Modelling site-dependent environmental impacts of nitrogen fertiliser use in life cycle assessments of crop cultivation

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Doctoral thesis
Swedish University of Agricultural Sciences
Uppsala 2019

Acta Universitatis agriculturae Sueciae
2019:70

ISSN 1652-6880
ISBN (print version) 978-91-7760-458-7
ISBN (electronic version) 978-91-7760-459-4

Print: SLU Service/Repro, Uppsala 2019

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Abstract

Use of mineral nitrogen fertilisers in crop cultivation has enabled substantial yield increases, strengthening global food security. High yields also allow better resource efficiency and result in higher organic matter inputs to soil, increasing the potential for soil carbon sequestration. However, nitrogen fertilisers cause substantial greenhouse gas emissions and nutrient losses to water bodies when the excess nitrogen leaves the field in reactive form. Thus nitrogen fertiliser can either increase or decrease the environmental impact of crop cultivation, depending on soil management, site characteristics and the aspects considered.

Life cycle assessment (LCA) is a commonly used tool to assess the environmental impact of crop cultivation. In LCA, the impacts of all or part of the life cycle of a product, process or service are compiled. For crop cultivation, this generally includes production of inputs, machinery use and soil emissions. However, reactive nitrogen emissions, yield response and soil organic carbon dynamics are highly dependent on site conditions, relationships often poorly depicted in LCAs.

This thesis examined the influence of nitrogen fertiliser rate and site on the climate impact and marine eutrophication of crop cultivation as determined by LCA. Methods for quantifying nitrogen emissions from crop cultivation and their impacts were compared, and new methods for assessing marine eutrophication impacts in Sweden and including soil fertility effects of yield increase were developed.

The results showed that nitrogen fertiliser rate influenced the climate impact and marine eutrophication of crop cultivation, but that the effect of site was generally stronger. Site affected the two impact categories differently and also affected the nitrogen rate that gave the lowest impact. The level of impact and the effect of nitrogen rate and site also varied considerably with methodological choices, including: emissions models for soil nitrous oxide and nitrogen leaching, marine eutrophication characterisation model and accounting for the symbiotic relationship between yield and soil organic matter dynamics. These findings highlight the importance of careful model selection and interpretation of results when using LCA to assess the environmental impact of crop cultivation.

Keywords: LCA, crop cultivation, greenhouse gases, climate impact, eutrophication, nitrous oxide, leaching, soil organic carbon, spatial differentiation

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Platsberoende modellering av kvävegödselanvändningens miljöpåverkan i livscykelanalyser av växtodling

Sammanfattning

Användningen av kvävegödsel i växtodlingar har bidragit till att förbättra den globala livsmedelsförsörjningen genom att höja skördarna, vilket också ger bättre förutsättningar för resurseffektiv odling. Hög avkastning ger dessutom större potential att binda in kol i marken. Å andra sidan har kvävegödslingen också negativ påverkan på miljön, både under produktionen av mineralgödseln och när kvävet som inte tas upp av växten släpps ut. Detta ger bland annat växthusgasutsläpp, som påverkar klimatet, och tillskott av näringsämnen till vattendrag, som orsakar övergödning. Hur stor miljöpåverkan blir beror på odlingsmetoder, markens egenskaper, den geografiska platsen och vilka aspekter som inkluderas när miljöpåverkan utvärderas.

Livscykelanalys (LCA) är ett verktyg som ofta används för att bedöma miljöpåverkan av växtodling. I en LCA sammanställs miljöeffekterna av hela eller delar av en produkts livscykel. För växtodling innefattar det oftast produktionen av insatsvaror (till exempel gödsel, bränsle och bekämpningsmedel), maskinanvändning och markutsläpp av till exempel kol- och kväveföreningar. LCA-studier tar dock oftast inte hänsyn till att mängden markutsläpp varierar på grund av odlingsplatsens egenskaper.

I denna avhandling användes LCA för att undersöka hur växtodlingens klimatpåverkan och bidrag till marin eutrofiering (övergödning) påverkas av odlingsplats och hur mycket kvävegödsel som appliceras. Metoder för kvantifiering av växtodlingens kväveutsläpp och dess miljöeffekter jämfördes. Dessutom utvecklades en ny metod för att bedöma marin eutrofieringseffekt i Sverige, samt en ny metod för att inkludera bördighetseffekten av ökad tillförsel av organiskt material när skörden ökar.

Resultaten visade att mängden kvävegödsel som appliceras påverkar både klimatpåverkan och den marina eutrofieringseffekten av växtodlingen, men att odlingsplatsen i allmänhet hade ännu större betydelse. Platsen påverkade dessutom de två olika miljöeffekterna på olika sätt, och påverkade vilken kvävegiva som gav lägst miljöpåverkan. Metodval för beräkning av lustgasutsläpp och kväveläckage på fältnivå, karaktäriseringsmodell för marin eutrofiering, samt hur sambandet mellan skörd och organiskt material i marken modelleras hade också en stor betydelse för den kvantifierade miljöpåverkan.

Nyckelord: LCA, växtodling, växthusgaser, klimatpåverkan, eutrofiering, övergödning, lustgas, kväveläckage, markkol, platsberoende

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Dedication

To my sister Kim,
who will not read this thesis but still be its most devoted supporter

Jag bor i en värld av sprakande solar, strålande stjärnor och frodiga planeter
Supernovor, svarta hål och vilda kometer
Öppna hav, forsande floder och heta vulkanaktiviteter
Sagoskogar, myllrande liv, mysterier och hemligheter
som fyller oss med tacksamheter, ödmjukheter och visioner om hållbarheter,
för vi vet att den här världen har begränsade kapaciteter.

Ändå bor vi i en värld där vi varje minut skövlar, plundrar och bränner ner flera kvadratkilometer
av samma värld, exakt samma värld.
Emil Jensen

Contents

List of publications

This thesis is based on the work contained in the following papers, referred to by Roman numerals in the text:

I Henryson, K.*, Hansson, P.-A. & Sundberg, C. (2018). Spatially differentiated midpoint indicator for marine eutrophication of waterborne emissions in Sweden. *The International Journal of Life Cycle Assessment* 23, 70-81.

II Henryson, K.*, Sundberg, C., Kätterer, T. & Hansson, P.-A. (2018). Accounting for long-term soil fertility effects when assessing the climate impact of crop cultivation. *Agricultural Systems* 164, 185-192.

III Henryson, K.*, Hansson, P.-A., Kätterer, T., Tidåker, P. & Sundberg, C. (in press). Environmental performance of crop cultivation at different sites and nitrogen rates in Sweden. *Nutrient Cycling in Agroecosystems* 114(2), 139-155.

IV Henryson, K.*, Kätterer, T., Tidåker, P. & Sundberg, C. Impacts of nitrogen soil emissions in life cycle assessment of cereal cultivation– a comparison of different modelling options. Manuscript.

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The contribution of Kajsa Henryson to the papers included in this thesis was as follows:

I Planned the paper together with the co-authors. Carried out the data collection and calculations. Wrote the paper with support from the co-authors.

II Planned the paper and developed the modelling approaches together with the co-authors. Carried out the data collection and calculations, with assistance from one of the co-authors. Wrote the paper with input from the co-authors.

III Planned the paper together with the co-authors. Carried out the data collection, calculations and analysis. Wrote the paper with input from the co-authors.

IV Planned the paper together with the co-authors. Carried out the model selection, data collection and calculations. Wrote the paper with input from the co-authors.

Abbreviations

AGTP	Absolute global temperature change potential
CH_4	Methane
CO_2	Carbon dioxide
CO_2eq	Carbon dioxide-equivalents
CU	Cereal unit
GWP	Global warming potential
ha	Hectare
HELCOM	Baltic Marine Environment Protection Commission
ICBM	Introductory Carbon Balance Model
IPCC	Intergovernmental Panel on Climate Change
LCA	Life cycle assessment
N_2	Di-nitrogen gas
N_2O	Nitrous oxide
Neq	Nitrogen-equivalents
NH_3	Ammonia
NH_4^+	Ammonium
NO_3^-	Nitrate
NO_x	Nitrogen oxides (NO and NO_2)
PLC	Pollution Load Compilation
SOC	Soil organic carbon
SOM	Soil organic matter

1 Introduction

Anthropogenic activities are imposing a heavy load on the environment, to the point where we now have pushed the Earth system beyond many of the estimated planetary boundaries (Steffen *et al.*, 2015). Agricultural activities contribute significantly to this load, so decreasing the environmental impact of agricultural activities while providing food, feed, fibre and energy for a growing global population is important to achieve a sustainable society (Rockström *et al.*, 2017). A fundamental challenge is fertiliser management (Mueller *et al.*, 2012). The invention of the Haber-Bosch technique and subsequent introduction and widespread use of mineral nitrogen fertilisers has completely changed the conditions for agricultural production systems and increased global food security (Sutton *et al.*, 2013; Smil, 1999). However, bringing new reactive nitrogen into the biosphere has also had large environmental costs (Sutton *et al.*, 2011). Added nitrogen can form nitrous oxide (N_2O), a powerful greenhouse gas, or nitrate (NO_3^-), ammonia (NH_3), ammonium (NH_4^+) and nitrogen oxides (NO_x), which can subsequently form N_2O or cause eutrophication in water bodies. The yield increase following fertilisation also has a substantial effect on soil organic carbon (SOC) accumulation in soil and the amount of land required to produce a certain amount of crop (Kätterer *et al.*, 2012; Balmford *et al.*, 2005). Fertiliser use in agriculture thus has many effects on the agricultural system and its environmental impacts, and adequate tools are needed to evaluate this environmental impact.

One tool frequently used to assess the environmental impact of products and services is life cycle assessment (LCA). In contrast to many other environmental assessment tools, LCA focuses on quantifying different environmental effects of all activities required to produce the product or service, rather than one specific emission or activity. One purpose of this wider perspective is avoiding actions that, instead of decreasing the overall environmental impact, shift the burden to other parts of the product's life cycle, other types of environmental impact or other geographical locations (Finnveden *et al.*, 2009). Considering the life cycle perspective when analysing the environmental consequences of a product was first done for beverage packaging, around 1970 (Hunt *et al.*, 1996). Today, LCA is used in product development, process design, education, marketing, product labelling and policy, including for agricultural products (Notarnicola *et al.*, 2017; European Commission, 2013; Hillier *et al.*, 2011; European Parliament, 2009; Cooper & Fava, 2006).

In terms of structure, LCA is a framework that needs to be filled with adequate data and models to produce relevant results. There are certain standards for how to perform LCA (ISO, 2006b; ISO, 2006c), but there is still substantial freedom for the practitioner to make methodological choices depending on the goal and scope of the study, availability of appropriate data and models, desired accuracy and time constraints. Quantifying emissions from crop cultivation can be particularly difficult due to the dominance of diffuse emissions, not least those related to nitrogen compounds. These emissions and their impact on the environment tend to vary depending on soil conditions, climate and geographical location (Rochette *et al.*, 2018; Tysmans *et al.*, 2013; Kyllmar *et al.*, 2006). However, models typically used in LCA to estimate these emissions often neglect the influence of cultivation site on emissions and their impacts. This introduces large uncertainties, which can limit the usefulness of the LCA results (Notarnicola *et al.*, 2017; Camargo *et al.*, 2013). Spatial differentiation in emissions modelling and impact characterisation has therefore been identified as an important step towards increasing the credibility of agricultural LCAs (Notarnicola *et al.*, 2017; Reap *et al.*, 2008).

2 Aim and structure

2.1 Aim and objectives

The overall aim of this thesis was to contribute to the development of life cycle assessment methodology that better represents actual climate and marine eutrophication impacts of crop cultivation, focusing in particular on the impacts of mineral nitrogen fertiliser use. The work included developing new methods that are specifically applicable for evaluating crop cultivation in Sweden. These methods were compared with existing methods, especially regarding their representation of differences between sites and fertiliser regimes. Specific objectives were to:

- Develop a new spatially differentiated characterisation method for waterborne emissions in Sweden contributing to marine eutrophication, to be implemented in life cycle assessments (Paper I).
- Explore how the secondary soil fertility effect of yield changes could be included in LCAs, and evaluate how including this would affect estimates of the climate impact of crop cultivation (Paper II).
- Identify how site affects the climate and marine eutrophication impacts of crop cultivation, especially in relation to the influence of fertiliser management (Papers I, III, IV).
- Compare available models of reactive nitrogen emissions from crop cultivation and their impacts (Paper I, IV).

2.2 Thesis structure

This thesis is based on four papers (I-IV), which in different ways describe effects of fertiliser use on environmental impact and examine how methodological choices in LCAs of crop cultivation affect the estimated impacts. During the course of the work, it became obvious that cultivation site fundamentally affected emissions and their subsequent environmental impact, and accounting for this factor thus became an additional theme (*Figure 1*). The focus in the thesis is on climate impact (Papers II-IV) and marine eutrophication (Papers I, III and IV), since these impact categories are highly affected by nitrogen fertiliser management.

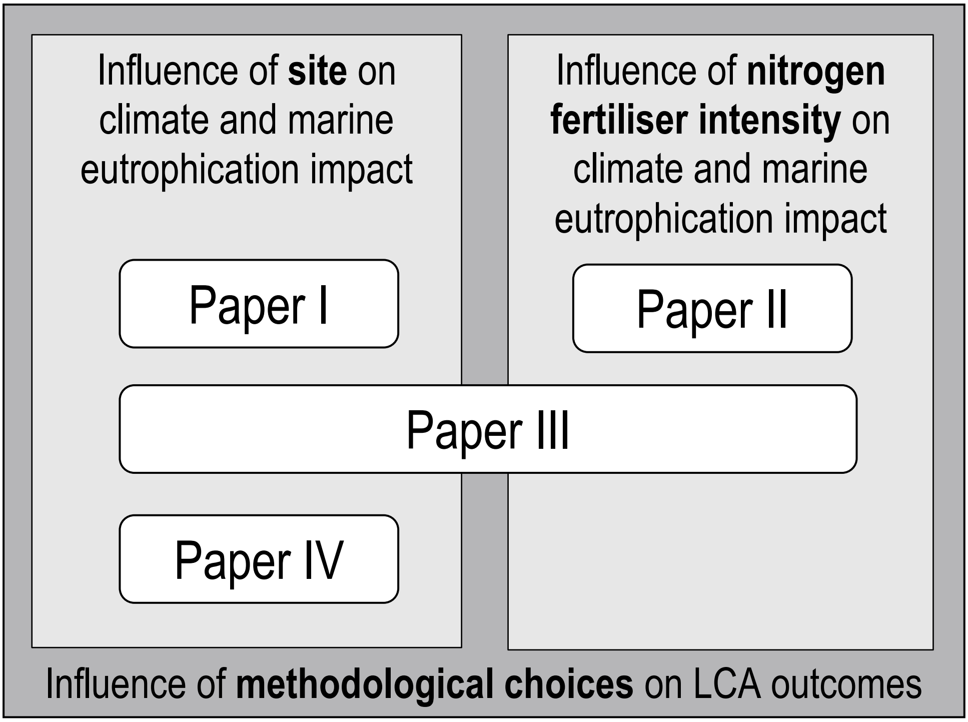


Figure 1. Illustration of the interrelated themes covered in Papers I-IV in this thesis.

In Paper I, a new characterisation model for marine eutrophication impact assessment of emissions to soil in Sweden was developed. With this model, it is possible to account for the site-dependent effect of nutrient losses on marine eutrophication. The model is based on nutrient transport data with high spatial resolution and accounting for the unusual condition (in a global perspective) of phosphorus limitation that occurs in the marine recipients surrounding Sweden.

Paper II presents a new framework for accounting for dynamic interactions between the crop and the soil in LCA, specifically the symbiotic relationship between yield level, soil organic matter (SOM) content and soil fertility. The framework was tested in a case study where a fertiliser-induced yield increase was simulated and the subsequent effects of this yield increase on estimated climate impact were accounted for at different levels of detail.

In Paper III, the climate impact and marine eutrophication effect of crop cultivation at three fertiliser levels at nine sites in Sweden were estimated using LCA. Data from long-term field trials were used as a basis for the assessment, together with site-dependent emissions models and the characterisation method for marine eutrophication developed in Paper I.

In Paper IV, different models for estimating soil N_2O emissions, nitrogen leaching and marine eutrophication impact were applied for winter wheat cultivation at two sites in Sweden. These models represented different approaches to accounting for the site-dependent nature of those emissions and impacts.

3 Background

3.1 Consequences of nitrogen fertiliser use

3.1.1 Global nitrogen fertiliser use

Nitrogen is the most abundant element in the Earth's atmosphere, constituting approximately 78%. However, most of this nitrogen exists in the form of di-nitrogen gas (N_2), which is not available for plant uptake. Unreactive N_2 can be transformed to reactive nitrogen forms by nitrogen fixation, mainly biologically by bacteria or industrially by the Haber-Bosch process (Sutton *et al.*, 2013). Nitrogen can enter agricultural systems by either of these routes. Nitrogen fixation by bacteria commonly occurs through their symbiotic relationship with leguminous plants such as beans, peas and clover (Sutton *et al.*, 2013). Crop residues from these plants can provide nitrogen to the soil or the crops can be used as animal feed, and thereafter returned to the soil as manure. Human excreta also contains nitrogen, but globally only a small fraction of this nitrogen is returned to agricultural soils, while the rest is either converted back to N_2 in wastewater treatment plants or lost to the environment in any of its reactive forms (Morée *et al.*, 2013).

Biological nitrogen fixation was the main nitrogen input to cropland until the 1960s (Galloway *et al.*, 2003). In the early 20th century, the Haber-Bosch process was discovered, allowing large-scale production of NH_3 from combining N_2 and hydrogen from fossil sources (Smil, 2001). The output of synthetic mineral nitrogen fertilisers increased substantially around the mid-20th century and was an important factor behind the 'Green Revolution', *i.e.* the dramatic yield increases achieved during the 1960s-1970s especially in developing countries (Smil, 2002; Khush, 1999). Today, approximately 30% of the global transformation of N_2 into reactive nitrogen occurs through the Haber-Bosch process, of which the majority is used for producing fertilisers (Fowler *et al.*, 2013).

Mineral nitrogen fertilisers have contributed to decreasing world hunger and have enabled global population growth (Davidson *et al.*, 2016). However, while the current growth in global nutrient use can mainly be attributed to the production systems in developing countries, biological nitrogen fixation is still the largest nitrogen input to cropland in Africa and South America (Sutton *et al.*, 2013; Liu *et al.*, 2010). This indicates both an uneven distribution of mineral fertilisers globally and a potential future increase in reactive nitrogen inputs in global agriculture (Sutton *et al.*, 2013).

Apart from contributing to increased food security, mineral fertilisers have enabled other transformations of the agricultural system. Crop cultivation and animal husbandry were previously closely interlinked through the necessity of circulating the nutrients in manure back to arable fields, which is no longer the case (Billen *et al.*, 2013). Geographical separation of animal husbandry and crop cultivation has provided opportunities for the specialist agricultural production systems that are common in the developed world today, but has also contributed to the disruption of local nitrogen cycles (Billen *et al.*, 2013; Naylor *et al.*, 2005).

3.1.2 Nitrogen fertilisers in soil

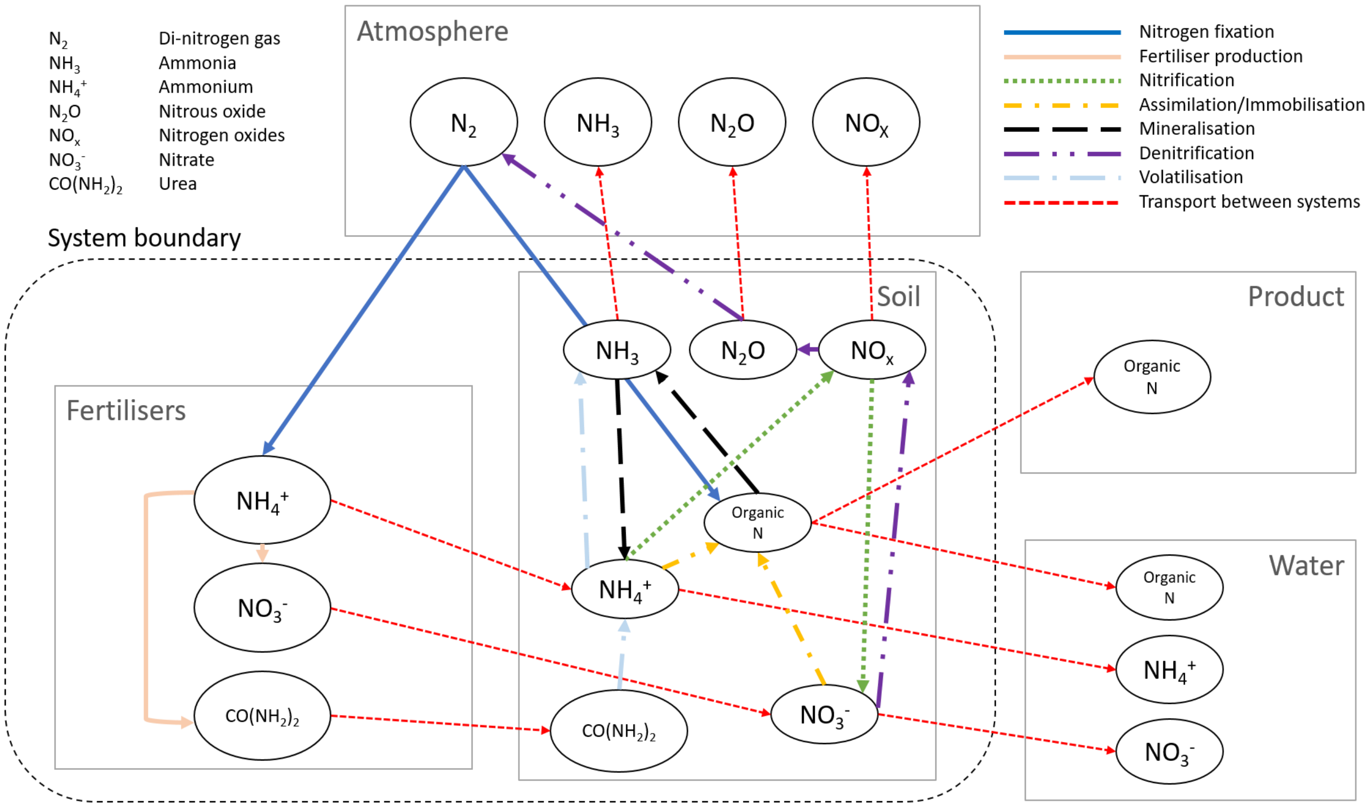
Once N_2 is converted to NH_3 through the Haber-Bosch process, it can be further treated to form mineral nitrogen fertilisers, most of them containing urea, ammonium or nitrate. Globally, urea accounts for about 50% of the mineral nitrogen fertilisers consumed, but it accounts for less than 1% of Swedish mineral nitrogen fertiliser consumption (IFASTAT, 2019). Instead, calcium ammonium nitrate is the most common type of nitrogen fertiliser used in Sweden (IFASTAT, 2019). Other inputs of reactive nitrogen to cropping systems include biological nitrogen fixation, atmospheric deposition, recycled organic material (manure, crop residues, sewage sludge etc.) and nitrogen stocks in the soil (Galloway *et al.*, 2003).

Only about half the nitrogen converted from N_2 to reactive nitrogen is incorporated into the crop, while the rest is lost during fertiliser production, transport and handling, or in the field (Galloway & Cowling, 2002). Some of the soil nitrogen that is not taken up by the crop is denitrified and re-emitted to the atmosphere as N_2 (*Figure 2*), thereby losing its reactive potential. However, some of it is lost to the environment as reactive nitrogen. Site conditions strongly influence both total nitrogen losses from cropland and the relative distribution of different nitrogen forms (Liu *et al.*, 2010). Reactive nitrogen emissions also depend on management factors such as tillage practices, type of fertiliser, timing of fertilisation and fertiliser placement depth in the soil profile (Pan *et al.*, 2016; Sutton *et al.*, 2013).

Gaseous N_2O, NH_3 or NO_x (*Figure 2*) can be emitted and accumulate or react with other compounds in the atmosphere, or be deposited back onto land or water bodies. Other fractions are lost by leaching through the soil profile, primarily in the form of NO_3^- (*Figure 2*). Waterborne nitrogen can eventually reach coastal waters, but a fraction is retained in the landscape or removed as gaseous emissions along the way (Billen *et al.*, 2013). The relative importance of these removal and retention mechanisms depends on factors such as climate, distance between point of emission and the marine environment, occurrence of lakes and wetlands, and river flow rates (Billen *et al.*, 2013; Howarth *et al.*, 2006). Different forms of reactive nitrogen have different effects on the system, and have different potential for being lost to other systems or being transformed and re-emitted to the atmosphere unreactive N_2 (Galloway *et al.*, 2003). However, reactive nitrogen is easily converted between different forms, and can therefore contribute to different environmental impacts (Galloway *et al.*, 2003). Nitrogen incorporated into crops can also eventually end up as reactive nitrogen in the atmosphere or in a water body after consumption by microbes, animals or humans.

Nitrogen cycling in soils is connected to other soil element cycles, notably carbon and phosphorus (Gruber & Galloway, 2008). Carbon and nitrogen are mainly stored in the soil in the form of organic matter, in which they are both fundamental components (Stevenson & Cole, 1999). Consequently, storing carbon as SOC requires immobilisation of nitrogen, while mineralisation (decomposition) of SOM releases both carbon and nitrogen (Luo *et al.*, 2006). Nitrogen also affects the carbon cycle through nitrogen deposition acting as a fertiliser in both terrestrial and marine ecosystems, potentially increasing carbon storage in organic matter (Gruber & Galloway, 2008). Soil organic matter also contains phosphorus, but a significant proportion of phosphorus in the soil is stored in inorganic form (Stevenson & Cole, 1999). Some of this inorganic phosphorus is strongly bound to soil particles and is difficult for plants to access, but it is also less mobile than the NO_3^- dissolved in soil water. Soil phosphorus losses at a certain point in time are therefore less dependent than nitrogen losses on recent inputs, and instead more dependent on the long-term phosphorus balance (Svanbäck, 2014).

Figure 2. Simplified illustration of nitrogen cycling processes associated with mineral fertiliser use in crop cultivation.



3.1.3 Environmental impact of nitrogen fertiliser use

The current level of anthropogenic transformation of N_2 into reactive nitrogen, mainly driven by cropping activities, is exceeding the estimated planetary boundary for a stable and resilient Earth system (Steffen *et al.*, 2015; Smil, 1999). Use of fertilisers inevitably causes losses of reactive nitrogen to the environment, resulting in a plethora of unwanted effects on the climate, ecosystems and human health (Galloway *et al.*, 2003). In addition, fertiliser production requires energy inputs (Kool *et al.*, 2012).

Contributions to aquatic eutrophication

Approximately 13% of the total nitrogen input to the Baltic Sea originates from Sweden, where agriculture is the largest anthropogenic source (Sonesten *et al.*, 2018). Both gaseous and waterborne nitrogen emissions from crop cultivation can eventually end up in a water body, but most of the nitrogen inputs to the Baltic Sea enter through rivers (Sonesten *et al.*, 2018). Nitrogen is lost from soil with the infiltrating soil water, mainly as NO_3^-, but some is also lost as NH_4^+ and dissolved organic nitrogen (Raave *et al.*, 2014; van Kessel *et al.*, 2009). Sandy soils are more prone to nitrogen leaching than clayey soils (Kyllmar *et al.*, 2006; Hoffmann & Johnsson, 1999).

Nutrient addition to aquatic ecosystems causes elevated nutrient levels, a state that is called eutrophication (Smith *et al.*, 1999). Eutrophication affects the ecosystem balance, for example by causing algal blooms and consequent oxygen depletion in aquatic environments (Diaz & Rosenberg, 2008). Eutrophication is a regional impact, but affects most major freshwater bodies and coastal marine ecosystems on Earth (Diaz & Rosenberg, 2008; Smith, 2003). The problem is also expected to increase if climate change increases precipitation and thereby nutrient losses from soils to water (Kanter, 2018).

The environmental damage caused by nutrient addition depends both on the transport from the emission source and the characteristics of the recipient. For example, phosphorus is generally considered to limit plant growth in freshwaters, while nitrogen is considered the limiting nutrient in coastal marine environments (Conley *et al.*, 2009). The Baltic Sea is the world's largest brackish ecosystem and is heavily affected by eutrophication (HELCOM, 2009; Diaz & Rosenberg, 2008; Swedish EPA, 2006). Due to the low salinity, primary production in the Baltic Sea is limited by both nitrogen and phosphorus, with variations between sub-basins and between seasons (Tamminen & Andersen, 2007).

Contributions to climate change

Of the total annual net anthropogenic greenhouse gas emissions of 52 Gt carbon dioxide equivalents (CO_2eq), direct emissions from agriculture are estimated to contribute approximately 12% (IPCC, 2019). The contribution of land use change, which is mainly driven by agriculture, is estimated to be almost as great (9.4%) (IPCC, 2019).

Carbon dioxide (CO_2), methane (CH_4) and N_2O are the most important greenhouse gases emitted from agricultural systems (IPCC, 2019). These emissions occur during the cultivation phase, but also during the production of input materials (although these are not accounted for as agricultural emissions assessments from the Intergovernmental Panel on Climate Change (IPCC)).

In crop cultivation, CO_2 emissions mainly occur during production of input materials, from combustion of fuels in agricultural machinery and as a consequence of SOM mineralisation. Mineralisation of SOM transforms organically bound carbon into CO_2, which is then released to the atmosphere. Since plants also take up CO_2 from the atmosphere and transform it into organic matter, crop cultivation can cause either a net loss of SOC or SOC sequestration. Soil organic matter dynamics are governed by the organic matter input to the soil and the organic matter decomposition rate (Andrén & Kätterer, 1997). The former is strongly influenced by site conditions such as climate and intrinsic soil characteristics, but also by soil management strategies (Andrén *et al.*, 2004). The organic matter input to cultivated soils is related to the productivity, which means that the fertilisation regime can affect the net SOC balance.

Carbon dioxide emissions also often occur when land is transformed to agricultural land. Crop cultivation can cause direct land use change, when crops are grown on land that was previously used for something else, or indirect land use change, when crop cultivation involves land use change at another location distant from the cultivation site (Ahlgren & Di Lucia, 2014). For example, using crops that were previously used for food or feed as biofuels increases demand for agricultural land, which can contribute to conversion of natural ecosystems into cropland elsewhere in the world (Ahlgren & Di Lucia, 2014).

The CH_4 emissions from agricultural systems are mainly caused by ruminating livestock (IPCC, 2019). Cultivated soils can both take up and release CH_4, but the climate impact of the net balance on mineral soils is generally small (Le Mer & Roger, 2001).

Direct N_2O emissions from soils induced by mineral fertiliser use account for 1.5% of the total climate impact in the Swedish greenhouse gas inventory (Statistics Sweden, 2018a). However, it should be noted that in the inventory, soil N_2O emissions are calculated as a function of nitrogen fertiliser use, but in reality also occur from unfertilised soils (Hergoualc'h *et al.*, 2019; Stehfest &

Bouwman, 2006). The N_2O emitted from soils is produced during microbial nitrification and denitrification (Bouwman *et al.* (2002) (*Figure 2*). The magnitude of these emissions varies substantially depending on factors such as availability of soil nitrogen, soil microbial activity, soil water content and temperature (Novoa & Tejeda, 2006). Indirect soil N_2O emissions arise when reactive nitrogen leaves the field as *e.g.* leached NO_3^- or volatilised NH_3 and is transformed to N_2O at another location (Hélias, 2019). Field emissions of several forms of reactive nitrogen thus contribute to the total soil N_2O emissions caused by nitrogen fertiliser. Apart from N_2O emissions from soils, there can be substantial losses of N_2O during mineral fertiliser production, especially if the production process does not use modern N_2O abatement technologies (Kool *et al.*, 2012).

Influence on soil quality

The concept 'soil quality' essentially refers to the ability of soils to deliver a function (Carter, 2002). In the context of agricultural production, this usually means sustaining plant growth. Different indicators have been proposed to quantify soil quality, but SOM is generally viewed as the most important indicator (Vidal Legaz *et al.*, 2017; Hauschild *et al.*, 2013; Garrigues *et al.*, 2012; Brandão *et al.*, 2011; Carter, 2002). Soil organic matter has multiple functions in soil, such as delivering nutrients to plants and microorganisms, and stabilising soil structure. A good soil structure protects soils from erosion, improves their water-holding capacity and provides good conditions for root growth (Carter, 2002).

Many agricultural soils located across different continents and areas with different economic levels have a negative nitrogen balance, *i.e.* the removal by crops and losses exceed the input (Liu *et al.*, 2010). The direct consequence of low nitrogen availability is lower crop yields, which leads to lower organic matter input to the soil (Kätterer *et al.*, 2012). Nitrogen depletion is therefore a threat to soil quality. Excessive nitrogen fertilisation, on the other hand, can cause acidification of soils, which is also a threat to soil quality (Tian & Niu, 2015).

Other impacts

Nitrogen is an essential element for plants and animals. Since reactive nitrogen is often growth-limiting in ecosystems, changes in nitrogen availability have the potential to alter ecosystem balance (Gruber & Galloway, 2008). Apart from biodiversity impacts caused by aquatic eutrophication, atmospheric nitrogen deposition affects terrestrial biodiversity by promoting growth of certain plant species and acidification, causing unfavourable conditions and decreased resilience to stress factors such as pathogens and drought for many species (Wallis de Vries & Bobbink, 2017).

In addition to the effects on the environment, reactive nitrogen can cause various problems for human health. Elevated NO_3^- concentrations in drinking water can cause oxygen deficiency in infants, while NH_3 and NO_x contribute to decreased air quality through promoting formation of fine particulate matter and photochemical smog (Damania *et al.*, 2019; Sutton *et al.*, 2013). However, the global contribution of airborne reactive nitrogen emissions from ammonium nitrate-based fertiliser use is relatively small compared with that from combustion of fossil fuels, manure management and urea use (Pan *et al.*, 2016; Fowler *et al.*, 2013; Sutton Mark *et al.*, 2013). After the sharp reduction in chlorofluorocarbon use during recent decades, N_2O is now the main contributor to stratospheric ozone depletion, increasing the influx of ultraviolet solar radiation with potential damage to both ecosystems and human health (Ravishankara *et al.*, 2009; Solomon, 2008).

3.2 Life cycle assessment

Life cycle assessment is a standardised methodology to assess the environmental impact of products, services or processes, with the purpose of improving environmental performance, internal or external communication, or informing decision-makers at various levels (ISO, 2006b). Life cycle assessment has been used in a wide range of applications (Hellweg & Milà i Canals, 2014). It is generally considered a scientifically sound approach, but it has also been criticised for the freedom of practitioners to make methodological choices and assumptions, which can have a large influence on LCA results (Curran, 2014; Reap *et al.*, 2008).

3.2.1 Life cycle assessment methodology

An LCA consists of four main phases; goal and scope definition, inventory analysis, impact assessment and interpretation.

In the goal and scope definition phase, the product system, the purpose of the study, assumptions and methodological choices are described (ISO, 2006b). Important concepts include:

- The *functional unit*, a quantifiable representation of the system function used as a reference point to which the inventory and subsequent assessed impact is related (ISO, 2006b). The functional unit for crop cultivation is usually related to the area used or yield obtained (Notarnicola *et al.*, 2017).

- The *system boundaries* define the activities that are included in the assessment and should be chosen with respect to the goal of the study (ISO, 2006c). Roer *et al.* (2012) demonstrated that some processes which are often neglected in LCAs of crop cultivation, such as SOM mineralisation, can have a large influence on the final results. The system boundaries also include the boundary between the production system (the 'technosphere') and the environment (the 'biosphere') (Schau & Fet, 2008). The distinction between the technosphere and the biosphere is not obvious when evaluating agricultural processes, since the production system and the environment are highly interlinked in these systems (Schau & Fet, 2008).
- *Allocation* is the process of partitioning the environmental burdens between the main product and one or several more co-products produced in the same process (ISO, 2006c). In an agricultural context, this is necessary *e.g.* when rapeseed is separated into oil and cake, or when crops are cultivated in rotation if the practitioner is interested in assessing the impact of only one of the products. Allocation can be based on *e.g.* monetary value, mass or energy content.
- The selection of *impact categories* should be consistent with the goal of the study, as should the *characterisation models* used to connect the inventory flows to the indicator results (ISO, 2006c). The definition of impact categories varies between studies. For example, effects of nutrient addition can be described as the total eutrophication potential, terrestrial vs aquatic eutrophication, or terrestrial vs freshwater vs marine eutrophication (Morelli *et al.*, 2018; Hauschild & Potting, 2005; Guinée, 2002).

The life cycle inventory process mainly consists of data collection and connecting emissions and resource use across the system boundary (elementary flows) to the functional unit. The elementary flows are then combined with characterisation models representing the connection between them and various types of environmental impacts in the life cycle impact assessment stage. A characterisation model can encompass the whole cause-effect chain, from emission to damage inflicted on the ecosystem or human health (endpoint modelling), or part of the cause-effect chain (midpoint modelling) (Bare *et al.*, 2000).

The interpretation phase includes identifying the main outcomes of the LCA, any weaknesses and their potential influence on the results (ISO, 2006c). The interpretation should analyse the sensitivity of results in relation to data uncertainties, methodological choices and assumptions (ISO, 2006c).

3.2.2 Life cycle assessment of crop cultivation

Application of LCA to agricultural systems poses certain challenges compared with the industrial systems where LCA was first applied (Notarnicola *et al.*, 2017; Keller *et al.*, 2014). The peculiarities of agricultural systems have consequences for LCA practitioners in the input data, emissions modelling and impact assessment phase. Some of these challenges are summarised in this section.

Agricultural products are produced in scattered production systems, with many different producers. This is a challenge for LCA since data collection becomes more time-consuming, but also because both site conditions and agricultural management can vary significantly between farms (Notarnicola *et al.*, 2017; Keller *et al.*, 2014).

For impact categories such as climate impact and eutrophication, the impacts of agricultural activities are often dominated by diffuse emissions occurring in the field, at least in countries such as Sweden where the fertilisers used are produced with modern technology (Ahlgren *et al.*, 2009; Hayashi *et al.*, 2006; Brentrup *et al.*, 2004). This makes emissions difficult to measure and results highly dependent on emissions models.

Agricultural productivity is highly dependent on the environmental conditions at the site, and in turn strongly affects the local environment. An example is the close relationship between crop management and the soil. Soil properties affect the crops that can be grown and how much fertiliser is needed, while crop management affects soil erosion, soil microorganism activity and SOM dynamics. The soil is thus part of the production system and part of the affected environment, making system boundary definition between these systems disputable (Notarnicola *et al.*, 2017).

Agricultural systems are multifunctional, meaning that two or more functions are co-produced. Crop rotations and integrated crop-livestock systems yield more than one marketable product and, even when production is separated in time or space within the farm, the production of one product will inevitably affect the other, and vice versa. Valuation of non-marketable outcomes of agricultural activities such as soil quality and biodiversity, often called ecosystem services, has also been proposed to be included in LCAs in various ways (Boone *et al.*, 2019; Notarnicola *et al.*, 2017; Oberholzer *et al.*, 2012). Multifunctionality affects both the choice of functional unit and the need for allocation of environmental burden between co-products (Notarnicola *et al.*, 2017; Karlsson *et al.*, 2014; Hayashi *et al.*, 2006).

Agricultural production practices are dependent on the conditions at the site at which the production occurs, as are the emissions and the impact of those emissions (Notarnicola *et al.*, 2017). Many of the factors responsible for this variability are connected to nitrogen fertiliser management, such as yield, nitrogen fertiliser amount and emissions from fertiliser production (Clavreul *et al.*, 2017). Earlier studies have reported spatial variations in the environmental impact of crop cultivation of up to an order of magnitude even within countries (Yang *et al.*, 2018). For example, a study of Swedish wheat cultivation showed that climate impact could vary by a factor of three between farms within the same region (Ahlgren *et al.*, 2012). The goal and scope of the study, the spatial resolution of data and the emissions models and characterisation models used also affect the magnitude of variation between studies (Yang *et al.*, 2018; Korsaeth *et al.*, 2014; Ahlgren *et al.*, 2012). Many LCA studies aim to quantify the environmental impact of crop cultivation at a specific site or in a specific region (*e.g.* Heidari *et al.*, 2017; Korsaeth *et al.*, 2012; Ahlgren *et al.*, 2009). However, while such studies apply site-specific data for describing the production practices and yield obtained, they often use site-generic models for estimating emissions and impacts, potentially limiting the usefulness of the results.

The temporal aspect is also important when assessing the environmental impact of crop cultivation. Yields and emissions vary with the particular inter-year weather conditions, and cultivation practices also vary over time. In addition, there is often a time lag between soil management change and soil responses such as soil acidification and SOM accumulation (Li *et al.*, 2019; Kätterer *et al.*, 2008). This means that some of the soil emissions during a particular year are highly dependent on previous soil management (Gan *et al.*, 2012b). For LCA applications, this poses challenges in data collection and methodological concerns regarding allocation of burdens between different land uses over time, *e.g*. accounting for carbon sequestration (Brandão *et al.*, 2013). The basic LCA principles involve temporal integration of emissions and impacts, which is not always suitable for dynamic systems such as land use. Time is also an important factor in climate impact assessment, since there is a significant time lag between emissions and climate response (Levasseur *et al.*, 2016).

3.2.3 Incorporation of the spatial dimension in life cycle assessments of crop cultivation

Life cycle assessment was originally a site-independent tool, but spatial differentiation has been proven to increase the environmental relevance of LCA results and broaden the ability to answer new research questions (Frischknecht *et al.*, 2019; Patouillard *et al.*, 2019). Ultimately, spatial differentiation aims to reduce the uncertainty originating from spatial variability, *i.e.* instead of deriving a site-generic result with large uncertainty, it involves more closely identifying where within that range production at a specific site or in a specific region is likely to be (Patouillard *et al.*, 2019).

Major progress towards spatial differentiation in LCA has been achieved in the past two decades (Patouillard *et al.*, 2018). Enabling spatial differentiation in LCA practices encompasses a wide range of research activities, since the spatial dimension affects several parts of the LCA methodology, from software development to new characterisation models (Frischknecht *et al.*, 2019).

The inventory can be regionalised, *i.e.* adapted to better reflect the situation at the site of production (Patouillard *et al.*, 2018). This can mean either adaption of the processes included or adjustment of their estimated emissions (Patouillard *et al.*, 2018). In a crop cultivation context, the former could be *e.g.* the amount of fertiliser used, while the latter could be the amount of soil emissions assumed to occur due to the cultivation. Ideally, all emissions should be perfectly representative of the (actual or simulated) process, but in reality it is necessary to prioritise regionalisation efforts in order to minimise the additional work needed to compile the inventory (Patouillard *et al.*, 2019). Representative process data can often be found in sources such as databases, public statistics and previous studies. Representative soil emissions are often more difficult to estimate due to the high spatial and contextual variability (Notarnicola *et al.*, 2017; Ahlgren *et al.*, 2012). In most cases, measured data for the specific site and management regime are not available and emissions then need to be modelled. The most common approach is to use simple emissions models such as the IPCC Tier 1 approach, which estimates N_2O emissions, nitrogen leaching and nitrogen volatilisation as fractions of the nitrogen available in fertilisers and crop residues. However, these models typically lack spatial differentiation, so other approaches are necessary if this is to be included. Process-based agroecosystem models such as DAYCENT, DNDC and CERES-EGC have been used in LCAs to estimate soil emissions (Goglio *et al.*, 2018; Goglio *et al.*, 2014; Dufossé *et al.*, 2013; Adler *et al.*, 2007). These models tend to correspond better to measured emissions, but require sufficient calibration data and expert knowledge to apply (Goglio *et al.*, 2018; Gabrielle & Gagnaire, 2008). Therefore, this is often not a viable option for LCA practitioners.

According to ISO14044, the characterisation model used in LCA should consider spatial and temporal differentiation, as well as the fate and transport of substances, if that is called for by the goal and scope and by the environmental mechanism described by the characterisation model (ISO, 2006c). Regionalised impact assessment is relevant for all impact categories where the impact of an emission depends on the emission site (Hauschild & Potting, 2005). Stratospheric ozone depletion and climate impact are considered to be global impact categories, *i.e*. where the impact of an emission is independent of the emission site (Potting & Hauschild, 2004). For other impact categories, accounting for the spatial dependency can significantly influence the results (Owsianiak *et al.*, 2018; Anton *et al.*, 2014).

Applying regionalised impact assessment requires access to regionalised characterisation models for the impact categories of interest. Regionalised characterisation models has been developed for many impact categories, including marine eutrophication (Cosme & Hauschild, 2017), freshwater eutrophication (Helmes *et al.*, 2012), acidification (Roy *et al.*, 2014) and toxicity (Rosenbaum *et al.*, 2008). However, development of new characterisation models is still ongoing and there is still no consensus on appropriate choice of characterisation models (Frischknecht *et al.*, 2019; Bach & Finkbeiner, 2017; Hauschild *et al.*, 2013).

Despite the improved possibilities for site-dependent LCA, few published studies employ spatial differentiation across all LCA phases (see Cosme and Niero (2017) for a notable exception). Considering the potential benefits of employing spatial differentiation in crop cultivation LCAs, there is a need to determine when site-dependent modelling is necessary and identify appropriate models that can be used in the applications.

4 Methods

This chapter provides a general overview of the approaches used in methodological development and evaluation and in the case studies presented in Papers I-IV. For a more detailed description of methods used, see the respective papers.

The calculations for Paper I were carried out in Microsoft Excel 2010, while the calculations for Papers II-IV was carried out in MATLAB (version R2015b and R2018b, The Mathworks, Inc.).

4.1 Methodological development and evaluation

4.1.1 Development of a new characterisation model for marine eutrophication

A new characterisation model for marine eutrophication impact assessment of waterborne emissions from soil in Sweden was developed in Paper I. The characterisation model was based on data from Brandt *et al.* (2009) on retention between emission sites and marine recipients for the whole of Sweden, divided into approximately 1000 zones. The retention data were combined with a binary nutrient limitation factor for each marine recipient on sub-basin level, based on literature data. An equivalency ratio that corresponds to the Redfield ratio was added to enable conversion of phosphorus emissions into the chosen unit, nitrogen equivalents (Neq). After removing zones without agricultural land, this yielded characterisation factors for nitrogen and phosphorus for 968 catchments in Sweden, calculated as:

$$CF_{i,j} = (1 - Retention_{i,j}) * Nutrient\ limitation\ factor_{i,j} * Equivalency\ ratio_i$$

where $CF_{i,j}$ is the characterisation factor for substance i emitted at site j, $Retention_{i,j}$ is the fraction of substance i that is removed or retained between point of emission j and the marine recipient, $Nutrient\ limitation\ factor_{i,j}$ is the sensitivity to substance i in the marine recipient for point of emission j (0 or 1), and $Equivalency\ ratio_i$ is the algae growth potential of substance i in relation to the algae growth potential of nitrogen (1 for nitrogen and 7.23 for phosphorus).

4.1.2 Including the influence of soil organic matter changes on soil fertility in life cycle assessment applications

In Paper II, a modelling framework was designed to incorporate the interactions between soil productivity and SOM dynamics in the assessment of climate impact of crop cultivation. The productivity is represented by grain yield and the SOM dynamics by changes in SOC content. The framework consists of three main modules; one for calculating yield, one for calculating SOC content and one for calculating climate impact (*Figure 3*). All modules use annual time steps, and all variables used in the modules can be modified to represent a specific case.

The yield module uses a reference yield and calculates a new yield depending on the difference in SOC content between the reference situation and the new situation, and a given yield response to that SOC change. The SOC content is simulated using the Introductory Carbon Balance Model (ICBM) (Andrén *et al.*, 2008; Andrén *et al.*, 2004; Andrén & Kätterer, 1997). It is a simple process model that estimates SOC content in the top layer (0-25 cm) of agricultural soils based on the carbon inputs and their characteristics. The model also includes parameters that depend on factors such as soil texture and climate (Andrén *et al.*, 2004; Andrén & Kätterer, 1997). The regional version of the model, ICBMr, where the parameters are dependent on regional conditions (Andrén *et al.*, 2004), was used in Paper II.

The climate impact assessment module in the new framework uses the emissions flows from the SOC dynamics module and other emissions arising during the crop cultivation (fertiliser and pesticide production, machinery and soil N_2O emissions) to calculate the climate impact. Two different climate metrics are included in the framework; global warming potential (GWP) and absolute global temperature change potential (AGTP). The main difference between these metrics is that GWP assesses the cumulative change in radiative forcing over a specific time period, assuming that all emissions occur at the same time, while AGTP assesses the instantaneous temperature change. Estimation of AGTP requires a time-distributed emissions inventory and delivers a time curve representing the climate impact as temperature change (expressed in Kelvin) at any point in time within the chosen time period. Estimation of GWP is based on the accumulated emissions during the study period and delivers a single value for the climate impact (expressed as CO_2eq).

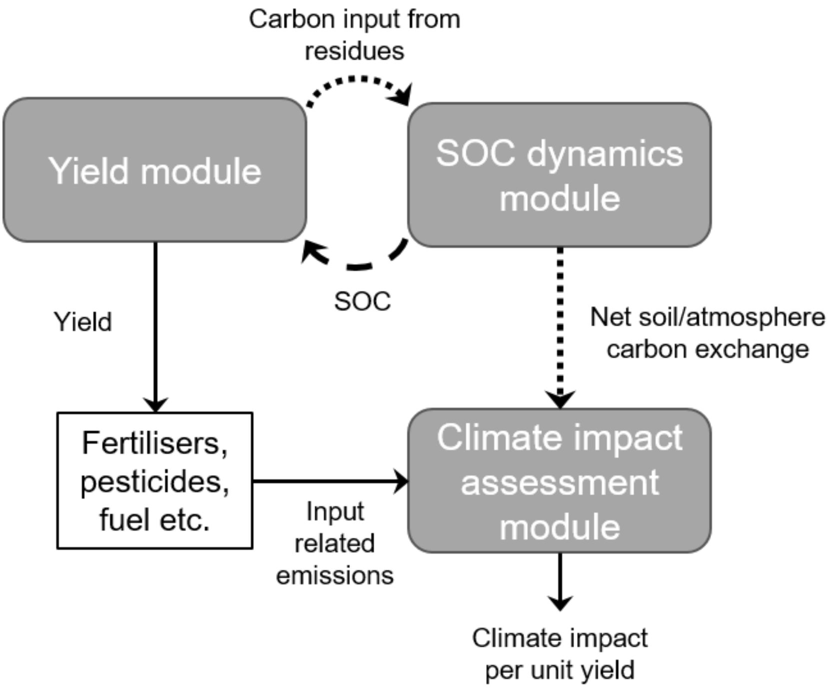


Figure 3. Illustration of the calculation modules (grey boxes), and the information flows (arrows) between these, in the modelling framework developed in Paper II. Solid arrows represent information flows included in all three approaches used in the case study, dotted arrows represent information flows in approaches that include soil organic carbon (SOC) change, and the dashed line indicates an information flow only used in an approach that included yield response to SOC change.

4.1.3 Comparison of models for direct soil N_2O emissions, nitrogen leaching and marine eutrophication impact assessment

Seven soil N_2O emissions models, seven nitrogen leaching models and five characterisation models for eutrophication (eutrophication potential and four marine eutrophication indicators) were compared in Paper IV. The comparison only included medium-effort models that could be applied with data typically available to an LCA practitioner, and which did not require expert knowledge in any agroecosystem process model. The models were compared by first applying site-generic models to wheat cultivation at two sites (see section 4.2), and then re-calculating the impact with each of the alternative models one at a time.

For soil N_2O emission, the aggregated version of the IPCC Tier 1 model (IPCC_agg) (Hergoualc'h *et al.*, 2019) was used as the site-generic model and compared with six site-dependent models: IPCC_disagg (Hergoualc'h *et al.*, 2019), Rochette_tot and Rochette_FI (Rochette *et al.*, 2018), StehfestBouwman_tot and StehfestBouwman_FI (Stehfest & Bouwman, 2006), and Lesschen (Lesschen *et al.*, 2011). The IPCC Tier 1 model also provides an alternative for estimating nitrogen leaching (IPCC) (Hergoualc'h *et al.*, 2019), which was used as the site-generic approach and compared with six site-dependent nitrogen leaching models: PooreNemecek (Poore & Nemecek, 2018), SQCB (Roches *et al.*, 2009), MITERRA (Velthof *et al.*, 2009), Brentrup (Brentrup *et al.*, 2000), NLeCCs_tot and NLeCCs_net (Johnsson *et al.*, 2016). The emissions models differ in overall approach and input parameters required (see Paper IV for an extensive description of the models).

The compared midpoint characterisation models for marine eutrophication compared in Paper IV and in this thesis were the site-generic models ReCiPe2008 (Struijs *et al.*, 2009) and ReCiPe2016 (van Zelm & Cosme, 2017), and the site-dependent models Cosme (Cosme *et al.*, 2017) and Henryson (Paper I). Eutrophication potential, which describes the total maximum eutrophying potential of all emissions, was also included, to enable analysis of the potential importance of elementary flows not covered by the other models.

4.2 Case studies

The case studies were designed to explore the effect of fertiliser management (Papers II, III and IV), site (Papers I, III and IV) and methodological choices (Papers I, II, III and IV), rather than evaluating the impact of a certain system. They are therefore not fully aligned with each other regarding product evaluated, system boundaries, functional unit, coverage of the cause-effect chain or impact categories, and are therefore not straightforward to compare. However, they describe similar systems and share common data sources and approaches, and are therefore described together in this section.

4.2.1 System

All case studies evaluated crop cultivation under mineral fertiliser, typical management practices and average yields for each region. An exception was the case in Paper III, where the nitrogen fertiliser amounts varied, and actual yield was measured in field trials.

The case study in Paper I included all of Sweden's agricultural land by assuming that one hectare (ha) in each sub-catchment was cropped with either spring barley or grass ley. In reality, the amount of agricultural land in each of the 8559 sub-catchments in Sweden varies between 0.1 and 72 000 ha (average 380 ha), so the case study did not represent the actual land use but was rather an example to illustrate the variation between sub-catchments.

The sites used in Papers III and IV are shown in *Figure 4*. The location of the site used in Paper II is only specified as Uppsala County, which is located around sites C4, C5 and East.

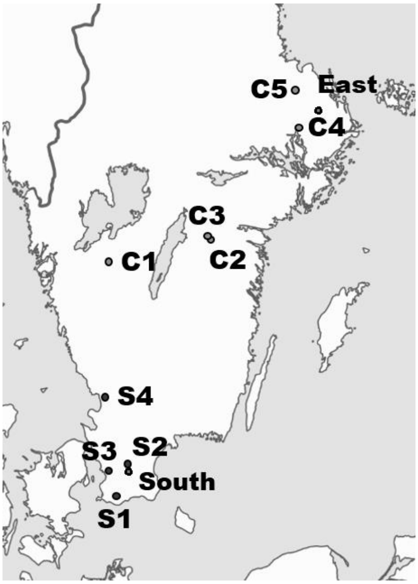


Figure 4. Map of southern and central Sweden showing the sites used in Paper III (S1-S4 and C1-C5) and Paper IV (South and East). The grey areas indicate major water bodies.

Spring barley was used as an example crop in Paper II, while winter wheat was used in Paper IV.

In Paper II, the initial yield was set as the average spring barley yield in Uppsala County in 2007-2016 (Statistics Sweden, 2016). Nitrogen fertiliser rates in Paper II was set according to management recommendations from the Swedish Board of Agriculture (Albertsson *et al.*, 2015). The yield response to SOM changes in Paper II was estimated based on measurements from the Ultuna continuous SOM field experiment in Uppsala (Kätterer *et al.*, 2011). The resulting yield response was 38.5% yield change per percent SOC change (Paper II). In the case study, we simulated a temporary fertiliser-induced 10% yield increase during year 1, and then ran the modelling framework for 60 years. In addition to the base scenario, the results for three other scenarios are presented in this thesis; amount of residues increase less than amount of grain yield, fertiliser rate decrease instead of increase, and lower yield response to SOM increase.

In Paper IV, the standard wheat yield in 2018 in Scania and Uppsala County was used for the South and East sites, respectively (Statistics Sweden, 2018b). The standard yield is the expected yield under normal weather conditions based on statistics from previous years. Mineral fertiliser rates in Paper IV were set according to statistics on average mineral fertiliser use in each region (Statistics Sweden, 2017b). The reason for choosing different data sources for yield and fertiliser rate in Papers II and IV was that the relationship between fertiliser rate and yield was more important for Paper II, while the focus of Paper IV was to depict average conditions. However, the yields and fertiliser rates from the different sources were approximately similar.

The assessment in Paper III was based on data from long-term field trials at different nitrogen fertiliser rates at four sites in southern Sweden (S1-S4) and five sites in central Sweden (C1-C5) (Carlgren & Mattsson, 2001). Impacts were assessed for crop rotations of spring barley-spring oilseed rape-winter wheat-sugar beet at sites S1-S4, and spring barley-oats-spring oilseed rape-winter wheat-oats-winter wheat at sites C1-C5. The average nitrogen fertiliser rates at the low, medium and high fertiliser levels were 50-100-150 kg N ha^{-1} at sites S1-S4, and 40-80-120 kg N ha^{-1} at sites C1-C5. Yield values were based on data from the field trials.

4.2.2 Goal and scope

System boundaries

The system boundaries were set at cradle-to-farm gate in Papers II-IV, which included the processes:

- Fertiliser and pesticide production
- Manufacturing, maintenance, fuel production and combustion emissions for agricultural machinery
- Soil emissions (including SOC changes)

Seed production was disregarded in Paper II and Paper IV, and was accounted for in Paper III by reducing the yield by the corresponding required seed rate. This approach is commonly used in LCAs (see *e.g*. Roer *et al.* (2012); Ahlgren *et al.* (2009), but is not ideal since in reality seeds are often produced on specialist farms and chemically treated after drying to maintain high quality and avoid the spread of pests (van Gastel *et al.*, 2002). This means that, in reality, production of grain for seeds probably causes higher impacts than production of other grain, but the influence of this process was assumed to be minor in relation to the other impacts.

Only the waterborne nitrogen and phosphorus emissions at field level were included in the case study in Paper I.

Functional unit

Several different functional units were used in this thesis, depending on the purpose of each study. In Papers I and IV, area used (expressed in ha) was employed as the functional unit to distinguish the influence of site and methodological choice on estimated emissions and impacts. However, a functional unit representing the amount of delivered product is usually a better choice for application in actual LCA studies, since it better reflects the actual function of the production system, *i.e*. delivering biomass for food, feed, energy, fibre *etc*. Harvested barley (expressed in kg grain) was therefore used as a functional unit in Paper II. Another functional unit was needed to express the impacts for several different crops at the same time in Paper III and in the summary of results in this thesis. For this purpose, cereal unit (CU) was used. It represents the animal feeding value of each crop in relation to the reference crop barley, and thereby accounts for one of the most important functions of crop production. Cereal unit was first introduced in an LCA context as a basis for co-product allocation between grain and straw or crops in rotation, but has also been used as a functional unit for crop rotations (Prechsl *et al.*, 2017; Brankatschk & Finkbeiner, 2015; Brankatschk & Finkbeiner, 2014).

Selection of impact categories

Climate impact and marine eutrophication were chosen as impact categories, since the contribution to those impact categories from crop cultivation is closely connected to nitrogen flows and yield level. Another reason for focusing on these impact categories is that agricultural activities are significant contributors to these environmental problems.

4.2.3 Life cycle inventory

Fertiliser and pesticide production

Emissions data for fertiliser production were taken from Brentrup *et al.* (2016), also presented as a full emissions inventory in the GaBi database (Fertilizers

Europe, 2018a; 2018b; 2018c; 2018d). The emissions inventory for production of pesticides was taken from the ecoinvent database (Nemecek, 2018).

Agricultural machinery

The field operations stubble cultivation, ploughing, harrowing, sowing, application of fertiliser and pesticides, and harvesting were included in Paper II-IV. Fuel consumption was estimated based on Lindgren *et al.* (2002) except for the ploughing in Paper III, which was differentiated by soil texture using an equation from Arvidsson and Keller (2011). Life cycle emissions for fuel production and use were taken from Gode *et al.* (2011). Machinery production and maintenance was included according to Tidåker *et al.* (2016) in Paper II, and estimated using ecoinvent data in Papers III and IV (see description in Paper III).

Soil emissions

Waterborne nitrogen and phosphorus emissions at field level were generally estimated using different versions of leaching data from the Swedish Environmental Emissions Data (SMED) (Johnsson *et al.*, 2016; Blombäck *et al.*, 2011; Johnsson *et al.*, 2008). In Paper III, nitrogen leaching was adjusted for nitrogen fertiliser rate and yield obtained according to the method presented by Aronsson and Torstensson (2004). In Paper IV, the two versions of the leaching data from Johnsson *et al.* (2016) (NLeCCs_tot and NLeCCs_net) were compared with other nitrogen leaching models (see section 4.1.3).

Airborne soil emissions of NH_3 and NO_x were estimated by the IPCC default value in Paper II (De Klein *et al.*, 2006), and by emissions models from EMEP/EEA (2016) in Papers III and IV.

Direct soil N_2O emissions were estimated by the IPCC Tier 1 model from 2006 (De Klein *et al.*, 2006) in Paper II, and by the model for total direct soil N_2O emissions from Rochette *et al.* (2018) in Paper III. The updated version of the IPCC Tier 1 model (Hergoualc'h *et al.*, 2019) and the model by Rochette *et al.* (2018) were compared with other models in Paper IV (see section 4.1.3). Indirect soil N_2O emissions were calculated with the emissions factors from IPCC Tier 1 2006 (De Klein *et al.*, 2006) in Papers II and III, and with those from IPCC Tier 1 2019 (Hergoualc'h *et al.*, 2019) in Paper IV.

Net emissions of CH_4 from soils were not included in any of the cases studied.

Soil organic carbon changes were estimated with the ICBM model in Paper II (see section 4.1.2). In Paper III, annual SOC changes were estimated as the slope of the linear regression of measured SOC content over the whole field trial period (approximately 50 years, with some variation between sites). Soil organic carbon changes were not included in Paper IV.

4.2.4 Life cycle impact assessment

The GWP_{100} metric (GWP with a 100-year time perspective) was used as the main characterisation model for climate impact in Papers II-IV. Characterisation factors excluding climate-carbon feedback (30 g CO_2eq g CH_4 and 265 g CO_2eq g N_2O) (Myhre *et al.* (2013) were used in Paper II. In Papers III and IV, characterisation factors including climate-carbon feedbacks (36 g CO_2eq g CH_4 and 298 g CO_2eq g N_2O) were used, since this gives a consistent accounting of the climate impact of different greenhouse gases (Myhre *et al.*, 2013).

The characterisation model developed in Paper I was used for assessing marine eutrophication in the case studies in Papers I and III. Eutrophication potential (CML2001; Guinée (2002)) was used as an indicator in the comparison of emissions models in Paper IV. These two models were also compared with three other characterisation models for marine eutrophication in Paper IV (see section 4.1.3).

In addition, GWP_{20} and AGTP (Myhre *et al.*, 2013) were applied for assessing climate impact in Paper II, and EDIP2003 (Hauschild & Potting, 2005) and ReCiPe2008 (Struijs *et al.*, 2009) were used for assessing marine eutrophication in Paper I. These characterisation models were included in the papers for comparison and the results are not presented in Chapter 5 of this thesis.

5 Results and discussion

5.1 Influence of site on environmental impact

Several of the emissions flows that dominate the climate and eutrophication impact of crop cultivation were strongly influenced by emission site (*Figure 5*, *Figure 7*, Paper III). This was highly relevant when evaluating the effects of fertiliser management, since impacts were dominated by emissions closely connected to fertiliser management. Site was also shown to affect which fertiliser rate that gave the lowest impact per unit produced (*Figure 6*, *Figure 8*, Paper III), which was a consequence of the site-specific dynamics between the magnitude of increased impact per unit area and the crop yield response to increased nitrogen rate (*Figure 5*, *Figure 7*, Paper III). Using models that can capture the differences in diffuse emissions between sites therefore improves the possibilities for relevant decision support for soil management and policy.

Figure 5. Climate impact per hectare (ha) of crop cultivation at different sites, fertiliser rates and methodological choices (see Chapter 4). Comparison of results between the different papers should be made with caution, since different methodological choices were made in

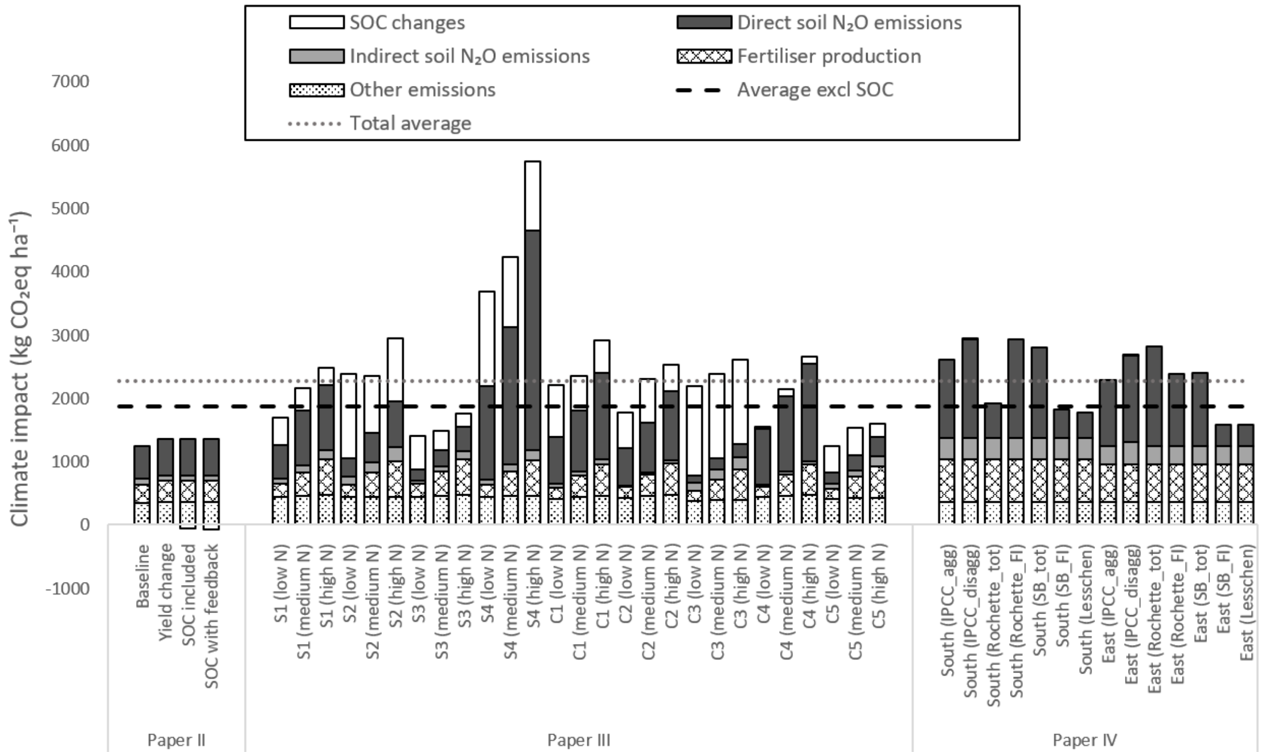


Figure 6. Climate impact per cereal unit (CU) of crop cultivation and yield at different sites, fertiliser rates and methodological choices (see Chapter 4). Comparison of results between the different papers should be made with caution, since different methodological choices were applied in each case.

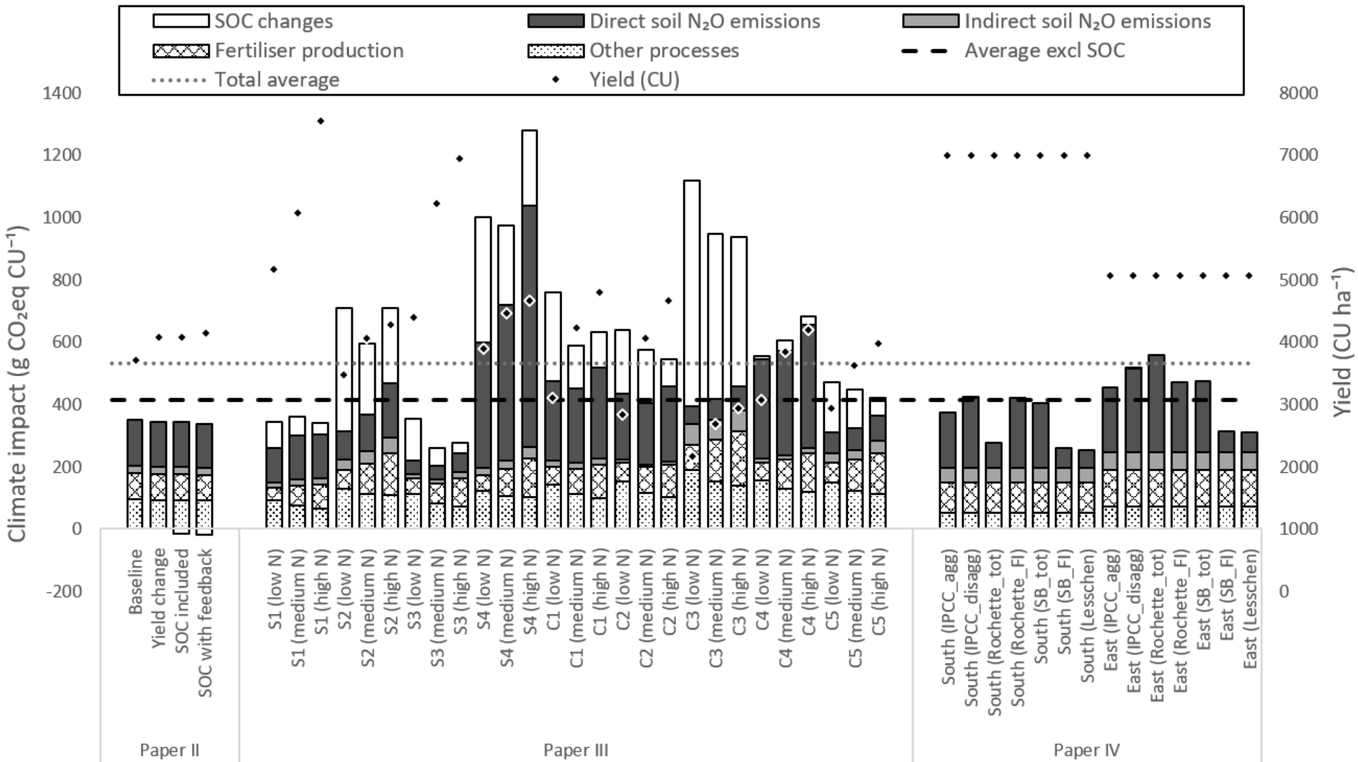


Figure 7. Marine eutrophication (Paper III and Paper IV – Characterisation models) or eutrophication potential (Paper IV – N leaching models) per ha of crop cultivation at different sites, fertiliser rates and methodological choices (see Chapter 4). Comparison of results between the different papers should be made with caution, since different methodological choices were applied in each case.

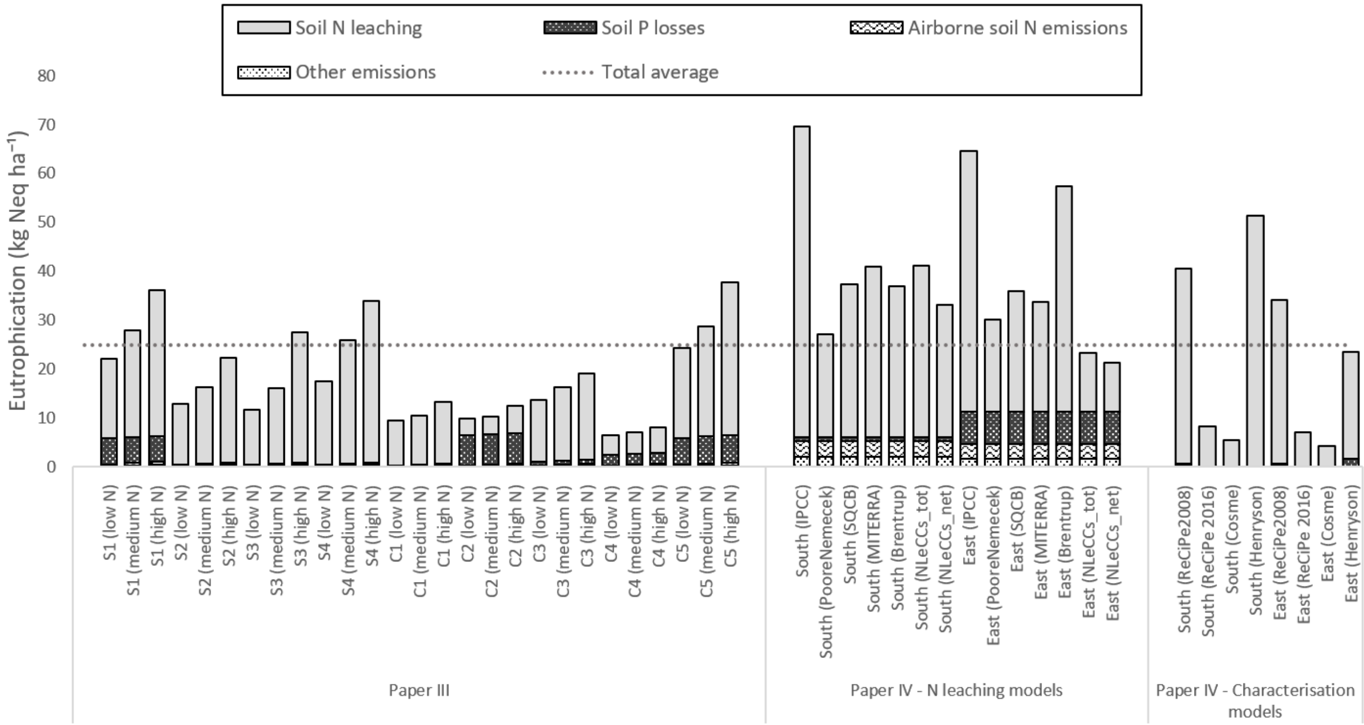
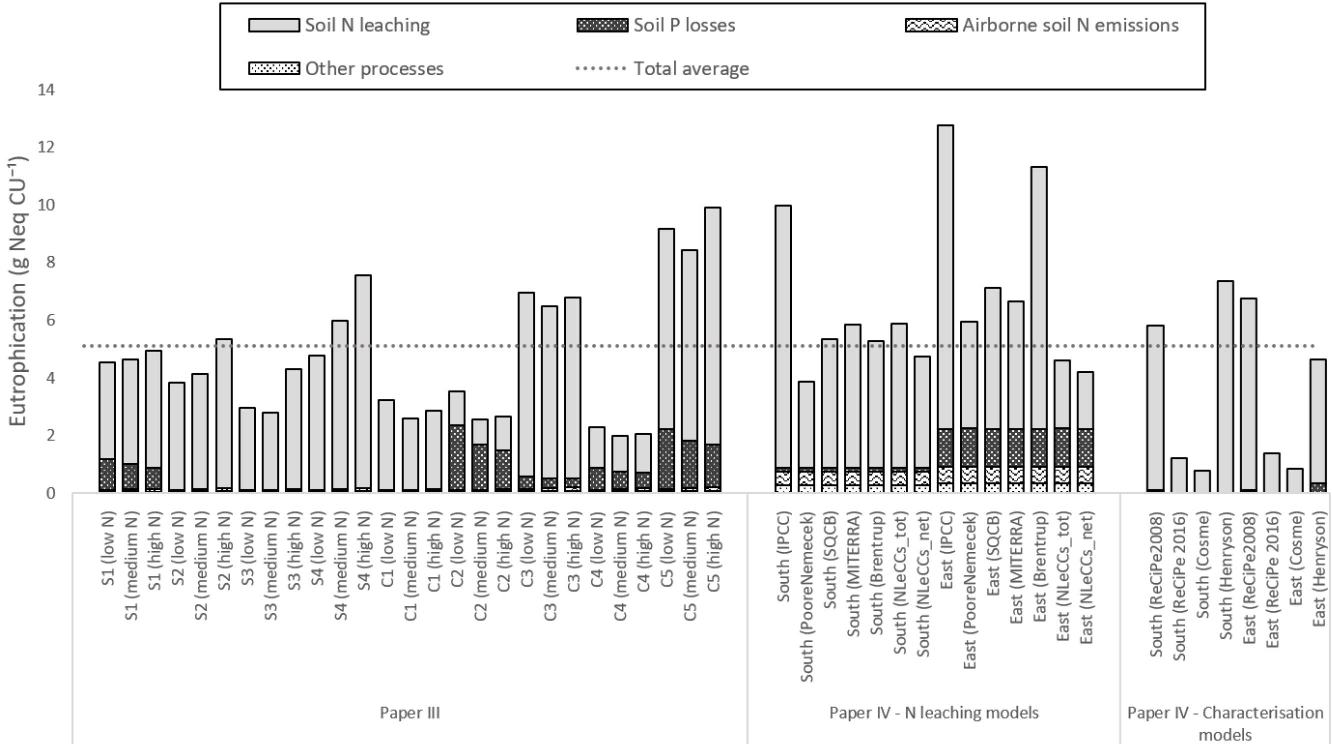


Figure 8. Marine eutrophication (Paper III and Paper IV – Characterisation models) or eutrophication potential (Paper IV – N leaching models) per cereal unit (CU) of crop cultivation at different sites, fertiliser rates and methodological choices (see Chapter 4). Comparison of results between the different papers should be made with caution, since different methodological choices were applied in each case.



5.1.1 Spatial variations in diffuse emissions at field level

Soil N_2O emissions

Soil N_2O was one of the processes with both a high contribution to climate impact (11-63% of total impact) and large variation between sites and nitrogen fertilisation rates (230-3600 kg CO_{2eq} ha^{-1}, 58-810 g CO_{2eq} CU^{-1} in Paper III) (*Figure 5, Figure 6*). Most of the impact from soil N_2O emissions came from the direct component (on average 81%), but this proportion varied between sites and fertiliser rates (47-96%). Generally, sites with higher direct soil N_2O emissions had lower indirect soil N_2O emissions, since clayey soils promote generation of N_2O emissions, but decrease the risk of nitrogen leaching (Rochette *et al.*, 2018; Kyllmar *et al.*, 2006).

Nitrous oxide emissions from agricultural fields are regarded as inherently difficult to quantify due to high temporal and spatial variability and a large number of influencing parameters, such as climate, soil type and soil organic matter content (Rochette *et al.*, 2018; Stehfest & Bouwman, 2006). The comparison in Paper IV of different models for quantifying soil N_2O emissions illustrated this difficulty. The estimated climate impact of direct soil N_2O emissions during wheat cultivation varied from 320 kg CO_{2eq} ha^{-1} to 1600 kg CO_{2eq} ha^{-1} (58-280 g CO_{2eq} kg^{-1} wheat at 14% moisture content) at one of the sites (East) in Paper IV, depending solely on which model was used (*Figure 5*). The commonly used IPCC Tier 1 model (Hergoualc'h *et al.*, 2019; De Klein *et al.*, 2006) estimated values close to the average of all models, which suggests that it provides a reasonable approximation of the direct N_2O emissions at larger scales. However, with the aggregated IPCC Tier 1 model the difference in impact of direct N_2O emissions between the sites was approximately 200 kg CO_{2eq} ha^{-1}, which can be attributed to the difference in fertiliser rate and amount of crop residues, while three of the site-dependent models predicted larger differences in impacts between the two sites (280-1020 kg CO_{2eq} ha^{-1}). The model that gave the largest difference between the sites was Rochette_tot (Rochette *et al.*, 2018), which was used in Paper III. In addition, and in contrast to the other six models, this model estimated higher emissions per ha at the East site (*Figure 5*). This could mean either that this model is better at capturing local differences, or that it overestimated the differences between these two sites. If the latter is true, it could mean that the difference between sites in Paper III appears larger than it is in reality. The lack of measurements and models validated for Swedish conditions limits the possibilities to judge which model gives the more accurate outcomes.

Nitrogen leaching

Nitrogen leaching dominated the marine eutrophication impact at most of the fertiliser rates, sites and model choices studied (*Figure 7*). The estimated nitrogen leaching also differed by up to 47 kg N ha^{-1} between the sites in Paper III (from 10 to 57 kg N ha^{-1} at the high nitrogen level) (*Figure 7*), and by up to 23 kg N ha^{-1} between the two sites in Paper IV (from 12 to 35 kg N ha^{-1} estimated by model NLeCCS_tot) (*Figure 7*). However, some of the models tested in Paper IV gave a much smaller difference in nitrogen leaching between the sites. The possibility to detect differences between sites was thus dependent on model choice.

In general, nitrogen leaching is promoted by sandy soils and higher precipitation. Since direct soil N_2O emissions are higher in clayey soils (Rochette *et al.*, 2018), site affected climate impact and marine eutrophication differently (*Figure 9*).

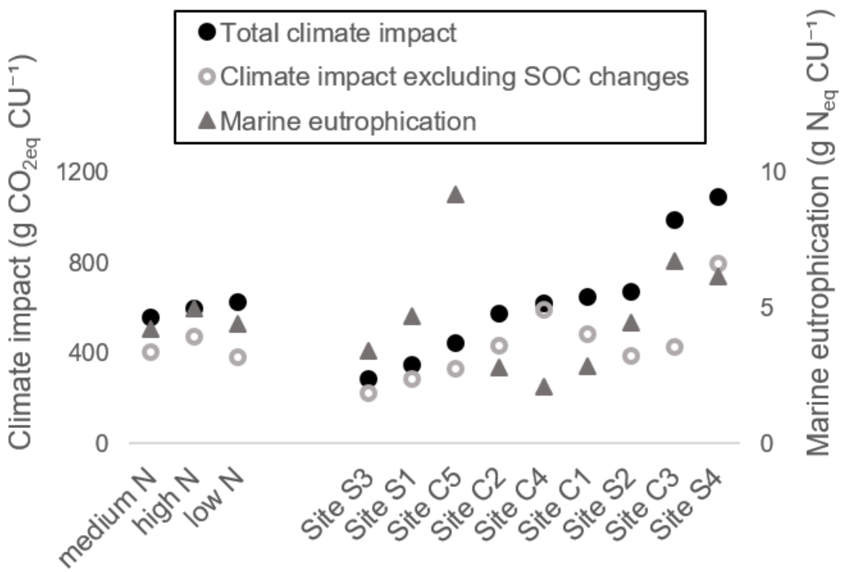


Figure 9. Mean total climate impact (dots), climate impact excluding soil organic carbon (SOC) changes (circles), and marine eutrophication (triangles) per cereal unit (CU), for the nitrogen fertiliser levels and sites in Paper III. The fertiliser rates and sites are ordered in ascending order according to total climate impact.

Soil organic carbon changes

The SOC stock decreased at all sites and nitrogen fertiliser levels studied in Paper III, with the average SOC loss at each site corresponding to between 25 and 560 g CO_2 CU^{-1}. The decrease differed more between the sites than between the fertiliser rates (Paper III), and also had a large influence on the differences in total climate impact between sites (*Figure 5*, *Figure 6*). However, cereal cultivation can also increase SOC stocks if the cultivation occurs on soils that have previously received low carbon inputs (Gan *et al.*, 2012a; Campbell *et al.*, 2005). The absolute SOC loss at a particular site under specific management may therefore be more dependent on soil history than the current soil management, and SOC loss might occur from some soils even if they are not cultivated. Comparisons between different sites intended to support decision-making should therefore also account for the SOC changes under a reference land use (Hammar *et al.*, 2016). It is thus more relevant to consider the SOC changes when comparing the climate impact at different nitrogen fertiliser levels than when comparing the sites (see also section 5.3.3 for a more extensive discussion on accounting for SOC change in LCA).

5.1.2 Spatial variation in marine eutrophication impact

Geographical location proved to have a large influence on marine eutrophication values. This was shown in Paper I, where the marine eutrophication characterisation factors for both nitrogen and phosphorus differed widely between sites, from 0.056 to 0.99 kg Neq kg^{-1} N and from 0 to 7.23 kg Neq kg^{-1} P (*Figure 10*). When these characterisation factors were combined with spatially differentiated data on estimated losses of nitrogen and phosphorus from agricultural fields in Sweden, divided into 8559 sub-catchments, impacts of these losses varied by an order of magnitude for grass ley (0.31-32 kg N_{eq} ha^{-1}), and even more for spring barley (1.2-58 kg N_{eq} ha^{-1}) (Paper I).

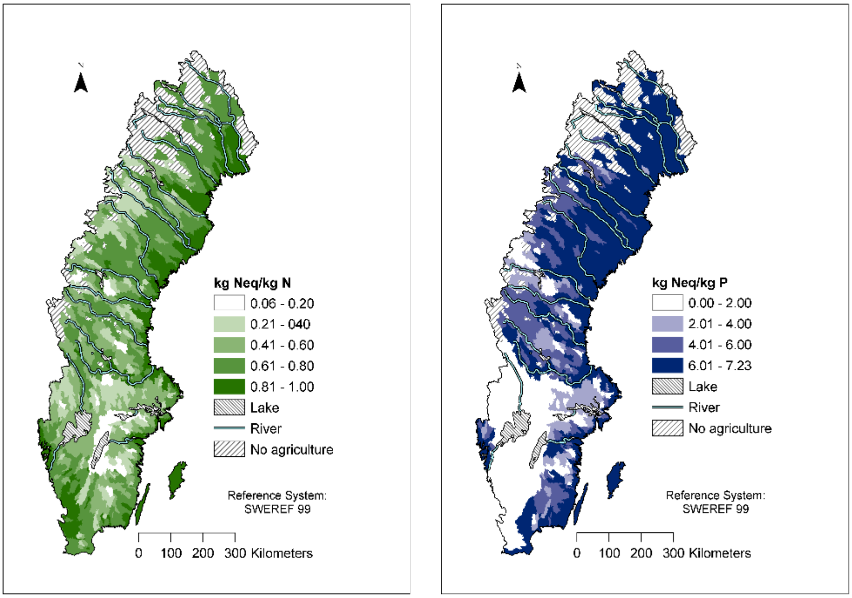


Figure 10. Marine eutrophication characterisation factors for nitrogen (N) and phosphorus (P) emissions to soil in Sweden derived in Paper I.

5.2 Influence of nitrogen fertiliser intensity on environmental impact

Increasing nitrogen fertiliser rate increased climate and marine eutrophication impacts per unit area at all sites in Paper III (*Figure 5*, *Figure 7*). The impacts per unit produced either increased or decreased with fertiliser rate, depending on site (*Figure 6*, *Figure 8*). The mean climate impact and marine eutrophication impact were lowest at the medium nitrogen level (*Figure 9*). However, the differences in mean impacts between nitrogen levels were not statistically significant for climate impact, and were only significant between the medium and high nitrogen rate for marine eutrophication (Paper III). This was due to the variation in impact magnitude and ranking of the nitrogen levels between the sites, and indicates that site conditions have to be considered when choosing an appropriate nitrogen rate for minimising environmental impact. However, nitrogen rate also affected the two impact categories differently within each site, with only one of the nine sites (C1) having the same ranking of nitrogen rates in terms of their contribution to the two impact categories. This was due to the different responses of emissions contributing to these two impact categories, in terms of increased impact per ha at increasing nitrogen rates.

For climate impact, the response to higher nitrogen rates was similar at all sites for the emissions from inputs and field operations, while the response for soil N_2O emissions and SOC change varied between the sites. The impact from soil N_2O emissions per ha increased with nitrogen rate, while the impact from SOC change per ha generally decreased with increased nitrogen rate (although the estimation of SOC change was very uncertain, see Paper III).

For marine eutrophication, nitrogen leaching dominated the impacts, but phosphorus losses at field level were also important at some sites. While the impact of nitrogen leaching per cereal unit generally increased with higher nitrogen rates, the impact of phosphorus losses decreased, since phosphorus losses per ha were assumed to be constant.

Impacts of direct soil N_2O emissions, SOC changes, nitrogen leaching and phosphorus losses were affected differently by site conditions (see section 5.1). Therefore the relationship between the climate impact and marine eutrophication impact at different nitrogen fertiliser levels was not the same at all sites studied in Paper III (*Figure 6*, *Figure 8*). The dominance of these site- and nitrogen rate-dependent emissions in both impact categories prompted further investigations of the robustness of models available for LCA practitioners to estimate soil N_2O emissions and nitrogen leaching (Paper IV).

5.2.1 Effects of fertiliser intensity on soil organic matter dynamics

The mean SOC loss was smaller at higher nitrogen fertiliser levels in Paper III, both per unit area and per unit produced crop. The mean climate impact of SOC loss was 780, 640 and 570 kg CO_2eq ha^{-1} (*Figure 5*) and 230, 150 and 120 g CO_2eq CU^{-1} (*Figure 6*) for the three nitrogen levels tested. However, all sites did not exhibit this pattern and the uncertainties were larger than the differences between nitrogen levels, so it is difficult to draw general conclusions based on these results. See also section 5.3.3 about uncertainties in SOC accounting.

A different perspective was investigated in Paper II. There, the influence of a crop management change rather than constant management was modelled, and the SOC changes were not directly measured but simulated based on a unique dataset from a long-term agricultural field experiment. The fertiliser-induced yield increase and subsequent increase in soil organic matter proved to have a small but non-negligible effect on the estimated climate impact, which was even larger if the soil fertility increase was included (*Figure 11*). In the base scenario, a higher fertiliser rate decreased the climate impact even if SOC accumulation was not accounted for, since the yield increased more (10%) than the estimated climate impact per ha (8%). However, accounting for SOC accumulation decreased the estimated climate impact even more, and also accounting for the subsequent effect of SOM on crop yield gave additional reductions of the impact. *Figure 11* displays the results from running the modelling framework from Paper II with different parameter settings for residue response to fertiliser change, fertiliser increase or decrease and yield response to SOM change. The quantitative effect of accounting for the yield response to SOM change differed depending on parameter choice, but the reinforcing effect persisted (*Figure 11*).

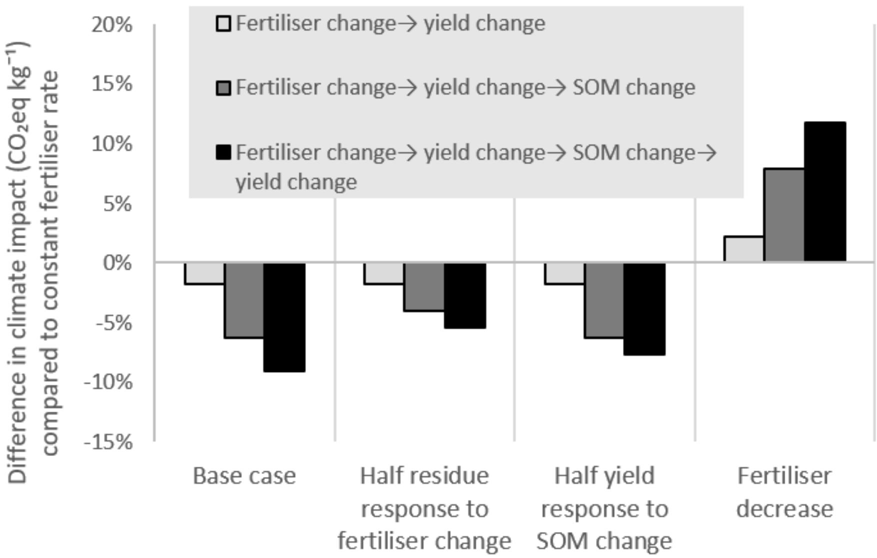


Figure 11. Difference in assessed climate impact (g CO_2eq kg^{-1}) between a constant fertiliser rate (base case), and different cases of fertiliser-induced yield change. Grey shades indicate modelling approach, *i.e*. how the soil organic matter (SOM)-yield dynamics were modelled.

5.3 Influence of methodological choices on LCA outcomes

Overall, the different comparisons between methods performed in this thesis showed that some methodological choices can be crucial for the outcome of an LCA. These choices range from the choice of system boundary to the choice of emissions models and level of spatial differentiation in impact assessment modelling.

5.3.1 Emissions model selection

There are many examples of LCA studies that compare crop cultivation at different fertiliser intensities, where crude models are used for estimating soil nitrogen emissions (*e.g.* Noorhosseini & Damalas, 2018; Reckling *et al.*, 2016; Krohn & Fripp, 2012). If soil N_2O emissions and nitrogen leaching are modelled using fixed and site-generic emissions factors related to fertiliser rates, such as the widely used IPCC Tier 1 model (Hergoualc'h *et al.*, 2019), then fertiliser use per unit mass of yield becomes crucial for the outcomes in these impact categories. Since yield per unit mass of nitrogen fertiliser tends to decrease with increasing nitrogen fertiliser rate, it is often concluded that minimising fertiliser rate will give the lowest impact per unit produced crop (Pahlmann *et al.*, 2013). However, the results in this thesis shows that site (Paper III) and the models chosen to assess diffuse nitrogen emissions (Paper IV) affect the comparison of impacts at different fertiliser rates.

In practice, the model choices in LCA are affected by the availability of relevant models, the data requirement and the effort needed to apply them. The synthesis and comparison of different site-dependent medium-effort models for direct N_2O emissions and nitrogen leaching at field level in Paper IV revealed that different models produced widely different results, in terms of emissions amount and also in terms of which site gave larger emissions (*Figure 5*, *Figure 7*). These emissions flows typically comprise a major part of the climate and marine eutrophication impact, respectively, of crop cultivation. Therefore, choice of model can alter the conclusions of the LCA (Paper IV) (*Figure 5*, *Figure 7*).

Kasimir Klemedtsson and Smith (2011) compared measured N_2O emissions at two Swedish sites and two mineral fertiliser rates at each site to emissions estimated using the aggregated IPCC model, the StehfestBouwman model (Stehfest & Bouwman, 2006) and a third model by Freibauer and Kaltschmitt (2003) that was not included in the comparison in Paper IV. None of the models had an uncertainty range that encompassed the emissions in all four treatments studied (Kasimir Klemedtsson & Smith, 2011). Those authors concluded that SOC content, rather than fertiliser rate, was the most important factor at one of the sites, and that soil nitrogen content was the most important factor at the other site. Another study based on measurements across European sites found that nitrogen fertiliser addition was the most important factor, but only explained 15% of the variance in the linear regression analysis (Rees *et al.*, 2013). The overall outcome of these studies is supported by the findings of the comparison in Paper IV, *i.e.* that using nitrogen fertiliser rate as the only variable when estimating soil N_2O emissions can give misleading results.

The IPCC guidelines for national greenhouse gas emissions reporting were updated in May 2019 (Hergoualc'h *et al.*, 2019). The previous version of the guidelines did not provide any site-dependent emissions factors for soil N_2O, although the option to use country-specific emissions factors (Tier 2) or more advanced models (Tier 3) was allowed (De Klein *et al.*, 2006). The 2019 guidelines provide the option to use different emissions factors depending on whether the cultivation is located in a wet or dry climate (implemented as IPCC_disagg in Paper IV and in this thesis). For Swedish conditions, the spatially disaggregated emissions factors are 1.6% of mineral fertiliser nitrogen and 0.6% of nitrogen in crop residues and mineralised SOM (Hergoualc'h *et al.*, 2019). For the case study in Paper IV, this meant that 1.3% of all available nitrogen was estimated to be emitted as direct N_2O emission. This can be compared with the aggregated site-generic emissions factor of 1% for all available nitrogen, also provided in the 2006 version of the guidelines (Hergoualc'h *et al.*, 2019; De Klein *et al.*, 2006). Disaggregation reduces the uncertainty (Hergoualc'h *et al.*, 2019) and probably produces more realistic estimations of national N_2O emissions. However, it does not represent a substantial improvement for field-level assessments, since the spatial resolution is still very low compared with the actual variation (Fitton *et al.*, 2017; Stehfest & Bouwman, 2006).

Considering the varying outcomes of the N_2O emissions models in Paper IV (*Figure 5*), it would have been useful to compare the model outcomes against measured emissions. There are some published N_2O measurements from crop cultivation on mineral soils in Sweden (Kasimir Klemedtsson & Smith, 2011; Nylinder *et al.*, 2011), but these measurements cover few sites and management practices. Without measured values against which to compare the modelled results, it is not possible to decide with certainty which of the models compared performed best.

Strong site-dependence of nitrogen leaching is widely acknowledged when nitrogen leaching is assessed in agronomic or water pollution contexts, *e.g.* in national reporting to international pollution prevention agencies (Brandt *et al.*, 2009). In contrast to soil N_2O emissions, there is much measured and modelled data on nitrogen leaching in Sweden (*e.g.* Johnsson *et al.*, 2016; Delin & Stenberg, 2014; Bergström *et al.*, 2008; Kyllmar *et al.*, 2005). Thanks to the regular Pollution Load Compilations (PLC) submitted to the Baltic Marine Environment Protection Commission (HELCOM), leaching models and modelling results are also continuously updated (Sonesten *et al.*, 2018; Brandt *et al.*, 2009). The nitrogen leaching data in the PLCs are produced by running the process model NLeCCS with different set-ups to derive nitrogen leaching coefficients per crop and soil type for each region. The resulting leaching coefficients are then readily available, *e.g.* for LCA practitioners to estimate site-dependent nitrogen leaching during typical soil management without having to run the process model themselves. Different versions of the data on nitrogen leaching from the PLCs were used in Papers I, III and IV, and have been used in other LCAs of Swedish crop cultivation (Henriksson *et al.*, 2012; Ahlgren *et al.*, 2009). The leaching coefficients derived by NLeCCS gave larger differences between the sites than the other models in Paper IV (*Figure 7*). NLeCCS is the only one of these models specifically verified for Swedish conditions and it is therefore likely that the NLeCCS results are more realistic. The other models probably underestimate the influence of site characteristics on the magnitude of nitrogen leaching. Comparing the mean impacts at the sites in Paper III calculated with IPCC Tier 1 and NLeCCS illustrates the considerable effect of using a site-dependent nitrogen leaching model (*Figure 12*).

A drawback with the NLeCCS leaching coefficients is that there is no straightforward model to adapt them according to nitrogen fertiliser rate. In Paper III, this was handled by applying region- and soil type-dependent correction factors based on the difference between the nitrogen fertiliser rate applied and the recommended fertiliser rate in relation to yield obtained (see Supplementary Material to Paper III). This approach was developed by Aronsson and Torstensson (2004) to demonstrate the effect of different management practices to farmers. Although there is no obvious reason why this would be an inappropriate method to derive nitrogen leaching for cropping system LCAs, it has not been used previously in LCAs.

5.3.2 Impact assessment model selection

Marine eutrophication

The selection of characterisation models for impact assessment should be consistent with the goal and scope of the study (ISO, 2006c). This indicates that a site-generic characterisation model for site-dependent impacts such as eutrophication is not suitable for quantifying the impact of production at a certain site or comparing the impacts of products produced at different sites. The model presented in Paper I was compared with several different characterisation methods (Paper I and Paper IV). Comparing the results for eutrophication potential and site-dependent marine eutrophication impacts (displayed on the right-hand side in *Figure 7*) showed that choice of characterisation model can have a large effect on the conclusions drawn from an LCA. Re-calculation of the mean marine eutrophication impacts at each site in Paper III using the site-generic characterisation model ReCiPe2008 illustrates this further (*Figure 12*). Comparing the results for the two approaches displayed in the middle and on the right-hand side in *Figure 12*, it can be seen that the results differ substantially for most of the sites. For some, *e.g.* sites S2 and C3, lower site-dependent characterisation factors for nitrogen decreased the impact compared with that determined using the site-generic model. For other sites, for example sites S1, C2 and C5, the inclusion of phosphorus flows in the site-dependent model instead increased the estimated impact compared with that produced in the site-generic impact assessment. Site-dependent modelling of both emissions and impacts of those emissions are thus needed to produce site-dependent results.

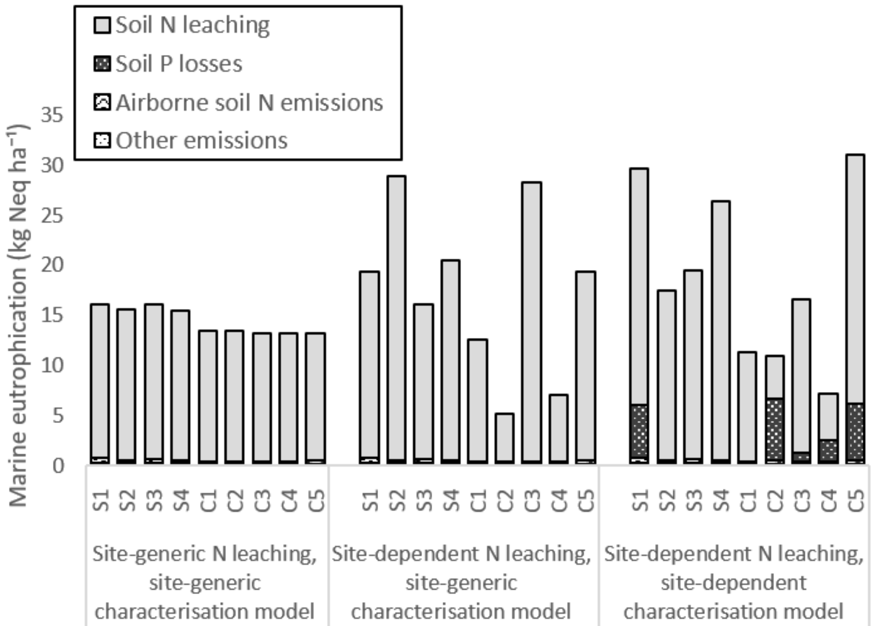


Figure 12. Mean marine eutrophication per ha at sites S1-C5 in Paper III, recalculated with either a site-generic (IPCC) or site-dependent (NLeCCs with correction for nitrogen (N) fertiliser rate), nitrogen (N) leaching model, and a site-generic (ReCiPe2008) or site-dependent (from Paper I) characterisation model for impact assessment. The three combined approaches shown represent different levels of spatial differentiation: (Left) a site-generic approach, (centre) a site-dependent inventory but site-generic impact assessment, and (right) a site-dependent inventory and impact assessment.

Around the time when Paper I was published, a spatially differentiated marine eutrophication characterisation model with global coverage was developed (Cosme & Hauschild, 2017). A site-generic version of this model was later also implemented in the ReCiPe2016 characterisation method (van Zelm & Cosme, 2017). Global coverage is a great benefit compared with the national perspective applied in Paper I, since supply chains are often multi-national. The characterisation model by Cosme and Hauschild (2017) also provides exposure, effect and damage factors, which enables inclusion of a larger part of the marine eutrophication cause-effect chain. However, this global model has lower spatial resolution, which means that the model presented in Paper I can detect differences between sites located closer to each other (Paper IV). Comparing these two models at the midpoint level, they also gave widely different results in terms of impact magnitude, spatial variability and specific characterisation factors at the two sites (*Figure 7*, Paper IV). The model by Cosme and Hauschild (2017) had characterisation factors of 0.085 and 0.078 kg N_{eq}/kg N at the South and East site, respectively, while the corresponding characterisation factors using the model in Paper I were 0.81 and 0.41 kg N_{eq}/kg N (Paper IV). There is no obvious explanation for this large difference. The data used to calculate retention, *i.e.* the fraction of emitted nutrients that will not reach the recipient, in the method presented in Paper I is used for the Swedish reporting to HELCOM and it seems unlikely that the calculated retention would differ from reality by an order of magnitude.

In terms of substance coverage, neither the model from Paper I nor the model by Cosme and Hauschild (2017) includes airborne compounds, which can give misleading conclusions (Bach & Finkbeiner, 2017). Airborne emissions only accounted for a minor part of the eutrophication potential in the case studies in Paper IV (*Figure 7*), but it could be essential to include them for other production systems. The characterisation model from Paper I is the only one of the characterisation models for marine eutrophication tested in Paper IV that includes phosphorus. The exclusion of phosphorus from the other models is most likely a deliberate choice, since nitrogen is the limiting nutrient in the majority of marine ecosystems globally. However, at the local scale, where phosphorus limitation can occur, models that do not include phosphorus can generate misleading outcomes. While nitrogen contributed to a larger part of the impact than phosphorus in most of the case study results in Papers I, III and IV, there were some exceptions. For example, waterborne phosphorus losses contributed to a larger impact than waterborne nitrogen losses in 15% of the sub-catchments simulated in Paper I and at one of the sites in Paper III (*Figure 12*). This means that more than half of the potential contribution to marine eutrophication would not be accounted for in these cases if the chosen characterisation model did not cover phosphorus. Considering that agriculture contributes almost half of the phosphorus loads to the Baltic Sea (Sonesten *et al.*, 2018), basing decisions on assessments that disregard this emission flow would not be ideal.

Spatial differentiation in impact assessment is generally considered an important step towards more representative LCA results (Notarnicola *et al.*, 2017; Azevedo *et al.*, 2013; Reap *et al.*, 2008), but is rarely applied in reality due to lack of appropriate impact assessment models and/or too high effort required to apply these models. The characterisation model presented in Paper I only covers waterborne emissions to soil in Sweden, and requires some effort to apply due to the high spatial resolution. For those reasons, widespread use of the model is unlikely. However, its use in the case study in Paper I, and its application in Papers III and IV, demonstrated that it can provide useful information in specific cases. For increased use of spatially differentiated impact assessment, there is a need for characterisation models with global coverage, relevant spatial resolution with options to use aggregated characterisation factors, inclusion of all relevant substances and integration with commonly used databases in accessible software tools (Bach & Finkbeiner, 2017; Njakou Djomo *et al.*, 2017; Mutel & Hellweg, 2009). Until characterisation models that fulfil these requirements become available, application of models such as that developed in Paper I can provide knowledge on appropriate spatial resolution and the importance of different substances.

Eutrophication potential and nitrogen footprint

There are broader indicators than those applied for marine eutrophication which can be useful when analysing the environmental impact of nitrogen fertiliser use.

The characterisation model for eutrophication potential included in the CML2001 impact assessment method (Guinée, 2002) was used in Paper I and Paper IV. This model translates the nitrogen and phosphorus emissions flows into a common unit, but does not account for the fate or impact of these flows. In Paper I, it was used to analyse whether the new characterisation factors were reasonable. In Paper IV, it was included to analyse the potential importance of emissions flows not covered by the marine eutrophication characterisation models. It was also used for the comparison of nitrogen leaching models, since the primary aim in that instance was to compare the estimated emissions rather than the impact of those emissions.

A similar indicator is the nitrogen footprint, which represents the total reactive nitrogen emissions to the biosphere (agricultural soils are considered part of the biosphere or part of the technosphere, depending on the chosen version of the indicator) throughout the life cycle (Einarsson & Cederberg, 2019; Leach *et al.*, 2012). The eutrophication potential and the nitrogen footprint can be useful for communication to wide audiences, identifying potential hotspots in the life cycle and ensuring that no flows are unintentionally overlooked in the impact assessment (Einarsson & Cederberg, 2019; Willett *et al.*, 2019; Leach *et al.*, 2012). However, they are complements rather than alternatives to marine eutrophication impact assessment, since they do not consider the mechanisms that cause environmental damage and therefore are poor predictors of the actual impacts.

5.3.3 Accounting for impacts on soil organic matter and soil organic carbon dynamics

The results from Paper II showed that accounting for the positive feedback mechanism between SOC and crop yield when assessing fertiliser management change could be almost as important for the calculated climate impact as accounting for the direct effect of SOC change (*Figure 11*). The approach used in Paper II requires the soil to be considered as both part of the technosphere delivering the product and part of the biosphere receiving the impact. This makes sense in that context, but violates the traditional system boundary concept defined by the ISO standards on LCA (ISO, 2006c).

The importance of preserving or increasing soil organic matter content to maintain crop yields is widely recognised within the agronomic field, but the fertility effect of SOM is usually ignored in LCAs, even if direct SOC changes are accounted for (Njakou Djomo *et al.*, 2015; Queiros *et al.*, 2015; Malca *et al.*, 2014). The fertility effect of SOM has been considered in some LCA contexts, *e.g.* by assuming that a sufficient amount of crop residues is left in the field to maintain soil fertility (Spatari *et al.*, 2010), or that the removal of crop residues is compensated for by increasing fertiliser input (Cherubini & Ulgiati, 2010). Soil organic matter has also been included in specific impact assessment methods for soil quality (Oberholzer *et al.*, 2012; Brandão *et al.*, 2011; Milà i Canals *et al.*, 2007b). Compared with these approaches, the modelling framework presented in Paper II offers more flexibility in terms of the management changes that can be modelled, and it also has the benefit of including the fertility effects on the system performance instead of a separate indicator. The modelling framework may be too complex and data-intensive to be feasible for application in most LCAs, but since the results in this thesis show that the effect on climate impact is non-negligible, it may be necessary to account for it in some way to avoid burden shifts. This would be especially relevant when evaluating large-scale or long-term implementation of *e.g.* intensification efforts or increased biomass outtake (in particular residues), both of which have been widely advocated (Garnett *et al.*, 2013; European Parliament, 2009).

While there is general consensus on the importance of SOM for maintaining crop yields, there is an ongoing debate about whether this can be attributed to the effect of SOC or the nutrients provided by SOM mineralisation (Oelofse *et al.*, 2015). The effect of SOM on yield in Paper II was derived by comparing the yields between two treatments, where cereal straw was added in one but not the other. Regression analysis revealed a statistically significant linear relationship between the yield difference and the SOC difference between these treatments. Although SOC was used as the indicator for soil fertility in Paper II, this does not necessarily mean that it is the SOC itself that improves productivity, but rather that an increase in crop residue input from a yield increase would have a similar effect as adding additional straw (as done in the field experiments). Here, SOC was used as the common 'unit' to relate the SOM increase in the simulations to the SOM increase in the field experiments. It is also important to note that the effect of SOM on yield is highly dependent on site characteristics and soil management (Oelofse *et al.*, 2015; Blanco-Canqui & Lal, 2009). The estimated yield response to SOC used in Paper II should therefore not be used as a default factor in other contexts.

In Paper III, a simple method was used to account for the CO_2 emissions arising from SOC changes, by attributing the average annual SOC loss during the whole course of the long-term field trials to the emissions inventory each year. The SOC measurements were highly variable, resulting in uncertain annual SOC change estimates (see Paper III). Modelling and including soil organic carbon dynamics in agricultural LCAs is also inherently complicated for several other reasons (Brandão *et al.*, 2013). Soil organic carbon dynamics are strongly dependent on soil history, as previous soil management can affect the SOC level for many decades after the management changes (Kätterer *et al.*, 2008). In addition, the long-term climate benefit of carbon sequestration depends on the duration of carbon storage (Brandão *et al.*, 2013). It is therefore not a fully justifiable decision to attribute annual carbon exchange between the soil and atmosphere to the crops produced in that year. For these reasons, the analysis in Paper III was carried out for both total climate impact and the climate impact when SOC changes were excluded. This affected the total climate impact values and the absolute differences between treatments, but generally did not alter the conclusions regarding the influence of site and nitrogen level on the impacts.

Despite the methodological difficulties, it is important to note that the uncertainty in SOC accounting does not justify ignoring SOC dynamics in LCAs, since they make a large contribution to total climate impacts.

5.4 Life cycle assessment as a decision support tool

5.4.1 Benefits of improving precision of life cycle assessment results in different contexts

The influence of methodological choices on LCA outcomes has been a topic of debate within the LCA community for a long time, and the lack of strict rules for method choices is still a common criticism regarding the credibility of LCA results as a whole (Curran, 2014; European Commission, 2013; Baumann & Rydberg, 1994; Tillman *et al.*, 1994; Guinée *et al.*, 1993). Several ambitious initiatives, such as the European Commission's suggested Product Environmental Footprint (PEF) (European Commission, 2013), the standard for Environmental Product Declarations (EPD) (ISO, 2006a) and the European Union's Renewable Energy Directive (RED) (European Parliament, 2009) aim to harmonise LCA methodology for specific applications so that results can be compared across studies in a fairer way. This is clearly an advantage when the results are used in *e.g*. marketing, where the recipient has limited possibilities to critically assess the underlying methodology used to achieve the final quantitative result presented. Applying LCA approaches in legislation also requires clear guidelines to ensure fair judgement of all actors. However, detailed rigid rules on appropriate choices for an LCA also limits the possibility to adjust the methodological approach to answer specific questions. They might also limit incorporation of new scientific insights on environmental mechanisms or new modelling tools. The results in this thesis show that more detailed modelling of certain processes can provide new insights into the mechanisms governing the climate and marine eutrophication impacts of crop cultivation. These insights can be used to improve the accuracy of assessed impacts, and improve the usefulness of LCA as a tool towards decreasing the environmental impacts of agricultural production.

The application of LCA has different objectives depending on who requests the information and what the results will be used for. However, if LCA is to be a useful tool in achieving a more sustainable food system, the overall goal should be to reduce the total environmental damage caused by production and consumption of food, while avoiding sub-optimisation and burden shifting between geographical regions, products and impact categories. With that goal in mind, this section explores how different stakeholders can use the information from more detailed LCA modelling to help achieve an overall reduction in the environmental impact of crop cultivation. The discussion focuses on environmental aspects, but other factors, such as agronomic, social, legal and economic aspects, are of course also important to consider when evaluating effects of crop management change.

Farm level

The results in this thesis indicate that improving modelling precision, *e.g*. by using site-dependent models (*Figure 5-8, Figure 12*, Paper IV) or several impact categories (*Figure 9*, Paper III) and the interaction between crop yield and SOM (*Figure 11*, Paper II), provides additional information that can be used to guide management at farm level towards decreasing impacts.

Farmers cannot change the intrinsic conditions of their land, such as soil type or climate, but they can take these factors into account when adjusting their practices and prioritising mitigation measures. If farmers were informed about their farm's specific environmental profile, it would be easier for them to prioritise the most effective measures to decrease their total impact. For example, reduced tillage can reduce nitrogen leaching while increasing N_2O emissions, which may or may not be an acceptable trade-off depending on the magnitude of those emissions, which are site-dependent (Mkhabela *et al.*, 2008), and the site-dependent eutrophication effect of the leached nitrogen. The results in this thesis showed that the optimal fertiliser rate to minimise emissions can vary between sites (*Figure 6*, *Figure 8*). Recommendations on adjusting soil management to minimise environmental impacts must therefore consider site characteristics, which has also been concluded by others (Hijbeek *et al.*, 2019).

Spatial variation is not restricted to variations between farms, but can also be relevant to consider at smaller scales. Considering the sometimes large variability in soil texture and other soil characteristics within farms and even within fields, measures such as precision fertilisation or relocating crops within the farm depending on nitrogen fertiliser requirement could have an important effect on the environmental impact (Delin & Stenberg, 2014; Delin *et al.*, 2005). If LCA is used to guide farm management towards decreasing environmental impact, enabling quantification of these effects could help identify additional emissions mitigation strategies.

There are currently several life cycle-based tools available for use at farm level, *e.g.* Cool Farm Tool (Hillier *et al.*, 2011), BioGrace (https://www.biograce.net/) and VERA (http://adm.greppa.nu/vera). Most of these tools focus on greenhouse gas emissions, but some also provide indicators for leaching, water use *etc*. They typically require farm-specific data, *e.g.* yield, crop rotation and fuel use, and apply built-in models for calculating emissions and impact assessment. The detail at which emissions are modelled varies substantially between different tools (Peter *et al.*, 2017). Adopting site-dependent emissions modelling and impact assessment, as partly done in *e.g.* the Cool Farm Tool (Hillier *et al.*, 2011), and including a broader range of impact categories and indicators could improve the usefulness of these tools. However, the different models applied in this thesis provided vastly different outcomes (*Figure 5*, *Figure 7*), so site-dependent medium-effort models may also entail large uncertainties. As noted by Hillier *et al.* (2011), these tools can therefore mainly provide an initial assessment of mitigation options, whereas more complex models or measurements may be needed to reduce the uncertainties.

Certification schemes

Some food brands have adopted life cycle-based approaches to guide their sustainability efforts, *e.g.* the international consumer goods company Unilever (https://www.unilever.com/) and the Swedish farmers' cooperative Lantmännen (https://www.lantmannen.se/). Implementation of the European Union directive on renewable energy (European Parliament, 2018) has also forced biofuel companies to perform a simplified LCA on their fuels (Ahlgren *et al.*, 2012). Implementation of life cycle management at this level is often connected to a certification scheme, sometimes associated with an eco-label and some kind of impact calculation tool (Keller, 2016). These calculation tools can be the same tools used to guide farm management decisions (Keller, 2016).

Benefits of certification, such as a price premium for certified products, can influence farmers to implement changes in their management (Fairweather, 1999), but the inherent variability of agricultural systems poses a challenge to sustainability certification schemes and sustainability labelling. One issue is whether farmers should be rewarded for the actual or average impact reduction achieved by a management change. Ideally, as changes ultimately occur at farm level, the system will be more efficient in reducing impacts if it promotes management changes that reduce the impacts most on each specific farm. Site-dependent LCAs can be used to achieve this, although they involve more demanding data collection and computing capacity. An issue in this context is that more site-dependent modelling could then potentially favour farms with intrinsic favourable conditions, whereas there is greater potential for absolute impact reductions if the highest emitting farms decrease their emissions. There is also a risk that cultivation at beneficial sites will be rewarded, while uncertified production will continue as usual at non-beneficial sites, without any actual change at either type of site. For certification, it may therefore be reasonable to apply site-generic models. However, management changes that are rewarded in the certification scheme should be validated across different farm types using site-dependent models to verify that the measures will not increase emissions or cause burden shifts to other impact categories under certain conditions. For example, eco-driving and precision fertilisation would most likely decrease greenhouse gas emissions across all farms without burden-shifting, although the magnitude of improvement would vary. Other measures, such as reduced tillage (Mkhabela *et al.*, 2008) and general adjustment of nitrogen level (Paper III), may have varying effects on environmental impact among farms and impact categories, and should therefore be more carefully evaluated for each farm. This is particularly important when the tools used for certification are also intended to be used to give guidance on farm management.

Public policy

Some of the challenges when using LCA in public policy design are similar to the challenges of certification schemes, *i.e*. to promote actual impact reductions without requiring unfeasible amounts of data and computing power. However, public policies by default need to have a broader perspective than certification schemes, since certification schemes can decide which farms to work with but public policy needs to consider all land use, at least within a certain geographical area.

Site-dependent LCA could potentially be useful when evaluating the efficiency of different measures towards achieving policy goals. The framework for the Swedish environmental quality objectives states that impact reductions in Sweden should be achieved without increasing environmental problems elsewhere (Swedish EPA, 2012). The inclusion of 16 different types of environmental quality and the ambition to avoid burden-shifting to other geographical regions makes LCA a suitable tool for evaluating measures for achieving the environmental quality objectives. Another example is the recently adopted National Food Strategy for Sweden (Swedish Ministry of Enterprise and Innovation, 2016), which calls for increased overall food production while achieving the national environmental quality objectives. Considering that the environmental quality objectives are currently not on track to being fulfilled (Swedish EPA, 2019), achieving the goals of the National Food Strategy would require substantial reductions in the environmental impact per unit produced. Site-dependent LCA could be a useful tool to identify where emissions mitigation efforts are most needed, and the measures and geographical locations that could be used to increase production without causing negative environmental effects. It might even be possible to derive recommendations based on farm or site archetypes, using data from site-dependent LCA.

Agricultural land under fallow is currently increasing in Sweden (Statistics Sweden, 2017c), which means that there is potential for relocation of production from currently used land causing high impacts to currently unused land causing lower impacts. Even in a future scenario when all agricultural land is needed to meet the demand for food, feed, fibre and energy, relocation by switching locations could be possible. Since the marine eutrophication impact of an emission varies substantially throughout Sweden (*Figure 10*), moving activities causing high leaching from high-impact areas to lower-impact areas could decrease overall nutrient additions to the Baltic Sea. Such activities could be *e.g.* potato production and animal husbandry with high livestock density (Kyllmar *et al.*, 2006). A modelling study by Hashemi *et al.* (2018) tested how nitrogen leaching could be reduced within two Danish catchments by relocating nitrogen leaching and spatially targeted mitigation strategies. They found that nitrogen load reductions of up to 15% could be achieved if relocations were restricted to occur only within soil types and within farms, and up to 30% if cover crops were also used and relocations within the catchment were not restricted. Site-dependent LCA could be used to expand that approach to include additional emissions and environmental impacts. Public policy aiming to relocate agricultural activities in order to reduce the environmental impact would not necessarily entail forcing certain management upon the farmers, but could instead use *e.g.* economic incentives, as already done to reduce other aspects of the environmental impact of Swedish agriculture (Swedish EPA, 2007).

Implications for life cycle assessment practice

Patouillard *et al.* (2018) noted that few LCA studies identify the need for spatial differentiation in the goal and scope phase. However, the results in this thesis show that the climate and marine eutrophication impacts are strongly linked to the site at which the cultivation occurs, and that model choice highly affects whether this is depicted in the results. This means that the conclusions from crop cultivation LCAs are highly dependent on the handling of spatial information in the LCA process.

Spatial differentiation is first and foremost important when the aim of the LCA is to map the environmental impact of agricultural activities at a certain site or in a certain region (see *e.g.* Avadí *et al.* (2017); Korsaeth *et al.* (2014); Wang *et al.* (2014). However, considering the influence of site on the LCA results can be important even if the explicit aim is broader than that. For example, the spatial variability means that using inventory data derived at other sites poses a risk of biased results, which is important to consider *e.g.* when compiling data for databases and when using these data (Notarnicola *et al.*, 2017). Regional inventories are also important for assessing marginal changes, which by definition occur only in some parts of the system under study (Yang *et al.*, 2018).

It is useful to consider the need for spatial differentiation when planning an LCA study. Ideally, the goal and scope, inventory analysis, impact assessment and interpretation of results should be aligned regarding the spatial resolution. This does not necessarily mean that the spatial resolution should be identical at all stages, since the production systems and environmental mechanisms vary at different scales. For LCAs to be feasible, it is also important that the amount of data needed is reasonable (Notarnicola *et al.*, 2017). However, there is a lack of guidance on how to prioritise spatialisation efforts (Patouillard *et al.*, 2018), and results from detailed modelling can be used for this purpose. As indicated by the results in this thesis, site-dependent modelling of *e.g.* direct N_2O emissions and nitrogen leaching, as well as the choice of characterisation model, can have a substantial effect on the climate impact and marine eutrophication values obtained, respectively.

Overall, it is important for LCA practitioners to differentiate between modelling at different scales and consider that emissions models are not necessarily transferrable between LCAs at different spatial resolutions. The best example is perhaps the IPCC Tier 1 model for soil N_2O emissions. This model was not developed to estimate field emissions, but for assessments at national level (Hergoualc'h *et al.*, 2019). This is a much lower level of resolution than often needed in LCAs, which usually focus on specific products. However, it was clear from the comparison of soil N_2O emissions models in this thesis that the site-dependent models gave diverging results, and therefore not necessarily better estimations of emissions. Therefore, the IPCC Tier 1 model may still be the best available choice for many applications. In those cases, being transparent about the uncertainties in the final results, explicitly discussing the geographical validity and making interpretations accordingly could prevent misinterpretation of outcomes.

5.4.2 Limitations and perspectives

Potential trade-offs in environmental impacts of crop cultivation

This thesis focused on climate impact and marine eutrophication and identified trade-offs between these impact categories regarding both low and high-impact sites and preferred fertiliser rate.

The emissions contributing most to the climate and marine eutrophication impacts of crop cultivation are highly affected by the site conditions (*Figure 5*, *Figure 7*, Paper III). Soil N_2O emissions, SOC change, nitrogen leaching and phosphorus losses are all affected by both the soil characteristics and climate, but their response to changes in these parameters differ. For example, high clay content generally increases soil N_2O emissions and phosphorus losses, but decreases the risk of nitrogen leaching and SOC loss (Rochette *et al.*, 2018; Lal, 2007; Kyllmar *et al.*, 2006; Ulén *et al.*, 2001). The outcome of these and other differing effects is that the crop cultivation at a certain site can cause a low climate impact, but high marine eutrophication, and vice versa (*Figure 9*). Comparing the results for each site in Paper III also showed that the preferred nitrogen fertiliser level to minimise impacts differed between the two impact categories. These trade-offs should be considered *e.g.* when giving recommendations on fertiliser management.

Apart from the two impact categories included in this thesis, other environmental aspects are also affected by fertiliser management and have to be considered if the LCA results are intended to be used in a decision-making context. Since fertiliser management in most cases affects the yield, the resources and emissions per unit yield will change even if the resource use and emissions per unit area remain constant. This affects the results of all impact categories if the functional unit is related to the yield obtained. However, some impact categories are affected by the use of fertilisers in more intricate ways. The most obvious example is freshwater eutrophication. The limiting nutrient in freshwaters is mainly phosphorus, although this also can vary between water bodies (Conley *et al.*, 2009; Elser *et al.*, 2007; Hecky & Kilham, 1988). Unlike nitrogen, there is no process that transforms phosphorus to an unreactive form and removes it to the atmosphere. Emissions from sites where most of the phosphorus is retained before reaching a marine recipient (*Figure 10b*) are therefore more likely to contribute to phosphorus enrichment in freshwaters. Consequently, there may be a trade-off between freshwater and marine eutrophication when comparing the environmental impact of crop cultivation at different sites, especially since soil texture also has a diverging effect on nitrogen and phosphorus losses from soils. Freshwater eutrophication was not assessed in any of the case studies in this thesis, but is important to include if the LCA is used for decision-support.

Fertiliser management is also connected to land use, by affecting both the conditions on the land where the cultivation occurs and the amount of land required to produce a crop unit due to the effect on crop yield. Half of the Earth's ice-free land surface is currently used for agricultural activities (IPCC, 2019), but future projections suggests that, even if some deforestation is allowed, the demand for land can exceed the availability as early as late 2020s (Lambin & Meyfroidt, 2011). Agricultural activities also cause land degradation, further exacerbated by climate change (IPCC, 2019). Agricultural land is thus a scarce resource that needs to be considered when assessing the environmental implications of different fertiliser management strategies. Paper III shows the amount of land required to produce one crop unit (m^2 year CU^{-1}) at each of the fertiliser levels and sites. This is a rather poor indicator of the direct impact of that land use (Milà i Canals *et al.*, 2007a), but is more connected to indirect land use change. Intensification, *i.e.* increasing the output per area unit, has been suggested as a measure to meet future crop demand without causing indirect land use change, sparing land for 'natural' vegetation (Searchinger *et al.*, 2018). However, other studies suggest that better profitability due to increased productivity might instead lead to agricultural land expansion (Lambin & Meyfroidt, 2011). The mechanisms governing global land use are thus complex, and beyond the scope of this thesis, but crucial to include when assessing environmental impacts of fertiliser management at a large scale.

Limitations of case study results

The case studies were performed in the context of mineral fertiliser-based crop cultivation on mineral soils in Sweden, which has some implications for the results. This section discusses how these implications affect the generalisability of the conclusions drawn from the case studies.

First, in the case studies, it was assumed that the mineral fertilisers used were ammonium nitrate or calcium ammonium nitrate. This is a reasonable assumption for Sweden and the other Nordic countries, but urea is more commonly used globally and even in many other parts of Europe (IFASTAT, 2019). Production of urea causes less greenhouse gas emissions per kg nitrogen than production of ammonium nitrates, but urea causes higher NH_3 emissions when applied in the field due to its chemical composition (Hergoualc'h *et al.*, 2019; Brentrup *et al.*, 2016) (*Figure 2*). Further, emissions inventory data for fertilisers produced in Europe, which generally generate less greenhouse gas emissions than fertilisers produced elsewhere (Brentrup *et al.*, 2016), were used in the case studies. Overall, using a different type of mineral fertiliser can influence the total estimated impact (Ahlgren *et al.*, 2009).

Secondly, no organic fertilisers were considered in the case studies, even though manure is commonly used as a fertiliser in Sweden (Statistics Sweden, 2017a). This choice was made to minimise the number of variables influencing the results. Animal manure is often regarded as a waste material in LCAs, *i.e.* no emissions from animal husbandry are allocated to the manure when used as a fertiliser in crop cultivation. Manure also causes higher NH_3 emissions per kg nitrogen applied and provides additional organic material, which affects the SOM dynamics (Hergoualc'h *et al.*, 2019; Kätterer *et al.*, 2011).

Thirdly, a limited range of fertiliser rates was tested in the case studies. In particular, the highest nitrogen fertiliser rate included in the case studies was close to the average rate applied in the respective regions (Paper III). Some studies suggest that N_2O emissions and nitrogen leaching increase exponentially with increasing nitrogen fertiliser rate (Delin & Stenberg, 2014; Snyder *et al.*, 2009). The conclusions from the case studies in this thesis may therefore not be valid for crop cultivation under high nitrogen fertiliser rates.

In addition, the importance of phosphorus emissions in contributing to marine eutrophication is a particular circumstance for Sweden, and does not occur at most other geographical locations. However, it is not unique. A number of countries contribute nutrient loads to the Baltic Sea (Sonesten *et al.*, 2018), and there are other partly phosphorus-limited marine environments in other parts of the world (Barba-Brioso *et al.*, 2010; Gallego *et al.*, 2010; Fisher *et al.*, 1999).

All these factors combined mean that both the absolute impacts and the relative influence of site and fertiliser rate might differ if the LCA were performed in a different context. In addition, field-level assessments accounting for a limited number of impact categories cannot themselves provide sufficient information to function as decision-support. The primary contribution of the case studies is therefore to explore the importance of accounting for site, fertiliser rate and modelling choice, rather than deriving fertiliser management recommendations.

6 Conclusions

- The optimal nitrogen fertiliser rate for minimising both climate impact and marine eutrophication impact depended on the site at which the cultivation occurred. Since the effect of changes in soil management on environmental impact differed between sites, site-dependent modelling of environmental impacts can be useful when using LCA to evaluate *e.g.* the effect of policy interventions.
- The relationship between climate and marine eutrophication impacts of crop cultivation at different sites and at different nitrogen rates were not consistent when site-specific characteristics were taken into account. This meant that some sites gave low climate impact but high marine eutrophication impact, and vice versa, and that the minimum climate impact and marine eutrophication impact per produced unit were not achieved at the same fertiliser rate at most of the sites.
- Direct soil N_2O emissions and SOC changes contributed most to differences in climate impact between sites, while differences in marine eutrophication impact between sites were due to nitrogen leaching, phosphorus losses and characterisation model. These are therefore the most important processes to model site-dependently when aiming to assess site-dependent impacts of crop cultivation using LCA.
- Site-generic models commonly used in LCAs for estimating N_2O emissions and nitrogen leaching at field level gave different results than the site-dependent emissions models. However, the site-dependent models tested also exhibited large variation, so it is not possible to give general recommendations on what easily applicable models to use instead based on these results.
- Including impacts of phosphorus in marine eutrophication indicators for recipients where biomass growth is at least partly limited by phosphorus can have a substantial effect on the estimated impacts. This applies

particularly to emissions to the Baltic Sea, but may also apply to other marine environments.

Methodological contributions

- A new spatially differentiated characterisation method for assessing the marine eutrophication impact of emissions to soil in Sweden was developed, applied in case studies and compared with other available methods. The case studies showed that applying this new characterisation method may alter the preferred alternative when comparing crops cultivated at different sites, compared with using a characterisation method with lower spatial resolution.
- A novel approach to include the effect of increasing soil organic matter content on soil fertility when assessing climate impact of a crop management change was developed and tested in a case study. The case study results revealed that including the this effect had a non-negligible effect on the estimated climate impact.
- Available site-dependent methods to quantify direct N_2O emissions and nitrogen leaching at field level with data typically accessible to an LCA practitioner were synthesised, applied to a case study and compared with regard to quantified emissions values.

7 Future research

The outcomes of this thesis highlight the benefit of applying appropriate models in relation to the goal of the study when estimating the climate and marine eutrophication impact of crop cultivation using LCA. The thesis also provides comparisons of some available options for modelling the impact arising from nitrogen fertiliser use and some new modelling options. However, many questions remain, both regarding the choice of appropriate models and application of the new methods. The following issues should be addressed in future research:

- Site-dependent modelling of emissions and impact characterisation would improve the quality of many LCAs. However, the lack of available models is currently a challenge. Developing an easy-to-use soil N_2O model that is applicable at field scale should be a top priority, considering the high contribution to climate impact of crop cultivation. Applying process-based agro-ecosystem models is one way of decreasing the uncertainties associated with the IPCC Tier I model, but is often not a feasible alternative for LCA practitioners. Using such models to derive site-dependent typical values, as was done for the Swedish nitrogen leaching coefficients in this thesis, could be a way forward. In that context, it would be useful to have more N_2O field measurements for Swedish conditions to verify the outcomes.
- The characterisation model presented in Paper I has high spatial resolution, deriving from the retention data. Aggregating the characterisation factors to a lower resolution would make the model easier for LCA practitioners to apply. It would also be useful to develop characterisation factors for direct emissions to water, so that the model could be used for assessing impacts of emissions from wastewater treatment. Ultimately, a spatially differentiated characterisation model with sufficient spatial resolution and global

coverage, which also provides characterisation factors for airborne emissions and phosphorus, should be the final goal. To achieve that, it would probably be more suitable to add models for phosphorus emissions (in locations where this is relevant) and airborne nitrogen emissions to the existing global marine eutrophication model than to build on the model developed in Paper I.

- The modelling framework presented in Paper II has so far only been applied for the case study described in that paper. It would be interesting to apply the framework to other management practices that affect SOM dynamics, for example harvesting straw to use as feedstock for bioenergy.
- Expanding the case study in Paper III to a more complete spatially differentiated LCA by including more impact categories and more sites and expanding the scope to include implications for land use change would make it more useful as decision support for adjusting nitrogen fertiliser rate. It would also be interesting to identify options for relocating agricultural activities within Sweden or within a smaller region, and assess their potential to mitigate environmental impact.

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Popular science summary

Nitrogen is one of the essential elements required by plants and therefore nitrogen fertilisers are applied to cultivated crops to obtain high yields. Nitrogen makes up a large proportion of the atmosphere (78%), but in the chemical form di-nitrogen gas (N_2), which generally does not react with other substances and therefore cannot be taken up by plants. However, it can be converted into reactive nitrogen by microorganisms or by adding energy in industrial processes. This industrial process gives mineral fertiliser, which is readily available to plants and therefore applied to crops. Not all of the nitrogen applied is used by the crop and the excess can be released into the atmosphere or removed with the soil water, with negative impacts on the environment. These impacts include emissions of the strong greenhouse gas nitrous oxide and nitrogen loads to marine environments, which can cause eutrophication. Manufacture of mineral fertilisers also cause emissions. However, mineral fertiliser use also has a moderating effect on the environmental impact of crop cultivation since it increases the yield, and higher yield means that fewer other resources (for example diesel) are required to produce the same amount of crop. Higher yield also usually means that the crop produces more plant residues, such as roots. These residues contain carbon, so increasing the amount of plant residues can increase carbon sequestration in the soil, thereby reducing the amount of carbon dioxide in the atmosphere and the climate impact. Therefore, a lower nitrogen fertiliser rate, *i.e.* a smaller amount of nitrogen fertiliser per unit area, will not necessarily lower the overall environmental impact of crop production.

Soil carbon sequestration, emissions of nitrogen compounds from the field and crop yield response to fertilisation vary greatly depending on the cultivation site. These variations are due to factors such as soil type, precipitation and temperature. Some types of environmental impact, such as eutrophication, also depend on the site where the emissions take place. All these effects need to be taken into account when assessing the environmental impact of nitrogen fertiliser use and determine *e.g.* which nitrogen fertiliser rate will cause the least environmental impact. A tool commonly used for this purpose is life cycle assessment (LCA). In agricultural contexts, LCA is used *e.g.* to provide guidance to farmers seeking to reduce the environmental impact of their operations and in EU legislation aiming to ensure that biofuel production does not cause large greenhouse gas emissions.

All emissions that occur during a product's life cycle are added up in LCA. For example, an LCA for crop cultivation includes emissions during the production of fertilisers and other input products required for cultivation, fuel consumption by field machinery, emissions of nitrogen and other compounds at the field level, and sometimes net soil organic carbon change. Simple models, which do not account for the influence of site, are often used to describe the relationship between fertiliser use and environmental impact in LCAs. However, due to the complex relationships between fertiliser management, site characteristics, amount of emissions and the environmental impact of those emissions, more sophisticated models are sometimes needed to give accurate results.

Contents of this thesis

The overall aim of this thesis was to improve LCA methodology so that the environmental impact of crop cultivation can be calculated in a relevant way. The focus was on climate and marine eutrophication impacts of crop cultivation in Sweden.

Three main themes were explored; the influence of cultivation site on the environmental impact of crop cultivation, the influence of nitrogen fertiliser rate on the environmental impact of crop cultivation, and how methodological choices affect LCA results. The work included method development and case studies, where data from long-term field trials were used.

Overall results

The case studies showed that cultivation site has a great influence on both the climate impact and marine eutrophication impact of crop cultivation, often greater than the nitrogen fertiliser rate. The difference in climate impact between sites was mainly due to differences in soil organic carbon changes and nitrous oxide emissions from the site where the cultivation takes place. The difference in marine eutrophication impact between sites was mainly due to differences in nitrogen and phosphorus emissions via soil water and in the proportions of these emissions reaching a marine environment, where they can cause eutrophication, which vary depending on the site where the emission occurs. The environmentally optimal nitrogen fertiliser rate was found to vary between sites, and differed for climate impact and marine eutrophication impact. This means that emissions models which do not take site into account can give misleading results, and that there may be a goal conflict between minimising climate impact and minimising marine eutrophication by adjusting the nitrogen fertiliser rate.

In addition to the case results, the thesis also presents new methods that can be used in LCAs of crop cultivation. One is a new model for assessing marine eutrophication impacts of emissions from agricultural land in Sweden. Another is a method to account for the relationship between yield and soil organic carbon when calculating the climate impact of crop cultivation. The thesis also compared different models for calculating nitrous oxide emissions and waterborne nitrogen emissions at field level, and different models for calculating the marine eutrophication effect of the emissions. These comparisons showed that different models give widely varying results, which indicates that it is difficult to calculate the magnitude and environmental impact of emissions with high precision.

Populärvetenskaplig sammanfattning

Kväve är ett av de ämnen som krävs för att växter ska kunna växa, och därför används kvävegödsel i växtodling för att bibehålla en god skörd. 78 % av atmosfären består av kväve, men där finns den i form av kvävgas, som vanligen inte reagerar med andra ämnen och därför inte kan tas upp av växter. Kvävgasen kan däremot omvandlas till reaktivt kväve av mikroorganismer, eller med hjälp av energi i industriprocesser. Det reaktiva kvävet kan tas upp av växter och sprids därför på åkrar i form av mineralgödsel, även kallat konstgödsel. All kväve som sprids tas däremot inte upp av grödan, och kan då släppas ut till atmosfären eller föras bort med markvattnet och orsaka negativ påverkan på miljön. Denna påverkan består bland annat i utsläpp av den starka växthusgasen lustgas och tillskott av kväve till marina miljöer, där det kan orsaka övergödning. Produktionen av mineralgödsel orsakar också utsläpp. Eftersom mineralgödseln ökar skörden har gödslingen dock också en positiv effekt på växtodlingens miljöpåverkan, eftersom mindre andra resurser (till exempel diesel) behöver användas per producerad mängd gröda. Dessutom innebär en högre skörd oftast att grödan producerar mer växtrester, till exempel rötter. Växtrester innehåller kol, och en större mängd växtrester kan därför öka kolinlagringen i marken och därmed minska mängden koldioxid i atmosfären, vilket minskar klimatpåverkan. Trots att kvävegödseln orsakar miljöpåverkan är det därför inte självklart att en lägre kvävegiva, det vill säga en mindre mängd kvävegödsel per areaenhet, ger en lägre miljöpåverkan.

Både kolinlagringen i marken, utsläppen av kväveföreningar från fältet och skördeeffekten av gödsling varierar stort beroende på vilken plats odlingen sker på. Dessa variationer beror på faktorer som jordart, nederbörd och temperatur. Vissa miljöeffekter, till exempel övergödning, beror också på vilken plats utsläppet sker. Alla dessa effekter behöver beaktas när man bedömer miljöpåverkan av kvävegödselanvändning för att till exempel beräkna vilken kvävegiva som är lämpligast för att minimera miljöpåverkan. Ett vanligt verktyg att använda för att beräkna miljöpåverkan av en produkt eller process är livscykelanalys (LCA). I jordbrukssammanhang används LCA till exempel för rådgivning till lantbrukare som vill minska miljöpåverkan från sin verksamhet, och i EU-lagstiftning som syftar till att säkerställa att produktionen av biodrivmedel inte orsakar så stora växthusgasutsläpp.

I en LCA summeras alla utsläpp som sker under en produkts livscykel. Till exempel innefattar en LCA för växtodling utsläppen vid produktion av gödsel och andra insatsprodukter som krävs vid odlingen, arbetsmaskinernas bränsleförbrukning, utsläpp av bland annat kväveföreningar på fältnivå samt ibland nettoomsättning av kol i marken. I LCAer används ofta enkla modeller för att beskriva sambandet mellan gödselanvändning och miljöpåverkan, som inte tar hänsyn till platsens påverkan på utsläppen och deras miljöpåverkan. På grund av de komplexa förhållandena mellan gödslingsstrategi, odlingsplatsens egenskaper, utsläppsmängd och utsläppens miljöpåverkan behövs dock ibland mer sofistikerade modeller för att ge rättvisande resultat.

Avhandlingens innehåll

Det övergripande syftet med denna avhandling är att bidra till utvecklingen av LCA-metodiken, så att växtodlingens miljöpåverkan kan beräknas på ett rättvisande sätt. Avhandlingens fokus är på klimatpåverkan och marin övergödning från växtodling i Sverige.

Avhandlingen är strukturerad kring tre huvudteman; odlingsplatsens effekt på växtodlingens miljöpåverkan, kvävegivans effekt på växtodlingens miljöpåverkan samt hur olika metodval påverkar LCA-resultaten. Avhandlingen omfattar både metodutveckling och fallstudier, där data bland annat från långliggande fältförsök användes.

Avhandlingens övergripande resultat

Resultaten från fallstudierna indikerar att platsen har en stor effekt på både klimatpåverkan och marin övergödning av växtodlingen, ofta större än kvävegivans effekt. För klimatpåverkan berodde skillnaden mellan platserna framförallt på platsens effekt på kolinlagringen i marken samt lustgasutsläppen från fältet där odlingen sker. För den marina övergödningen berodde skillnaden mellan platserna främst på skillnader i utsläpp av kväve och fosfor via vattnet i marken, samt att andelen utsläpp som når en marin miljön där de kan orsaka övergödning varierar beroende på var utsläppet sker. Vidare konstaterades att den miljömässigt optimala kvävegivan varierade mellan platserna, och dessutom var olika för klimatpåverkan och marin övergödning. Detta innebär dels att utsläppsmodeller som inte tar hänsyn till odlingens plats riskerar att ge missvisande resultat, dels att det kan finnas en målkonflikt mellan att minimera klimatpåverkan och att minimera den marina övergödningen genom att justera kvävegivan.

Utöver de tillämpade resultaten presenteras i avhandlingen också nya metoder som kan användas i LCA av växtodling. Den ena är ny modell för att bedöma den marin övergödningen av utsläpp som sker från jordbruksmark i Sverige. Den andra är en metod att ta hänsyn till sambandet mellan skörd och markkol i beräkningar av växtodlingens klimatpåverkan. I avhandlingen jämförs också olika modeller för att beräkna lustgasutsläpp och vattenburna kväveutsläpp på fältnivå, och olika modeller för att beräkna marin övergödningseffekt av utsläppen. Jämförelserna visade att modellerna ger kraftigt varierande resultat. Det är alltså svårt att beräkna utsläppens storlek och miljöeffekt med hög precision.

Acknowledgements

Although I am the sole author of this thesis, it would not exist without the support of a whole bunch of other people. I wish to express my gratitude to all of these people.

First and foremost, thanks to my supervisors Cecilia Sundberg, Per-Anders Hansson and Thomas Kätterer, who contributed their time and expertise along this journey, and my co-author Pernilla Tidåker, who in many ways acted as a mentor to me.

Other colleagues at SLU and elsewhere also took the time to share their knowledge on specific topics and thereby contributed to making this thesis possible. These people include Karin Blombäck, Faruk Djodjic, Elin Widén Nilsson, Holger Johnsson, Anders Larsolle, Helena Aronsson, Thomas Keller, Gunnar Börjesson, Claudia von Brömssen, Håkan Jönsson and Mary McAfee.

Thanks to all my colleagues at the Department of Energy and Technology, for showing that the best results are achieved when we take the time to share our skills. Special thanks to Hanna, for guiding me through the noble art of achieving a PhD, and Emma and Torun, for invaluable friendship.

My deep gratitude to those who don't walk this Earth anymore, but have contributed to making me who I am: Mormor, who taught me the difference between persistence and perseverance. Farmor, who taught me to be proud, but humble. Farfar, who sparked my interest in agriculture, even though our involvement in the field (pun intended) is separated by several decades.

To my family, thanks for being there during the good and the bad times. Mamma, who taught me the art of thoughtfulness. Pappa, who encouraged my curiosity and inspired me to become an engineer. My sister Kim, where all words seem too trivial to describe how important you are to me. Håkan, for being an excellent flat mate (most of the time), and for taking care of my sister and nephew in the best possible way.

And finally, to my nephew Sam, who constantly reminds me of why my research is important.