler et al. [23].

Source	Rate Tg N y^{-1}	Uncertainty range Tg N y^{-1}
Ammonia synthesis	120	110–130
Agricultural BNF	60	40–80
NO_x from fuel combustion	26	
Natural BNF, terrestrial	128	44–290
Natural BNF, marine	120	100–200
NO_x from lightning	4	

The global rate of fixation is small compared to the total stock of reactive nitrogen on Earth. Terrestrial ecosystems contain somewhere around 100,000–300,000 Tg N, or about 1,000 years' worth of natural terrestrial fixation [18, 24]. About 90 % of the terrestrial N is found in dead organic matter in soils and the remainder is found in living biomass and inorganic nitrogen in soils [18, 19]. Perhaps 6,000–10,000 Tg N is found in plants and 2,000–6,000 Tg N in soil microbes, but especially the microbial biomass is hard to estimate [18, 19, 25]. Thus, despite the central role of microbes in the nitrogen cycle, they account for only one or a few percent of the soil nitrogen pool.

The atmosphere contains, in addition to 10^9 Tg N_2, a little more than 1,000 Tg N as N_2O and maybe 5 Tg N in other gases and particles [18]. Aquatic ecosystems are a much larger store of reactive nitrogen than terrestrial ecosystems, although the amounts are poorly known. The oceans may contain about 1,000,000 Tg N, of which at least half is dissolved NO_3^- [18, 26]. In microbes below 8 m depth under terrestrial systems and in sediments under the oceans there might be 4,000–100,000 Tg N [25, 27]. Finally, bound in rocks, deep sediments, and coal deposits, there are many orders of magnitude more reactive nitrogen, but this nitrogen is for all practical purposes unavailable to life on the Earth surface [28].

2.3 Unintended consequences

What happens to the planet as humans add increasing amounts of reactive nitrogen to soils and to the atmosphere? Where does the nitrogen go? What are the unintended consequences?

The nitrogen cascade

As nitrogen is easily transported and transformed by biological and chemical processes, a single nitrogen atom may have multiple consequences over time and in different places. Imagine for example how a newly fixed ammonium ion may be added to soil, nitrified to nitrate, washed out with rain to a water body, taken up by a microbe, assimilated to a protein, later mineralized, taken up by a tree, stored for weeks or years, and so on, until it is eventually denitrified to N_2 and returned to the atmosphere. The constant flow through different places and chemical forms has famously been called the nitrogen cascade [29, 30]. The nitrogen cascade is a compact expression for a core idea in research on nitrogen in the environment: although nitrogen fixation is an important driver of environmental change, what matters in the end is not the source of the nitrogen but rather the transformations and the concentrations in different parts of the environment.

Figure 2.2 illustrates the processes that will be covered in the following pages.

Nitrogen in the atmosphere

Several sources emit oxidized nitrogen to the atmosphere. Section 2.1 mentioned microbial processes that produce NO_x and N_2O which may both escape into the atmosphere. In addition, burning of biomass, both natural and anthropogenic, releases substantial amounts of NO_x as organic nitrogen in the biomass is oxidized. Together with the NO_x from fuel combustion and lightning (Table 2.2), the NO_x emissions amount to about $40\,\mathrm{Tg\,N\,y^{-1}}$ [22]. Denitrification contributes some $15\text{–}20\,\mathrm{Tg\,N\,y^{-1}}$ as N_2O [12, 23].

Input to the atmosphere of reduced nitrogen comes mainly as NH_3 and mainly from agriculture. About half of the emissions come from animal manure during storage and handling, and the rest from synthetic fertilizer spreading, soil processes, biomass burning, and

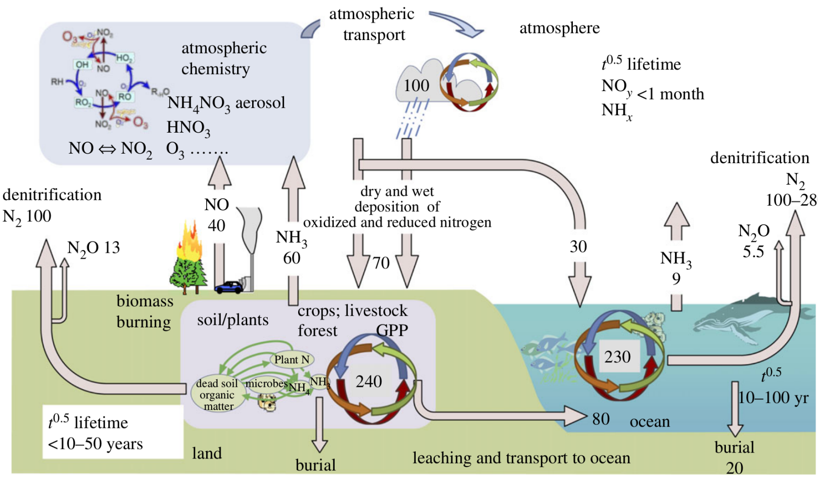

Figure 2.2: Transformation and transport of nitrogen through terrestrial and marine environments. Fluxes are given in Tg N y^{-1}. Figure by Fowler et al. [23], reused with permission.

processes in the oceans [31]. In total, these processes emit about 60 Tg N y^{-1} from land and 9 Tg N y^{-1} from the oceans [23].

NO_x and NH_3 in the atmosphere

Both NO_x and NH_3 are major contributors to formation of smog, i.e., a toxic mixture of ozone, hydrocarbons, small particles, and other substances. Through various chemical reactions, NO_x contributes to formation of ozone and to formation of small organic particles. In addition, both NO_x and NH_3 contribute to formation of small inorganic particles (salts) such as ammonium sulfate and ammonium nitrate. Smog, ozone, and particles are known contributors to asthma and other respiratory diseases as well as cancer [14].

Atmospheric concentrations of NO_x and NH_3 do not increase forever as both gases are eventually deposited back to the Earth's surface in gas or particle form (dry deposition) or dissolved in precipitation (wet deposition). But the gases, especially NO_x, and their reaction products can sometimes travel long distances, even hundreds of kilometers, before deposition. Thus, NO_x or NH_3 emissions in one country may be deposited in the oceans, or in other countries.

N_2O in the atmosphere

N_2O is much less reactive than NO_x and therefore remains longer in the atmosphere. Some rises all the way to the stratosphere where it contributes to depletion of stratospheric ozone and thereby to skin cancer in humans [14]. Since a number of heavily ozone-depleting substances were abolished with the Montreal protocol in 1987, N_2O has become the single most important ozone-depleting chemical [14].

N_2O is also a potent greenhouse gas and thus emissions contribute to climate change. Since preindustrial times the atmospheric N_2O concentration has increased about 20 % [14], and the anthropogenic N_2O that has currently accumulated in the atmosphere has been estimated to contribute about 6 % of the anthropogenic radiative forcing [32]. However, increased radiative forcing from N_2O is only one in a long list of nitrogen-related effects on the climate, some warming and some cooling, e.g., as nitrogen fertilization can lead to increased biomass growth. The combined effect on the climate of anthropogenic nitrogen emissions is highly uncertain, but likely to be net cooling [14, 33]. That said, decreasing the emissions of N_2O specifically would clearly reduce global warming [33].

Nutrient enrichment and acidification

Terrestrial ecosystems

Atmospheric deposition provides large amounts of nitrogen to some terrestrial ecosystems. As an example, it is estimated that in preindustrial times forests received deposition of about 2 kg N ha^{-1} y^{-1}, but now many areas in Europe receive 10–20 kg N ha^{-1} y^{-1}, sometimes up to 60 kg N ha^{-1} y^{-1} [19, 34]. By comparison, in a growing European forest it would be typical to find an uptake of 60–100 kg N ha^{-1} y^{-1} in the vegetation, but almost as much is returned in turnover of leaves and fine roots, leaving a net uptake of 5–10 kg N ha^{-1} y^{-1} [19]. Thus, nitrogen deposition in European forests may easily be larger, sometimes several times larger, than the whole net uptake in vegetation.

In addition to its fertilizing effect, nitrogen contributes to acidification of terrestrial environments. Through a combination of these effects, large nitrogen inputs result in a complex set of changes, often including changes in the composition of species in plant, animal, and microbe communities. Increased nitrogen supply often leads to decreased biodiversity [14, 19]. In many sensitive ecosystems, nitrogen deposition above 5–10 kg N ha^{-1} y^{-1} has been established as a so-called critical load that cannot be exceeded without risk for reduced plant biodiversity [14].

Aquatic ecosystems

Increased mineral nitrogen concentrations in soils tends to increase water-borne nitrogen losses through a process called leaching. Since soil particles are often negatively charged, they repel the negative NO_3^- ion which is therefore easily transported downwards with water flows. While other nitrogen forms, including organic nitrogen compounds, may also be leached, the process is particularly fast if nitrification leads to high concentrations of NO_3^-. Leaching is quite common on agricultural land, but also occurs in some other ecosystems. Depending on hydrological conditions, leached nitrogen may be stored in groundwater for long periods, or quickly reach a stream or lake, and eventually it flows to the ocean.

While leaching is the largest contributor of nitrogen to aquatic ecosystems [23, 35], there are also substantial flows entering aquatic environments through atmospheric deposition and in wastewater. Atmospheric deposition is special both because gaseous reactive nitrogen can travel long distances in the atmosphere and because it contributes especially to acidification, particularly in freshwater ecosystems [14]. Wastewater is special as it is a point source and can therefore have strong effects locally although it is a smaller part of total nitrogen input. In summary, while deposition and wastewater contribute less nitrogen to aquatic ecosystems than leaching, they are both significant and they are also qualitatively different.

Excessive nutrient loading in aquatic ecosystems is also known as eutrophication, and just like in terrestrial ecosystems it leads to a complex set of ecological changes. Severely eutrophicated waters may experience extreme growth of algae or cyanobacteria, also known as algal blooms, which depletes oxygen and may suffocate other organisms. Decomposition of dead microbes and animals then leads to further oxygen depletion in bottom waters, in extreme cases leading to so-called dead zones. This process is well-known from freshwater systems, but is also increasingly occurring in coastal marine waters [14, 36]. Eutrophication of freshwater is common. For example, a

European assessment of nitrogen eutrophication in surface waters found that more than half of Europe fell into the highest of three risk classes for aquatic eutrophication, and essentially only the boreal areas in northern Europe fell in the lowest risk class [36].

Nitrogen and biodiversity loss

In summary, nitrogen fertilizes and acidifies ecosystems, sometimes with dramatic effects on biodiversity. There is wide scientific agreement that nitrogen is an important driver of biodiversity loss and that biodiversity loss is a main threat to the so-called ecosystem services that nature provides and humans rely on for food production, drinking water provision, climate control, recreation, and many other things. The exact impacts are hard to judge today, but it may well turn out in the long run that biodiversity loss is the most serious and long-lasting effect of anthropogenic nitrogen fixation.

2.4 Closing the nitrogen budget

When researchers attempt to measure or estimate all the flows of reactive nitrogen entering and leaving a geographical area, it is more common than not that the measured inflows are substantially larger than the measured outflows. This may be explained by one or several of following factors: (1) some measurement method is systematically incorrect, (2) there is some substantial outflow that has not been measured, (3) denitrification inside the area accounts for the difference, or (4) nitrogen is being accumulated inside the area. All of these factors are plausible explanations and all of them are difficult to check. Therefore it is a difficult task to close a nitrogen budget, i.e., to make an accurate breakdown where all the flows and accumulations add up.

This section is not an attempt to solve any of these puzzles, but to introduce and draw attention to a few specific uncertainties that are especially relevant for the research I present in Chapter 3.

Denitrification

Measuring and understanding denitrification is very difficult. The process occurs in soils as well as in wetlands, lakes, rivers, and the oceans. The diversity of microbial mechanisms for denitrification is actually quite a bit larger than I indicated in Section 2.1, and recent discoveries of microbes show that we should not be surprised to find more [37]. Furthermore, different measurement methods give different results, and variations in weather, soil type, availability of different substrates, and other factors are known to influence denitrification rates [37].

Both in studies of whole catchments and of agricultural systems, denitrification is often estimated as a balancing term, i.e., as the difference between the known inputs and outputs [38, 39]. However, this builds on the assumption that the reservoirs of reactive nitrogen do not change over time. This assumption might be reasonable for natural ecosystems that show little change over long timescales, but this is not often the case. Furthermore, there are at least two types of direct evidence against the constant reservoir assumption, which I will now outline.

Nitrogen accumulation in groundwater

One line of evidence for nitrogen accumulation in groundwater is aquifers that show increasing nitrate concentrations over time. This has been observed both in Europe and in the US [36, 38, 40]. However, since the measurements only specify the concentrations, and given the complexity of underground water transport, few have even tried to estimate how large the accumulations might be and how long time will pass before they exit into surface water [38].

Nitrogen accumulation in soils

Nitrogen accumulation in soils and vegetation seems to be more reliably estimated than in groundwater. A review by Galloway et al. [12] suggests that about 60 $\mathrm{Tg\,N\,y^{-1}}$ might be accumulating in terrestrial systems, and based on a similar review Fowler et al. [23] suggest a range of 25–100 $\mathrm{Tg\,N\,y^{-1}}$.

While such an accumulation rate would be substantial compared to other flows, perhaps equal to 40 % of the natural terrestrial fixation, remember that it is still a small annual accumulation compared to the total stock of soil organic nitrogen (Section 2.3). This stock has evidently accumulated in some way, and therefore it does not seem too strange if some soils are currently accumulating organic nitrogen.

Agricultural soils are especially relevant since agriculture is the main focus of this thesis. It is poorly known both what sign and what magnitude the rate of nitrogen stock change may have in agricultural soils. While several model-based assessments assume zero or estimate negative stock changes in EU agricultural soils [39, 41], there are estimates from the River Thames basin, as well as from Canada and USA that suggest positive change [42–44]. In 1999, Smil [45] reviewed several sources of evidence and suggested that while agricultural soils of the world accumulate perhaps 3–6 kg N ha^{-1} y^{-1} on average, those soils that receive plenty of nitrogen fertilizer may accumulate 25–35 kg N ha^{-1} y^{-1}. One recent publication, based on long-term measurements of agricultural soil nitrogen in the Mississippi river basin (1957–2010), suggests that soils are currently (1980–2010) accumulating organic nitrogen at a rate of perhaps 30–50 kg N ha^{-1} y^{-1} [38]. This would explain about half of the difference between nitrogen inputs and outputs in the Mississippi river basin, and it is twice the amount of nitrogen that reached the ocean in the Mississippi river in the same period [38]. Although these few indications of substantial accumulation should not be extrapolated to all agricultural soils in regions of high nitrogen input, they do suggest that soil nitrogen accumulation can play a significant part in closing the nitrogen budget in agricultural landscapes.

CHAPTER 3

Present work

This thesis makes two principal types of contributions to the understanding of nitrogen flows in European agricultural systems:

1. The thesis estimates and analyzes flows of nitrogen in European agriculture. Specifically, Paper A [46] provides spatially explicit estimates of where and how much manure and crop residues are produced in the EU, as well as an analysis of how these substrates could be used for biogas production. Paper B [47] uses farm-level data to describe and analyze nitrogen flows in Swedish milk production, comparing organic and conventional production systems.
2. The thesis describes and discusses different indicators for nitrogen use efficiency and nitrogen pollution from agricultural systems. In Paper B we calculate a common indicator called nitrogen surplus as well as a more recent indicator called nitrogen footprint, for organic and conventional milk production in Sweden, and discuss the merits of these indicators in some detail. In Paper C [48], I critically discuss some aspects of the nitrogen footprint in further detail.

The following sections goes through the appended papers one by one, providing motivations, research questions, method descriptions, results, and some discussion and conclusions. I focus on highlighting results and methodological difficulties that I find particularly interesting, or that were not clearly discussed in the papers.

3.1 Paper A

Motivation

This project originally had to do with EU policy for greenhouse gas emissions from transport fuels. To make a long story short, there are reasons to believe that biofuel production on cropland can affect demand for land through international markets and thus cause indirect land-use change, potentially releasing large amounts of greenhouse gases and hence weakening or reversing the intended greenhouse gas savings of the biofuels. In response, EU politicians have adjusted the renewable energy policies to provide extra incentives for biofuels that supposedly do not compete for land, one example being biogas produced from crop residues and manure.

One hypothesis we had was that biogas production from crop residues and manure might be limited by the geographical distribution of these substrates in combination with their physical and chemical properties. Manure is often handled in liquid manure systems where feces and urine are mixed, possibly with additional water, in a tank. Liquid manure typically has dry matter concentrations of 6–10 % and is therefore expensive to transport and expensive to digest to biogas in isolation since the reactor is mostly processing water. Furthermore, some manures that are too nitrogen-rich, i.e., have too low C:N ratio, can cause ammonia poisoning of the digesting microbes, decreasing its efficiency. By comparison, crop residues are dry and rich in carbon, which makes them cheaper to transport and potentially highly productive in terms of biogas. A problem, though, is that the crop residues are actually too dry and have too high C:N ratio to be easily digested. A mixture of manure and crop residues is therefore to digest than either one in isolation. But European agriculture is rather segregated: the intensive crop production preferentially happens on the most fertile soils, while livestock production is most concentrated along the coasts and in regions with less fertile soils.

Research questions and method

Our research questions were the following: How much manure and crop residues are produced annually in the EU? How are the substrates geographically distributed? Can they be reasonably transported to biogas plants of sufficient size to be economically viable? Would the chemical composition of the substrate mixtures limit the biogas production?

To answer these questions, we estimated the amounts and geographical distribution of available substrates using a combination of several datasets:

- subnational agriculture statistics from Eurostat on livestock populations and crop production,
- national data on manure management practices in EU countries from the national reporting to the UN Framework Convention on Climate Change,
- a land cover map showing where arable land is located [49], and
- a livestock density map, showing a statistics-based estimation of how pigs, cattle, and poultry are geographically distributed [50].

We then designed a mathematical model of hypothetical biogas plants all over the EU to assess what fraction of the substrates could be utilized.

Results

In Paper A we focused on the biogas production potential. In this thesis I instead focus on the substrate quantities and the nitrogen associated with them.

Figure 3.1 shows the spatial distribution of the available substrates. The segregation of crop production and livestock production is seen for example in the UK, where the eastern half has more crop production and the western half has larger livestock populations. As a result, the combined substrate resources are in some regions very dry and carbon-rich and in other regions rather wet and nitrogen-rich (see Figure 3.2).

One significant outcome of this work from a nitrogen perspective is that it says something about the amount of nitrogen that could plausibly be put through biogas digestion. Table 3.1 shows estimates of nitrogen in the available substrates and the substrates that could be utilized for biogas production in the base scenario. These numbers are not included in Paper A but follow directly from the original calculations.

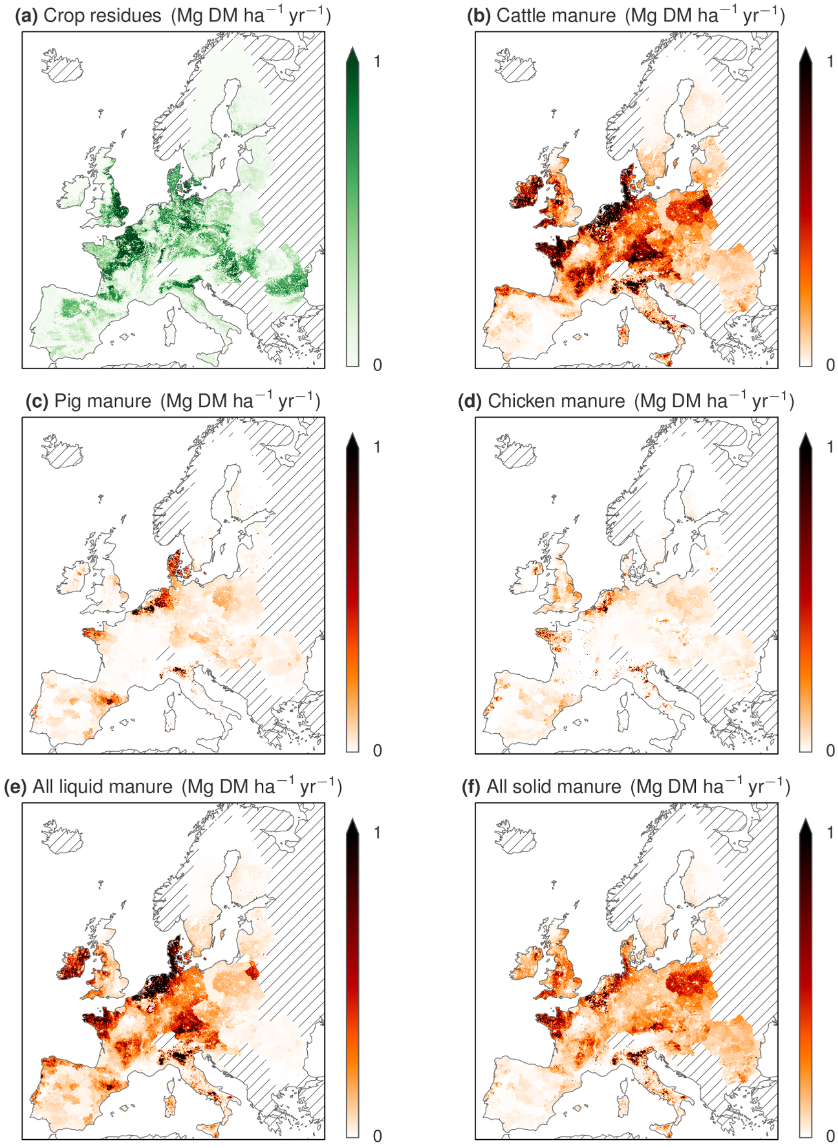

Figure 3.1: Estimated amounts of crop residues and manure available for biogas production in the EU. Figure by Einarsson and Persson in Paper A, under Creative Commons license.

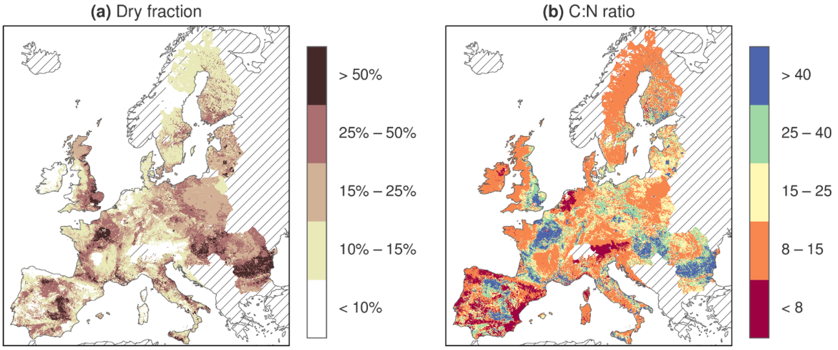

Figure 3.2: Dry matter content (panel a) and carbon:nitrogen ratio (panel b) of the combined substrates shown in Figure 3.1. Figure by Einarsson and Persson in Paper A, under Creative Commons license.

Our estimate of the nitrogen quantity in manure available for biogas production is about 50 % larger than an estimate of nitrogen in applied manure by Leip et al. in the European Nitrogen Assessment [34, Figure 16.1]. One explanation for the different sizes of these estimates is that our estimate included all manure nitrogen excreted in animal houses, whereas the estimate by Leip et al. accounted for ammonia losses up to and including manure application to fields. Another partial explanation is that our estimate of nitrogen in solid manure includes the nitrogen in straw that is used for animal bedding, and it is not clear whether this is comparable to their method. In any case, it is not surprising to find a discrepancy of this magnitude considering the difficulties in assessing manure amounts and manure management systems (for details, see Paper A, pp. 6–7). Another difference which is harder to reconcile is that our estimate of nitrogen in available crop residues is about 0.4 $\mathrm{Tg\,N\,y^{-1}}$, whereas Leip et al. [34] estimate a flow of 3.9 $\mathrm{Tg\,N\,y^{-1}}$ in crop residues. I can see three possible explanations for this discrepancy. First, our estimate is the nitrogen in crop residues available for biogas production; our estimated total production of residues from cereals, sunflowers, and sugar beets is about 1.3 $\mathrm{Tg\,N\,y^{-1}}$. Second, it is likely that Leip et al. included a larger set of crops, but this it is not clear from their method description. Third, it is likely that we have assumed different crop-to-residue ratios due to different definitions or data sources, but this is also not clear. In summary, the estimates of manure nitrogen agree reasonably well, but the estimates of nitrogen in crop residues are harder to reconcile.

Table 3.1: Available and utilized substrates in Paper A, expressed in terms of nitrogen content. Available substrates refers to the amount of manure and crop residues that could be collected from animal houses and cropland. Utilized substrates are the substrates that were used for biogas production in the base scenario.

	Available	Utilized	
	$\mathrm{Tg\,N\,y^{-1}}$	$\mathrm{Tg\,N\,y^{-1}}$	%
Cattle manure	3.8	3.0	80
Pig manure	1.2	0.8	66
Chicken manure	0.7	0.2	31
Crop residues	0.4	0.1	32
Total	6.1	4.2	68

Discussion

Biogas digestion has several effects on nutrient cycling since the digestate, i.e., the slurry that remains after the biogas has been removed, is usually used as fertilizer. I will mention two types of effects.

First, the digestate is chemically quite different from the incoming substrates. Digestion typically leads to nitrogen mineralization: the digestate might have more than 75 % of its nitrogen in ammonium form [51]. Another important difference is that the digestate contains much less carbon, since a major portion has left as biogas (CO_2 and CH_4). As a result of the high ammonium concentration, digestate nitrogen is more readily available for uptake in plants. A review by Möller and Müller [52] concluded that that nitrogen use efficiency is higher when digestate is applied than a corresponding amount of nitrogen in crop residues or green manure, but that the same result does not necessarily hold for manure. The lower organic carbon content compared to manure or crop residues means that the digestate is less useful as an energy source for soil biota, and research suggests that microbes and earthworms are comparatively at a disadvantage when soil is fertilized with digestate rather than a corresponding amount of manure slurry [53–55]. The goal here is not to review the literature on digestate and soils, but to point to this as an important concern.

Second, it is possible to mechanically or chemically process the digestate to obtain other fertilizer products. For example, a centrifuge separates the digestate into a relatively nitrogen-rich liquid fraction and a relatively phosphorus-rich fiber fraction. The fiber fraction may contain only about 5–10 % of the dry matter but 70 % of the phosphorus [51]. In regions with high livestock density, this may be an attractive option for farmers since standards for maximum phosphorus application are often the limiting factor for manure application. An example is the Maabjerg biogas plant in livestock-rich western Jutland region of Denmark which outputs about 10 MW higher heating value of biogas—the largest size we considered in Paper A—and exports the fiber fraction to eastern Denmark where there is less livestock [56]. The liquid fraction can then be further processed to denitrify the nitrogen (e.g., using anammox bacteria) or convert it into a concentrated mineral fertilizer [51].

Conclusions

My view is that Paper A makes two contributions to the aims of this thesis. The first one is the most obvious. Since manure in particular is a major transporter of nitrogen and an important contributor to nitrogen losses, it is useful to track these flows if it is a goal to reduce nitrogen losses to the environment. However, estimates of nitrogen flows in crop residues and manure are not readily available, as national statistics primarily measure useful products, not by-products that circulate within agricultural systems.

The second contribution is to contribute an order-of-magnitude estimate of how biogas production potentially can affect nutrient cycling in European agroecosystems. This contribution was certainly not spelled out in Paper A, but was in fact one important reason that I was interested in biogas to begin with. Paper A and the further elaboration in this thesis show that biogas production from crop residues and manure is not to be seen only as an energy technology, but also as a change to the nutrient cycling in an agroecosystem. Paper A suggests that current technology allows biogas production with an energy content of 0.7 $\mathrm{EJ\,y^{-1}}$ and a nitrogen turnover of 6 $\mathrm{Tg\,N\,y^{-1}}$ in the EU, a change that would only make a marginal contribution to EU’s energy balance, but could have a large effect on its cycling of nitrogen, phosphorus, and carbon.

3.2 Paper B

Motivation

Livestock production is a central part of European agriculture. It has been estimated that in 2002, agricultural land in EU27 produced a total of 18 Tg N in crop products and feed while EU27 livestock consumed 12 Tg N in feed [34]. Hence, although I'm skimming over some details about trade balances, it is clear that livestock production uses a majority of the agricultural land. Livestock production is associated with substantial losses of nitrogen during manure storage and handling, in cultivation of feed crops, and in food waste among consumers [34].

Milk production is an important part of European livestock production. In 2013 there were over 23 million dairy cows in EU28, making up about 18 % of the livestock units (LSU) in EU28 [57]. EU dairies annually collect about 150 Tg of milk which contains about 0.8 Tg N (assuming a crude protein content of 3.4 %). Hence, in order to understand nitrogen losses to the environment it is important to understand the nitrogen flows in milk production.

To describe the efficiency, or the pollution intensity, of a product or an agricultural system, much research has used some variant of indicators called nitrogen surplus or nitrogen use efficiency (NUE). Nitrogen surplus is the difference between nitrogen inputs and outputs, and NUE is the quotient between nitrogen outputs and inputs [58, 59]. Thus, the two indicators are different ways to give the same information since $\text{surplus} = \text{inputs} \cdot (1 - \text{NUE})$. However, there are different ideas about the appropriate system boundaries, or concretely, what exactly should be counted as inputs and outputs [59–63]. An important example of system boundary decisions is how to account for nitrogen losses that occur outside a farm, e.g., when dairy farms purchase feed produced on other farms. Several of the above-mentioned references propose something like a "chain nitrogen surplus", which in addition to nitrogen surplus on the dairy farm also accounts for nitrogen surplus associated with feed production on other farms, so as to make the results comparable across farms that produce their own feed or purchase feed.

Another indicator with system boundary wider than the farm gate is the nitrogen footprint, which has been defined by Leach et al. [64]

as the total amount of reactive nitrogen losses to the environment associated with consumption of products. The nitrogen footprint has been proposed to be used in a number of different ways [65, 66, and Paper C in this thesis], one of which is to compare different food products or production systems.

Research questions and method

The research questions we asked in Paper B are the following: How do nitrogen flows differ between conventional and organic milk production in Sweden? How are the differences reflected in the nitrogen footprint and in two variants of the nitrogen surplus with different system boundaries? What are major uncertainties in the calculations? What are the indicators useful for?

To answer these questions we worked mainly with a dataset of farm-gate nitrogen budgets from Swedish dairy farms. The data was extracted from a large database of Swedish farm-gate nutrient budgets collected since 2001 by the Swedish Board of Agriculture within its nation-wide advisory program Focus on Nutrients. We designed selection criteria to extract only specialized dairy farms, which we believe quite well represent most Swedish milk production, and extracted 1566 conventional and 283 organic farms. The farm-level data is detailed and of high quality (see Paper B for details), but since it does not specify what happens outside the farm we used various other data sources to estimate the nitrogen flows associated with feed production outside the dairy farms.

We combined these data sources to estimate flows in a model as illustrated in Figure 3.3 and to estimate three indicators for the nitrogen pollution associated with the milk: farm-gate nitrogen surplus, chain nitrogen surplus, and the nitrogen footprint. All three indicators were expressed in terms of surplus or losses per unit of milk nitrogen. The nitrogen footprint calculation was truncated to include only the dairy farms and the production of feed that they purchase. Hence, our footprint calculation is not as comprehensive as the original definition by Leach et al. [64], but similar in scope to a calculation of nitrogen footprint of food products in the EU by Leip et al. [67].

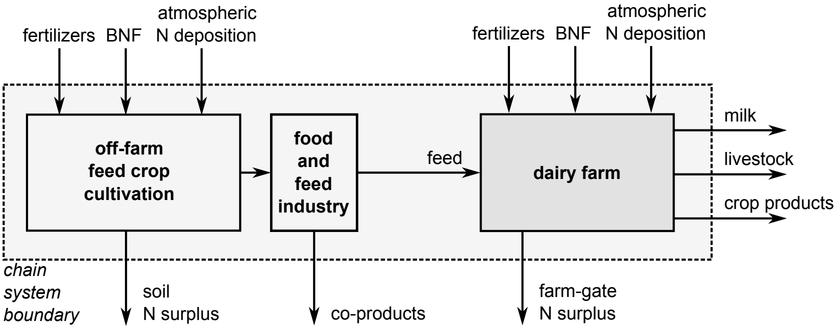

Figure 3.3: Illustration of the system boundaries used in Paper B. The farm-gate nitrogen surplus has the dairy farm as system boundary (darker gray box). The two other indicators, chain nitrogen surplus and nitrogen footprint, both use the wider chain system boundary (lighter gray with dashed border). Figure by Einarsson, Cederberg and Kallus in Paper B, under Creative Commons license.

Results

In both organic and conventional Swedish milk production, a majority of the nitrogen surplus occurs on the dairy farms: in the conventional and organic systems, about 90 % and 95 %, respectively, of the milk's chain nitrogen surplus was on the dairy farms (see Figure 3.4). In other words, the chain surpluses were only about 5–10 % higher than the farm-gate surpluses. Our best estimate is that all three indicators have statistically significantly smaller values for the organic milk production than the conventional. See Paper B for further elaboration on differences between the indicators and between the systems.

Another central result, which came out of the uncertainty analysis, is that there are important uncertainties in the estimated BNF rates in legume cultivation, that might change some of our conclusions as BNF estimates are improved in the future. We included several uncertain parameters in a Monte Carlo uncertainty analysis (see Paper B, supporting information) which identified a potential bias in BNF estimates as an important uncertainty. If BNF estimates would change, then also the difference in indicator values between conventional and organic would change since the organic system has a much larger input of BNF and thus is more sensitive to a potential bias. Assuming that BNF is underestimated by 15 % or more, the conclusion would even change to that conventional and organic milk are not significantly different in terms of nitrogen footprint (see Figure 3.5).

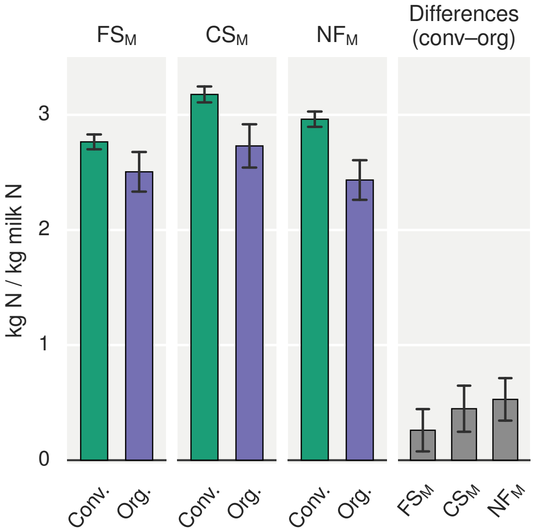

Figure 3.4: Comparison of farm-gate nitrogen surplus (FS_M), chain nitrogen surplus (CS_M), and nitrogen footprint (NF_M) between conventional and organic milk. The error bars show 95 % confidence intervals. The differences between conventional and organic are significantly positive in all cases; in other words, the conventional indicator values are significantly higher. Figure by Einarsson, Cederberg and Kallus in Paper B, under Creative Commons license.

Finally, Paper B also discusses the qualities of the three different indicators. We concluded that, in contrast to the chain nitrogen surplus, the nitrogen footprint has a clear physical meaning connected to a product. However, at least the farm-gate nitrogen surplus still has a role to play when expressed per unit area since it gives different information than the nitrogen footprint. We also highlighted that all three indicators aggregate nitrogen flows over time and space, and of different chemical forms, which have different effects in the environment. Thus, none of the indicators should be mistaken for measures of actual environmental impacts.

Discussion

Which potential errors should be investigated?

Paper B demonstrates the effects of a potential bias in BNF estimates on indicator values. Although this analysis is a bit speculative,

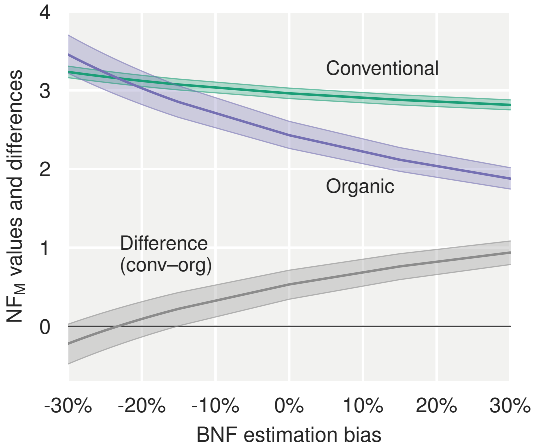

Figure 3.5: Sensitivity of the N footprint to a possible bias in the BNF estimates. The shaded bands around each line show 95 % confidence intervals. If the model overestimates BNF (positive bias values), the N footprints are lower than our best estimates (Figure 3.4) for both systems. Conversely, if BNF is underestimated (negative bias), the N footprints are higher than we think. The effect is stronger for organic milk since organic farms have more BNF input per unit milk, and therefore, the difference between organic and conventional N footprint is not significant at the 95 % confidence level if BNF has been underestimated by 15% or more. Figure by Einarsson, Cederberg and Kallus in Paper B, under Creative Commons license.

it is valuable as it draws attention to an important uncertainty that is not reflected in the confidence intervals shown in Figure 3.4. If there is remaining uncertainty in estimates of BNF rates, there is also a remaining uncertainty in the comparison of organic and conventional systems, regardless of how many farm-gate nitrogen budgets are available.

But why focus specifically on a potential bias in BNF estimates? One can come up with any number of such potential errors that could invalidate any scientific result. My best answer to this question is as follows. It is useful to investigate potential errors in any analysis, but it is not possible to investigate them all. First of all, the only candidates are the “known unknowns” — the things that we know or suspect to be errors. Second, there is always limited time to put into an analysis. Thus, choosing which potential errors to investigate is a judgment call, where it is reasonable to consider (1) which potential errors do we believe to be most severe or relevant, (2) which potential errors do we have a method to investigate, and (3) how much work would it be to investigate them?

Judging by these standards, I think the uncertainty analysis in Paper B was a good use of time: (1) we believed that it could be substantial and we understood that it would affect the organic and conventional system differently, (2) we did have a method and we did have some indication of the possible size of the bias, and (3) it was not too much work to do. In the end, I think the analysis highlighted an important uncertainty. However, there are two other important uncertainties that Paper B does not fully acknowledge, namely those surrounding soil nitrogen stocks and denitrification.

Denitrification and soil nitrogen stocks

How large changes might there be in agricultural soil nitrogen stocks? Extrapolating from the estimates of soil nitrogen accumulation from the Mississippi river basin [38, Section 2.4], it seems reasonable to consider accumulations up to 30–50 $\mathrm{kg\,N\,ha^{-1}\,y^{-1}}$. On the other hand, according to the models in the above-mentioned review [39], European agricultural nitrogen stocks are constant or decreasing by as much as 3.5 $\mathrm{Tg\,N\,y^{-1}}$. Translated to an average over the EU UAA, this is a decrease of 20 $\mathrm{kg\,N\,ha^{-1}\,y^{-1}}$, so the potential error in the constant stock assumption is substantial. But it is far from obvious how to analyze these possibilities. Should we expect the same rate of change on organic farms despite the lower rate of inputs there? If not, what basis is there for assumptions about differences in soil nitrogen accumulation on organic and conventional farms?

Denitrification to N_2 is difficult to measure and therefore N_2 is often calculated as a balancing term, according to a review of nitrogen budget models by De Vries et al. [39]. Estimates of N_2 emissions from European agriculture in the IDEAg, INTEGRATOR, MITERRA, and IMAGE models vary in the range 2.5–7.2 $\mathrm{Tg\,N\,y^{-1}}$, or an average of 14–41 $\mathrm{kg\,N\,ha^{-1}\,y^{-1}}$ if averaged on the $1.7 \cdot 10^8$ ha of utilized agricultural area (UAA) in the EU [39, 68]. These emission estimates are substantial, but note that the N_2 emission estimates are balancing terms in models with zero or negative accumulation of soil nitrogen. De Vries et al. [39] concluded the discussion on N_2 emissions that “it would be worthwhile to put more emphasis on the measurement of N_2.”

System boundaries, denitrification, and stock changes

In Paper B we estimated nitrogen footprints using a mass balance approach, assuming no change in soil nitrogen stocks and not accounting for possible denitrification to N_2. Hence, if there is soil nitrogen accumulation or denitrification that leads to decreased nitrogen losses to the environment, then the nitrogen footprint estimates in Paper B are inflated.

However, the solution to this puzzle depends not only on the physical flows, but also on the system boundaries. The nitrogen footprint is defined as “the total amount of Nr [reactive nitrogen] released to the environment as a result of an entity’s resource consumption” [64]. Thus, the definition relies on a distinction between the environment and the non-environment, and on tracking the flows of reactive nitrogen between them. This observation is relevant both for stock changes and denitrification to N_2.

If an activity leads to nitrogen accumulation inside the non-environment, then that accumulation does not increase the nitrogen footprint of the activity. For example, if the activity accumulates ammonia in a steel container, it is clear that there is no nitrogen footprint of the activity. But what about nitrogen accumulation in agricultural soil? Is that to be considered a loss of nitrogen to the environment, or as an accumulation in the non-environment?

A similar argument can be made for denitrification. For example, nitrogen footprint calculations usually account for denitrification to N_2 that occurs in sewage treatment systems [64, 69–71]. This denitrification occurs inside the non-environment and therefore decreases nitrogen losses to the environment compared to releasing untreated sewage. In contrast, denitrification to N_2 in wetlands or rivers is not deducted from nitrogen footprints, since wetlands and rivers are clearly part of the environment. But again, what about denitrification in agricultural soils, or in buffer strips between cropland and a water course? If and only if such areas are considered part of the non-environment, then their denitrification to N_2 should be deducted from nitrogen footprints just like for sewage treatment systems.

In summary, the choice of system boundary is crucial because it determines how soil accumulation and denitrification in the agricultural landscape contribute to the nitrogen footprint. These issues were largely ignored in Paper B, but I think it would be valuable to discuss them further.

Conclusions

Paper B contributes to the understanding of nitrogen flows in agricultural systems by analyzing and comparing nitrogen flows in Swedish organic and conventional milk production, and quantifying three indicators for the nitrogen pollution associated with the milk. While the best estimate is that the organic system leads to less nitrogen surplus and smaller nitrogen footprint per unit milk, this conclusion is sensitive to a potential bias in BNF estimates. The paper also highlights that the simplifications made in all three indicators hide many complexities with environmental relevance, which means that they can be misleading for decision-makers.

3.3 Paper C

Motivation

The nitrogen footprint definition by Leach et al. [64] was published in 2012, so the nitrogen footprint is a relatively young invention compared to the more well-known ecological, carbon, and water footprints [72]. However, a number of publications have already proposed various applications of the nitrogen footprint, mainly to inform consumers and policy-makers in different ways about the nitrogen pollution associated with consumption [65–67, 69, 70, 73–76].

While some footprint metrics have been quite successful in raising awareness of environmental pressures among the public [72, 77], they have also been heavily criticized in recent years, not least by members of the life cycle assessment (LCA) community who perceive footprints as confusing and misleading since they have overlapping scopes and sometimes measure environmental flows without regard to their actual environmental impacts [78–80]. An example of this last point is the water footprint, which has been criticized for comparing apples and oranges as it sums up, e.g., rainwater use in Norway and groundwater use in Morocco into one number. In LCA terminology, an indicator that does not measure equivalent units of environmental impacts lacks environmental relevance. A task force of the UNEP-SETAC Life Cycle Initiative has proposed to require environmental relevance of every footprint [80].

Since the nitrogen footprint aggregates reactive nitrogen flows over time, space, and of different chemical forms (see Paper B) it is possible to argue that it compares apples and oranges just like the water footprint. Thus, it is timely and relevant to look closer at the nitrogen footprint and the concept of environmental relevance and consider whether and how the two ideas should be reconciled.

Research questions and method

In the Introduction of Paper C I ask the following questions: "What is environmental relevance, and is it a reasonable goal? What limits are there to the environmental relevance of the nitrogen footprint? Is it relevant enough for its proposed applications? What sort of changes, if any, would be appropriate to make in the definition of the nitrogen footprint?"

The approach is (1) to use previous research on the carbon and water footprints to understand the concept of environmental relevance, (2) to review proposed applications of the nitrogen footprint, (3) to analyze whether the nitrogen footprint is relevant enough for its proposed applications, and (4) to consider what the previous points say about the possibility to improve the environmental relevance of the nitrogen footprint.

Results and conclusions

Paper C demonstrates that the concept of environmental relevance is not as straightforward as some members of the LCA community have implied. First, the debate over water footprints has clarified that a one-sided focus on environmental relevance is much too narrow compared to the original intention with the water footprint, and this is a reason that well-meaning attempts from the LCA community to adjust the water footprint [81, 82] have not necessarily been seen as improvements by the authors behind the original water footprint definition [83, 84]. A further complication is that environmental relevance is ultimately a subjective concept: even the de facto standard to weigh different greenhouse gases in the carbon footprint in terms of 100-year global warming potential is a design choice that cannot fundamentally be motivated on objective grounds.

In summary, Paper C is an attempt to learn from the history of other footprints to investigate what the limited environmental relevance of the footprint means for its proposed use cases, and to consider ways forward. I argue in the paper that the proposed use cases of the nitrogen footprint can be placed on a scale, roughly speaking, between less and more demanding in terms of environmental relevance, and therefore the way forward depends on what use cases are envisaged. In any case, it must be acknowledged that the work has a large subjective component.

CHAPTER 4

Discussion and outlook

The assessment of reactive nitrogen flows in agricultural systems relates to a wider ongoing debate on sustainable development and food provision for humanity. Much of this debate has been neatly summarized by Struik et al. [85] as an intersection of three themes: the right to food, agricultural intensification, and sustainability.

The right to food was pronounced by the United Nations Special Rapporteur on the Right to Food [86] as "the right of every individual [...] to have physical and economic access at all times to sufficient, adequate and culturally acceptable food that is produced and consumed sustainably, preserving access to food for future generations."

The intensification and sustainability components are often bundled in the term "sustainable intensification", which is according to the Royal Society a process where "[crop] yields are increased without adverse environmental impact and without the cultivation of more land" [87]. While this aim seems to be widely supported, it is a relatively narrow formulation of sustainability: it does not mention issues of global equity, sufficient nutritional quality, rural economies, or animal welfare, just to name a few [85, 88]. There is much room for discussion on the sustainability component.

One view is that sustainability must be seen as a hierarchy of considerations: first, there are biophysical limits to how much environmental change humanity can accept; and second, only within those limits, are different options for achieving long-term viability of economic and social systems [89]. One attempt to formalize and quantify this notion are the so-called planetary boundaries, a set of quantitative parameter ranges defining a "safe operating space for humanity" [90, 91]. But the planetary boundaries have been criticized for being neither planetary nor boundaries: not planetary since many environmental problems are of local character [92, 93]; and not boundaries since many forms of environmental degradation are gradual rather than abrupt [92, 94], and if there are indeed tipping points the parameters might not capture the complexity of the matters anyway [95, 96]. Although further refinements of the planetary boundaries [91] have tried to address the local character of some problems—the boundary for anthropogenic nitrogen fixation being one example [97]—I still find the concept problematic since there is ultimately no objective way to determine an acceptable level of environmental degradation.

Nevertheless, the planetary boundaries concept has received much attention. I interpret this as (1) a broad agreement that environmental protection must be prioritized, and (2) a demand for tangible scientific guidance to set policy targets. So what shape should that guidance take if not planetary boundaries? I would like to propose two starting points, at least for the nitrogen-related issues:

First, I think it is useful to put more emphasis on the regional character of nitrogen pollution and mitigation options. Policies to control nitrogen losses to the environment are in many cases already specified and monitored at the appropriate local or regional level, for example the EU Water Framework Directive and the HELCOM Baltic Sea Action Plan. In designing and understanding such policies, global, continental-scale, or national assessments may be useful tools. The refinement of the planetary nitrogen boundary [97] took a step in this direction by proposing a globally applicable method for deriving regional boundaries that also accounts for different chemical forms of nitrogen losses. A notable European example which is policy-relevant, but without defining boundaries, is the European Nitrogen Assessment which has collated spatially explicit data and model results on reactive nitrogen flows across the EU [34]. Research is also making progress on methods for impact assessment with high spatial resolution. For example, Hansen et al. [98] have explored the possibility of more spatially targeted regulation to decrease nitrogen loads from agriculture in Denmark, accounting for natural reduction of nitrate in groundwater; specifically interesting is their analysis of how merely relocating existing agricultural practices could decrease nitrogen loads from a catchment. Another interesting development is that the LCA community is working on more regionalized impact assessment, e.g., spatially differentiated tools to assess marine eutrophication [99, 100].

One way that I and my colleagues could contribute to spatially differentiated information is to use the large amount of detailed farm-level data—almost 19,000 farm-gate nutrient budgets—from the Focus on Nutrients project (see Section 3.2) to build a more detailed picture of Swedish agriculture than national statistics provide, and hopefully we could use this detailed picture to reason about how nitrogen loads to the environment might change through technology, structural change in agriculture, and dietary shifts.

Second, I think that much more information is needed on the trade-offs between yields and different types of nitrogen-related environmental effects. More generally, strong arguments in favor of analyzing trade-offs in agriculture have been given by, e.g., Struik et al. [85], Garnett et al. [88] and Garnett and Godfray [101]. Taking a step back from the ambition to define boundaries—to instead identify and describe relevant trade-offs in an accessible way—is to open up for discussion in a wider community of stakeholders. Researchers have a central role in identifying and describing relevant effects of anthropogenic nitrogen use, and to do so in a way that fosters informed and balanced debate among those that ultimately decide what actions to take.

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