Technical University of Denmark

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Quantification and critical analysis

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Preface

The research for this PhD thesis was carried out at the Department of Environmental Engineering of the Technical University of Denmark (DTU) under the supervision of Associate Professor Charlotte Scheutz and the co-supervision of Associate Professor Thomas Fruergaard Astrup. Funding for the project was provided by the Danish Council for Strategic Research (Innovation Fund) as part of the IRMAR project (Integrated Resource Management & Recovery; grant no. 11-116775).

This thesis is organised in two parts: the first part puts the findings of the PhD project into context and serves as an introductory review. The second part consists of the papers listed below, referred to in the text by their number given as the Roman numerals **I-III**.

I Klinglmair, M., Lemming C., Jensen L.S., Rechberger H., Astrup T.F., Scheutz C. 2015. "Phosphorus in Denmark: National and Regional Anthropogenic Flows. *Resources, Conservation and Recycling* 105 B (Special Issue: Losses and Efficiencies in Phosphorus Management): 311–324. doi:10.1016/j.resconrec.2015.09.019.

II Klinglmair, M., Zoboli O., Laner D., Rechberger H., Astrup T.F., Scheutz C. 2016. The Effect of Data Structure and Model Choices on MFA Results: A Comparison of Phosphorus Balances for Denmark and Austria. *Resources, Conservation and Recycling* (109): 166–175. doi:10.1016/j.resconrec.2016.03.009.

III Klinglmair, M., Vadenbo C., Astrup T.F., and Scheutz C.. 2016. An MFA-based optimization model for increased resource efficiency: phosphorus flows in Denmark. Submitted to *Resources, Conservation & Recycling*.

Acknowledgements

It has occurred to me that embarking on a PhD is, most of all, an exercise in humility, and often a rather solitary enterprise. Still, looking back, I can't help but notice all the ways in which one might just as well see it as a group effort. I certainly wouldn't be writing this paragraph now, had it just been me, toiling away on my own those last few years.

First and foremost, thanks are due to my supervisors, Charlotte Scheutz and Thomas Fruergaard Astrup, for encouraging me to come to Denmark and giving me the chance to start this PhD project at DTU in the summer of 2012. Thank you so much for giving me the feeling that, even when you were putting on some pressure, you were on my side and there to help.

I further wish to thank my co-authors, Camilla Lemming, Ottavia Zoboli, Carl Vadenbo, Lars Stoumann Jensen, David Laner, and Helmut Rechberger for being constructive as well as, when necessary, unsparing in their criticism, and all of whom were indispensable for making sure that my research and writings made any sense.

This work would have been much more difficult (and less interesting) without the people at the Institute for Water Quality, Resource and Waste Management at TU Vienna. During those short research stays in Vienna, now and then, I got so many insights and good ideas – be it in meetings or just through informal conversations. I'd especially like to thank Johann Fellner, who supervised my Master's thesis at TU Vienna and brought me on the track that eventually led to this PhD thesis.

I am immensely grateful to my wonderful family in Austria, who often had to put up with a rather absentminded Manfred during visits, and whose belief in my abilities always provided encouragement and assurance. Quite often it seemed that you were surer about me than I was myself! You're role models, all of you.

And, of course, what would a workplace be without nice colleagues! Thanks to the Solid Waste Research Group, and in fact the entire department, for creating a friendly and inviting workplace, providing interesting discussions, and never making coming to DTU in the morning seem like a drag. Moreover, thanks to the good people in office 129 for those all-important meetings at the end of the work week.

Mens sana in corpore sano, and so, as a final note, I'd like to express my gratitude to my climbing/walking/beach companions – Raphael, Bentje, Klaus, Pernille,

Paul and anyone who ever came along. Summer has begun again now it appears, and boy do I look forward to it!

Summary

Phosphorus (P) is an essential plant nutrient mined from the earth's crust as phosphate rock. It cannot be substituted, making it a crucial resource for food production. For the EU, future phosphate scarcity is a potential geopolitical and strategic threat. An increasing worldwide phosphate demand is coupled with dependence on imports from a limited number of suppliers outside the EU-28, so that the EU updated its list of critical raw materials in 2014 to include phosphate rock. As a plant nutrient, P is not destroyed by human use, but dissipated into the environment, where it is a pollutant contributing to eutrophication of water bodies and soils. The anthropogenic P is open on the global scale, with global shipments of animal feed, fertiliser, and food; and on the local scale, through the inefficient use of fertiliser or animal manure by application in excess of plant P demand, and losses in waste and wastewater treatment due to insufficient recycling.

The focus of this PhD project was on the resource aspect, as opposed to the pollution aspect, of P in Denmark. The overall goal was to quantify and evaluate the country's anthropogenic P flows, i.e. those flows caused or significantly influenced by human action, based on a comprehensive material flow analysis (MFA). MFA is a method widely applied to establish resource budgets within a spatial – such as a country – and temporal system boundary, establish a material balance, and handle data uncertainties and data conflicts.

When looking at P from a resource efficiency perspective, the most important flows to consider are those linked to agriculture, as a consumer and producer of large P flows, and waste/wastewater management, as the key processes for treating the resulting P-containing wastes. Country-wide average values regarding these processes hold limited informative value. Moreover, it became clear at the outset of the study that there were distinct differences between the P flows across regions of the country, especially between the east, with the largest urban agglomeration, and the northwest. Apart from population and industrial density, a contrast also exists in agricultural practice, with animal husbandry concentrated in the west and northwest, and the east being dominated by crop production. For the agriculture and waste management processes, the MFA was divided into 3 "typical" regions between the northwest (North Jutland), the east (Zealand and the capital region), and a middle part with more mixed characteristics (Mid-Jutland and Southern Denmark); the regional subdivisions formed a part of a complete country-scale MFA. As is typical for a European country, the Danish P budget showed a strong dependence on P imports in fertilisers and animal feed; with food products being the dominant export of P. The regional contrasts in agricultural P budgets were pronounced

as expected, with a slight P deficit in the east and the largest per-hectare surplus, due to large amounts of manure, in the northwest. Manure was shown to hold the most salient potential for P recovery, yet stays quite local and adds to the surplus in the country's northwest, posing a environmental problem. In the waste management system, two streams were identified to hold significant potential for P recovery. Sewage sludge, while already applied to land on a considerable scale, still holds potentially recoverable P not yet utilised today; and vegetable and animal kitchen and food waste from consumption currently not collected separately, with residues being lost to P recovery. These amounts are furthermore located in the east, with a slight P deficit in agricultural soils, suggesting themselves for substituting some fertiliser imports in the future. The total P quantities in these streams amounted to approximately 35% of concurrent mineral P imports.

Since MFA for regional resource budgets is often the groundwork for further analysis, the robustness and comparability of MFA studies' outcomes when using them as sources of information is important. To this end, the MFA for Denmark was compared to a recent and methodically similar P MFA for Austria, and the effects of the structure of the data material and an MFA practitioner's modelling choices on the outcomes identified and measured. It was demonstrated that the data available do, in fact, influence model layout. Moreover, the approach to assess uncertainty is subject to a certain degree of arbitrariness, and reflects the modeller's belief in the quality of the data material. This, however, leads to incomparability of data quality between MFAs, as the comparison showed, since data uncertainties can be only evaluated against those in the same model. Lastly, data conflicts are normal in country-scale MFA; the extent of the necessary reconciliation of conflicting data provides a useful proxy measure for the quality of an MFA. Metadata matter; this comparison showed the quantitative effects of those aspects of MFAs not resulting from the real-world systems studied. The results thus gave a quantitative basis to requiring a transparent system definition and data characterisation in regional MFA beyond the case of P in Denmark.

A third part of this PhD project consisted of exploring the potentials for increased P recovery efficiency in the Danish anthroposphere based on the results of the initial MFA. An aspect of a secondary resource (recovered P fertiliser) is its ability to fulfil the functions of the resource substituted (mineral P fertiliser). For this purpose, the MFA system and values obtained in this study were adapted to reflect the typical values for availability of P from various material flows to crops, and to allow for transport of less bulky secondary-P material flows. An optimal distribution of recovered P flows (from sewage sludge and composted organic household waste) was then determined by formal optimization via linear programming. The outcome showed a gradual decline of both mineral P inputs, and net additions to soil P stocks, stabilising at a distinctly lower level than evident from the static MFA, due to P applied gradually becoming available for plants over time, showing a significantly higher (82%) potential for substituting mineral P imports than evident from the initial, static MFA (35%). While the potential improvements in closing the P cycle could be shown, this can, however, not be expected to change the reliance on imported P on one or another form.

Dansk sammenfatning

Fosfor (P) er et essentielt plantenæringsstof, der udvindes fra jordskorpen som råfosfat. Fosfors rolle i fødevareproduktionen er afgørende, idet der ikke findes noget erstatningsstof. Fremtidig fosfatmangel udgør derfor en potentiel strategisk og geopolitisk trussel mod EU. En stigende verdensomspændende efterspørgsel kombineret med afhængighed af import fra et begrænset antal leverandører lokaliseret i et fåtal af lande uden for EU har ført til tilføjelsen af råfosfat på EU's liste over kritiske råstoffer i 2014. Den intensive anvendelse af P i landbruget medfører at P spredes i miljøet, hvor det fører til øget nærringsstofbelastning og eutrofiering af vand- og jordområder. Det antropogene P-kredsløb er på globalt plan åbent via foder-, gødnings- og fødevaretransport; og på det lokale plan via ineffektiv brug af kunst- eller husdyrgødning ved anvendelse, der overstiger planternes P-behov, og tab i behandlingen af affald og spildevand pga. utilstrækkelig genanvendelse.

Det overordnede mål med dette PhD-projekt har været at kvantificere og evaluere Danmarks antropogene P-strømme, dvs. de strømme der enten er forårsaget af eller under væsentlig menneskelig indflydelse. Projektet har haft et ressourcemæssigt fokus fremfor et forureningsorienteret og er udført på baggrund af en omfattende massestrømsanalyse (material flow analysis/MFA). MFA er en metode, der ofte anvendes til at udarbejde ressourcebudgetter indenfor en geografisk—f. eks. et land—og temporal systemgrænse, etablere en massebalance, og håndtere datausikkerheder og datakonflikter.

Når man ser på P ud fra et ressourceeffektivitetsperspektiv er de væsentligste strømme dem, der er forbundet med landbrug, som både forbruger og producerer store P-strømme, samt affalds- og spildevandshåndtering, som er nøgleprocesserne for behandlingen af det deraf følgende P-holdige affald. Landsdækkende gennemsnitsværdier for disse processer har begrænset informationsværdi. Det blev endvidere klart fra undersøgelsens begyndelse, at der var tydelige forskelle mellem P-strømme på tværs af landets regioner, særligt mellem det østlige Danmark, hvor den urbane agglomeration er størst, og den nordvestlige tyndere befolkede del. Ud over befolknings- og industriel tæthed findes også forskelle i den landbrugsmæssige praksis, da husdyravl er koncentreret i det vest/nordvestlige Danmark, mens planteavl er mere udpræget i det østlige Danmark. I MFA'en blev landbrugs- og affaldshåndteringsprocesserne inddelt i tre "typiske" regioner; den nordvestlige (Nordjylland), den østlige (Sjælland og hovedstadsregionen) og midten med mere blandede træk (Midtjylland og Syddanmark), og denne regionale underinddeling blev integreret i den landsdækkende MFA. Det danske P-budget

viste en stor afhængighed af P-import i form af mineralsk gødning og dyrefoder, hvilket er typisk for et europæisk land, mens fødevare udgjorde den største P-eksport. Den regionale kontrast i landbrugets P-budget gav det forventede udslag med et let P-underskud i den østlige region, mens det største overskud pr. hektar fandtes i den nordvestlige region pga. store gødningsmængder fra dyreavl. Husdyrgødning viste sig at have det væsentligste potentiale for P-genvinding, men forbliver lokalt og bidrager til det nordvestlige overskud, hvorved det udgør et transportproblem. I affaldshåndteringssystemet identificeredes to strømme med signifikante potentialer for P-genvinding; spildevandsslam, som på trods en betragtelig eksisterende anvendelse på jord stadig indeholder P til potentiel genvinding; og organisk affald i dagrenovation, som på nuværende tidspunkt ikke indsamles separat hvorved restmateriale til P-genvinding går tabt. Disse mængder er endvidere placeret i den østlige region med et let P-underskud i landbrugsjorde, hvilket indikerer en mulighed for fremtidig erstatning af P mineralsk gødningsimport. De totale P-mængder i disse materialestrømme blev kvantificeret til ca. 35% af den samlede nationale import af mineralsk P.

Eftersom MFA af regionale budgetter ofte danner baggrund for yderligere analyse er det vigtigt for brugen af deres udfald som informationskilde, at de er robuste og sammenligningsegnede. Med dette formål foretoges en sammenligning mellem MFA'en for Danmark og en aktuel og metodisk lignende Østrigsk P-MFA, og effekterne af datamaterialets struktur og forfatterens præferencer i forhold til MFA-modellens opbygning blev målt og identificeret. Det blev demonstreret, at den tilgængelige data faktisk har indflydelse på modelopbygningen. Endvidere er tilgangen til bedømmelse af usikkerheder underlagt en hvis grad af tilfældighed og afspejler forfatterens tiltro til datamaterialets kvalitet. Dette vanskeliggør sammenligning af datakvalitet mellem MFA'er, hvilket denne sammenligning også viste, eftersom datausikkerheder kun kan evalueres i forhold til samme model. Endeligt er uoverensstemmende data forventelige i en landsdækkende MFA; omfanget af nødvendig afstemning af uoverensstemmende data giver en brugbar proxy-måling for en MFAs kvalitet. Metadata er afgørende; denne sammenligning afslørede kvantitative effekter af de aspekter af MFA'en, som ikke stammede fra de undersøgte systemers faktiske karakteristika. Resultaterne gav et kvantitativ grundlag for at kræve en transparent systemdefinition og datakarakterisering i regionale MFAer generelt, ikke kun i forhold til P i Denmark.

Den tredje del af projektet undersøgte potentialet for øget P-effektivitet i den danske antroposfære baseret på resultaterne af den indledende MFA. Et aspekt af sekundære ressourcer (genvundet P-gødning) er dets egnethed til at opfylde den erstattede ressources (mineralsk P-gødning) funktioner. Med henblik herpå

tilpassedes MFA-systemet og dets opnåede værdier for at afspejle de typiske plantetilgængelighedsværdier for P fra diverse materialestrømme til afgrøder samt at tillade transport af mindre pladskrævende sekundære P-materialestrømme. Optimal distribuering af genvundne P-strømmer (fra spildevandsslam og kompostering af organisk husholdningsaffald) bestemtes på baggrund af optimering via lineær programmering. Resultaterne viste et gradvist fald i behovet af mineralske P-input samt akkumulering af P i landbrugsjord , som stabiliseredes på et væsentligt lavere niveau end i den statiske MFA pga. den gradvise tilførsel af P og følgende tilgængelighed for planterne over tid. Således blev det vist, at ved at genvinde P fra spildevandsslam og organisk husholdningsaffald kan importen af minarals P gødning reduceres med 82%, hvilket er væsentlig højere end de 35% fundet i den statiske MFA. Selvom der kunne påvises et potentiale for at opnå et mere lukket P-kredsløb end i dag, vil man dog kunne eliminere afhængigheden af importeret P i en eller anden form.

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Abbreviations

AT	Austria
CV	Coefficient of variation
DK	Denmark
GIS	Geographic information system
LCA	Life cycle assessment
LCI	Life cycle inventory
MFA	Material flow analysis
K	Potassium
N	Nitrogen
P	Phosphorus
SFA	Substance flow analysis

1 Introduction

1.1 Background and research motivation

The flows, stocks, and losses of a given natural resource are commonly registered at a disparate multitude of points within a country or region. The nature of the information obtained and recorded depends very much on the stakeholders involved, e.g. a business, a specialized government agency or consulting firm, or a national statistics office. On the scale of larger geographical areas, such as countries, material flow analysis (MFA) — often called substance flow analysis (SFA) when studying a substance[1] — allows for visualizing the anthropogenic turnover of materials. The approach, as defined by Baccini and Brunner (2012), has become a widespread tool for establishing a quantitative basis on which to build resource conservation efforts.

In 2014, the European Commission included phosphate rock in its list of raw materials critical for the European Union (European Commission 2014). Phosphorus (P), mined from phosphate rock, is an essential plant nutrient and cannot be substituted; its role for food security is crucial. Scarcity of P for the EU is a potential economic and strategic threat, since European rock phosphate reserves are extremely limited; in 2014, the EU-28 exported 27 Gg and imported 5949 Gg of phosphate rock (IFA 2016). At the same time, the P cycle is wide open on the global scale (Ashley et al. 2011), with shipments of food, livestock feed and fertiliser adding to the P balance of exporting regions, while necessitating the replenishment of soils in regions with agricultural production. Dissipative losses, inefficient use in agriculture, or insufficient recycling cause this open cycle to be mirrored at the local scale, both reducing our ability to recover P from waste, and adding to the pollution potential of P losses to the environment. MFA can here offer a comprehensive grasp of how P is cycled through the anthroposphere of within a specific; with a view to existing and potential future policies regarding the subject, a country-level analysis suggests itself.

Like most EU member states, Denmark has no noteworthy phosphate deposits. Despite the country's homogeneous topography, the anthropogenic P cycles are

[1] The more generic term "MFA" will be used throughout this thesis.

clearly different between the Danish regions. Its intensive and industrialised agriculture and modern waste management system offer a good data background, so that the Danish example lends itself to an exploration of existing improvement potentials based on a detailed quantification of P flows in the country. Annual reports on the nutrient balances (for nitrogen, phosphorus, and potassium) in Danish agriculture are readily available from the Danish Centre for Food and Agriculture at Aarhus University (e.g. (Vinther & Olsen 2014); however, a complete picture of the Danish P household necessitates the inclusion of all other significant flows in the anthroposphere. Flows to and from the agri-food system tend to be an order of magnitude larger than other anthropogenic flows of P, while the relevant waste flows offer significant potential for recovering a much higher fraction of P than is presently the case. A focus on the agri-food and waste management sectors is therefore pertinent from a resource-efficiency standpoint. In waste management, the assessment of P recovery potentials, and the treatment options on hand, are closely connected to the substitution potential of recovered materials. The question is to which extent recovered P can actually substitute primary P imports in agriculture in a given system – or whether, as was the hypothesis of recent research by Hanserud et al. (2015) on Norway, recovered P could even lead to full independence of primary P imports. The optimal redistribution of recoverable P is, then, always constrained by its value as a fertiliser, and its ability to be transported.

When establishing a detailed material balance of a region, such as the Danish P balance, the question of representing the system in an unbiased and accurate manner poses itself. In MFA studies at the regional and country level, usually based on secondary data sources such as official statistics, technical reports, scientific literature, etc., instead of measurements, the practitioner enjoys considerable freedom in the definition of flows and processes, resolution of the model, assessment of data quality, and system layout. When drawing on an MFA study as a source of information, the data material, assumptions, and subjective choices become important because of their influence on results. The question arises, then, to which extent the outcome of an MFA depends on the arbitrary choices taken by the MFA practitioner – is it reasonable, when drawing upon data reported in an MFA, to take these at face value?

1.2 Research objectives

A comprehensive quantification and analysis of anthropogenic phosphorus flows in Denmark (Klinglmair et al. **I**) forms, *first*, the quantitative groundwork of this PhD thesis. The objectives were to:

- establish a comprehensive MFA of P in the Danish anthroposphere;
- integrate sub-national, regional-level analyses of P in the agriculture and waste management sectors;
- identify and quantify the imbalances of the anthropogenic P household within the country and the regional amounts of potentially recoverable P.

Second, the robustness of regional MFA studies was of interest (Klinglmair et al., **II**), i.e. those aspects not resulting from the physical properties of the system studied. The objective in this regard was to:

- compare the P MFA study forming a part of this thesis to a recent P MFA study for Austria of similar scope;
- demonstrate and quantify the influence of the MFA practitioner's choices on the outcome of a given study;
- suggest an approach to identify these effects from an MFA's metadata when using an MFA as a source for information.

Third, the question of optimisation potentials (Klinglmair et al. **III**) is inherent in the regional MFA presented in this thesis. In the case of P, the aim was to

- adapt the static P MFA of Denmark for optimization using a formal linear-programming approach to minimise mineral fertiliser imports, taking into account transport and P plant availability of recycled P products;
- to outline the utility of various pools of potentially recoverable P in substituting mineral fertiliser;
- suggest an optimal redirection of the P flows identified in the MFA.

2 State of the field

The application of MFA to study flows and stocks of P in the anthroposphere, on the scale of countries or large regions, has seen a distinct increase in interest in recent years. Without claiming exhaustiveness, several studies from the European context shall be noted here. Ott & Rechberger (2012) established a P balance of the year 2011 for the EU-15 as an aggregate, encompassing all major anthropogenic P flows and the pertaining processes. On the national level, Binder et al. (2009) published a comprehensive P balance of Switzerland, while Egle et al. (2014a) did the same for Austria. Zoboli et al. (2015) expanded the latter, with minimal changes to the system layout, to a 22-year timeline, tracing historical changes in the waste flows over time and identifying potential systematic errors in the model. Other studies have a narrower focus on individual economic sectors. Antikainen et al. (2004) and Sokka et al. (2004) published detailed P balances for the forestry sector and waste management in Finland, respectively. Cooper & Carliell-Marquet (2013) studied the P flows in the food production and consumption system in the UK. In the context of Danish agriculture, Vinther & Olsen (2014) have annually been updating a 20-year timeline of nutrient balances and nutrient surpluses (N, P, K) in Danish agriculture, concentrating on the overall balances of inputs and outputs of nutrients. Senthilkumar et al. (2011) focussed their study both geographically and by sector and demonstrated, in their study of P flows in the food production sector of several selected French regions, the importance of agricultural production systems on regional P budgets. An overview of regional P balances (including outside of Europe) is provided by Chowdhury et al. (2014).

From an overview of studies such as the above, MFA studies on a regional, country, or multi-country level appear quite consistent with respect to system boundaries and use of data to determine overall in- and outputs (e.g. external trade statistics). In the case of P, Seyhan (2009), with Austria and Turkey as examples, suggests a template of standard system boundaries, flows, and stocks to be followed in a country-scale P balance for resource conservation; by and large, this layout is also followed, albeit not explicitly, by the P balance for the EU-15 by Ott & Rechberger (2012) and the meta-study of the EU-27 based on national P balances by van Dijk et al. (2015). Yet it can be observed e.g. from the above-mentioned review of P balances across the globe by Chowdhury et al. (2014) that the metadata and modelling choices by the authors of MFA studies, not necessarily limited to P, are disregarded while data reported in an MFA are taken at face value. This issue pertains, to a large part, to the data material used in establishing an MFA and the quality thereof, which is closely linked with how uncertainties in a model are assessed. In short, the assessment of uncertainties in regional MFA studies are subject to a degree of arbitrariness. Laner et al. (2014) proposed a step-by-step framework for handling uncertainty assessment in MFA, depending on data quality and the goal and scope of the MFA in question. Laner et al. (2015a) demonstrated an approach to estimating the uncertainty of MFA input data, building on a so-called "pedigree matrix" originally put forward by Weidema & Wesnæs (1996) in a life-cycle inventory (LCI) context. In this matrix, data quality is assessed according to a set of quality indicators such as reliability, completeness, or temporal and spatial correlation. Schwab et al. (2016) presented a framework for the systematic characterization of the data material used in an MFA by tagging each datum with a set of attributes found in a more detailed characterization matrix, while Laner et al. (2015b) explored the application of two different uncertainty concepts to MFA, namely, normal distributions such as are used in STAN, and fuzzy numbers, stressing the usefulness of the latter for poor information.

The suitability, and handling or treatment options, of various waste streams for recovering P have likewise come to prominence in recent years. Egle et al. (2014b; see also Lederer & Rechberger 2010) assessed the environmental, economic, and technological challenges in a range of treatment options for sewage sludge with a view to P recovery. The Danish Environmental Protection Agency (Miljøstyrelsen 2013) published a comparable report focussing on the Danish situation; both reports point, among other things, to the potential inherent in long-term storage ("banks") and later treatment of sludge mono-incineration ash. The potential of P in waste streams to substitute primary P import has seen attention e.g. in the UK, where Bateman et al. (2011) explored this potential, as well as that for remedying imbalances in P budgets within the country and identify transport as a major hindrance. Senthilkumar et al. (2014) pointed to the potential for P recovery in wastewater and municipal waste in France.

3 Materials & methods

3.1 Material flow analysis: data sources

The first step to assess the improvement options in the anthropogenic turnover of any resource is its quantification. In determining the flows and stocks of a resource on the country scale, the use of primary data, i.e. data measured first-hand by the investigator, is generally impractical, or rendered a practical impossibility by resource and time constraints. Instead, the MFA practitioner in this case relies on data collected from widely heterogeneous secondary sources, such as official statistics on external trade or domestic production and consumption of goods, technical reports and studies from universities or official authorities, peer-reviewed scientific literature, information from businesses, or expert estimates.

For the MFA of Danish P flows presented in Klinglmair et al. (**I**), the data sources drawn upon for determining both mass flows and P concentrations were official statistics on external trade and agricultural production from Statistics Denmark, and data from the Danish Environmental Protection Agency (Miljøstyrelsen), the AgriFish Agency (Naturerhvervstyrelsen), the Danish Nature Agency (Naturstyrelsen), Eurostat, the European Environment Agency, and the Danish Food Composition Databank. This information was supplemented by official reports from the Danish authorities, peer-reviewed journal publications, and data published by businesses (e.g. in the case of agricultural biogas production). A complete overview of flows and data sources for both mass flows and P concentrations in those mass flows is listed in the Supplementary Information for Klinglmair et al. **I**.

Furthermore, the comparison of country-scale P balances (Klinglmair et al. **II**) was based on the outcomes presented in Klinglmair et al. **I** and on a recent MFA of P in Austria by Zoboli et al. (2015), conducted by the Vienna University of Technology.

3.2 Material flow analysis: approach

Material flow analysis (MFA), as it was used in the context of this thesis, is based on Baccini & Brunner (2012), and is elaborated further by Brunner & Rechberger (2004). These works outline MFA primarily as a tool to design a model of the physical flows and stocks of a material or substance within defined spatial and temporal boundaries. Often, this is applied to the anthropogenic (caused by human activity) metabolism of a particular resource in a certain geographical area, such as a city or country. Since the work in this thesis concerns a substance (P), the term “substance flow analysis” is a more exact term.

The software STAN version 2.5 (Cencic & Rechberger 2008; www.stan2web.net) was used to establish the MFA systems in this work. STAN 2.5 is freeware available from Vienna University of Technology. It allows for visualisation of MFA diagrams, the calculation of unknown flows and stock changes by way of balancing the system, the consideration of uncertainties, error propagation, and reconciliation of conflicting, uncertain data. Especially in the case of complex MFA studies over a large geographical area, these values are based on a variety of sources (see section 3.1), which differ in quality; these data can be expected to contradict each other in many instances, so that the inputs and outputs of processes will not balance. For the purposes of STAN, all uncertain flows are assumed to be random, normally distributed variables defined by mean value and standard deviation. If an MFA system is overdetermined, i.e. more variables are known than necessary for solving the balance equations, unknown quantities (flows and stock changes) can be calculated, while Gaussian error propagation allows for the reconciliation of conflicting uncertain input data. The problem of data conflicts is solved in STAN by a least-squares optimisation of the changes of input data necessary to balance the system. These changes in input data are weighted by the inverse of the measurement variance (square of the standard deviation), i.e. values with high uncertainty will change more strongly compared to values with low uncertainty (cf. Laner et al. 2014). Data reconciliation furthermore results in the reduction of the uncertainties of reconciled flows due to the additional information available in the entire system.

Since the data material drawn upon for establishing the Danish P MFA in this thesis consisted of isolated, separate values from sources of varying quality, a part of the task, as a crucial part of any regional MFA, was to assess uncertainties in a transparent and structured way. The approach put forward by Hedbrant & Sörme (2001) for estimating the uncertainties in urban metal stocks was adapted for the case of P. Data sources were categorised into 6 types, ranked according to their reliability, and assigned an uncertainty level corresponding to an “uncertainty factor”. Since multiplication and division by this factor leads to increasingly asymmetrical intervals as uncertainty increases, we draw upon the adaptation of Laner et al. (2015b): the uncertainty factors are converted into coefficients of variation (CV), with the mean value multiplied with the uncertainty factor defined as the mean value plus two standard deviations, and the symmetric interval around the mean value corresponding to a 95% confidence interval. An uncertainty level (and resulting CV) is assigned both to the material (bulk) flow and the P concentration of the flow, with one CV resulting for the P flow shown in the MFA. In Klinglmair et al. **I**, 5 uncertainty levels corresponding to a type of data source were used; the combinations of uncertainties for material flows and P concentrations resulted in 11 discrete CVs (uncertainty intervals) for characterising the uncertainties of the P flows shown in the model.

3.3 Adaptation of the MFA and optimisation

To assess the optimisation potential inherent in the results shown in Klinglmair et al. (**I**), a relatively simple linear programme was set up using GAMS 24.4 (General Algebraic Modelling System, www.gams.com). GAMS is a modelling environment for mathematical programming and well-suited to optimization. The STAN model from Klinglmair et al. (**I**) was simplified to display only flows concerning the agri-food and relevant waste management processes, resulting in an MFA system with 28 processes and 75 flows (in total, showing 3 regional subsystems for the regions defined in Klinglmair et al. **I**). The pertaining code and process-product matrix are available in the supplementary information for Klinglmair et al. (**III**).

Since P applied to the soil becomes adsorbed to soil solids and becomes gradually plant-available in the soil solution, it can remain in the soil for years and keep adding to the plant available P pool over time (Syers et al. 2008; Ylivainio & Turtola 2013). Soil pH, crops, climate, and pest control furthermore influence plant P availability. For the adaptation of the MFA in this thesis, a strongly simplified approach was adopted by using generic literature values for Europe (see Klinglmair et al. **III**).

The optimisation model is to minimise mineral fertiliser import while satisfying defined agricultural P demand; waste streams from consumption are held constant, as e.g. a sudden change in diet is not accounted for in the model. The decision variables of the model, then, were considered the throughput (activity levels) and locations of a set of handling or treatment options for material flows holding potentially recoverable P: 1) manure, 2) sewage sludge applied directly on land, 3) sewage sludge incineration ash, and 4) compost from composted organic household waste. The case of the latter two options do not yet represent widespread current use in Denmark; organic household waste is generally not collected separately, while ash stored from sludge mono-incineration is considered to hold considerable potential for future treatment and use (e.g. Lederer & Rechberger 2010;

Miljøstyrelsen 2013). Garden waste does not form a part of the organic household waste; it is currently collected separately, composted, and mainly used in private gardens. The model is constrained by agricultural demand for P and the amount of P in relevant waste streams, as reported in Klinglmair et al. (**I**), the recovery rates of the respective handling options, and crop availability of P from each input over a 1-year timestep, with 3 yearly time steps chosen for this study.

4 Results & discussion

The results of this study are provided and discussed in three parts: 1) the results of a comprehensive MFA of the Danish anthropogenic P household, including regional MFAs for agriculture and waste management; 2) the outcomes of a comparison of the Danish MFA with a recent P MFA for Austria, to identify and quantify the influence of modelling choices and data structure on an MFA outcome; and 3) the results of an adaptation of the Danish MFA via linear programming, taking into account the improvement potentials of recovered P flows.

4.1 MFA of the Danish phosphorus cycle

4.1.1 System layout and assumptions

The system boundary was defined as the land area of Denmark, without the Faroe Islands and Greenland, and excluding the hydrosphere. Since the focus of this study was on resource recovery, it did not concern itself with the fate of P losses after they had entered water bodies. The temporal system boundary was one year (2011); this was due to highest availability and completeness of recent data material at the time of writing. It is to be noted that this presents a snapshot in time not intended to discuss longer-term trends and fluctuations in the anthropogenic P cycle. A 3-year average value was used when data for adjacent years were available, to account for annual variations especially in agricultural production.

While the topography in Denmark is relatively homogeneous, it became clear after the outset of the MFA study that differences in agricultural practice, as well as population and industrial density would render a purely country-scale MFA less meaningful than a regional consideration. This was especially the case for the two sectors mostly of interest from a resource-efficiency viewpoint, i.e. agriculture and waste management. Such regional subdivision is dependent on the system under study, and other divisions may yield more insight in other countries. Yet, agriculture and waste management are invariably key sectors for resource-efficient P use, showing the largest anthropogenic turnover of P, and flows of potentially recoverable P, respectively. For these sectors, the system was divided along administrative borders into three regions (Table 1), designated A, B, and C. Region A, the eastern island of Zealand (Sjælland) includes the capital region (Hovedstaden) which in turn encompasses the island of Bornholm in the Baltic Sea, and is the most densely populated region. It is dominated by arable crop production. Region C (Nordjylland, or North Jutland) shows the sharpest contrast, being dominated by intensive livestock production (mostly dairy and pigs) with accompanying fodder crop production, and the lowest population density. Region B shows a mix of crop and animal production in the eastern and more intensive animal production in the western part, with a population density also between A and C. Since these distinctions in region B do not occur as clearly along administrative borders, limiting data collection, the regions Midtjylland (Mid-Jutland) and Syddanmark (South Denmark) were combined in one region, region B, of mixed characteristics.

Table 1. System overview showing basic background information relevant to this study, and the region designations (A, B, C) used. Livestock units are numbers of livestock normalised by excretion (of nitrogen) of different types of livestock (Miljøministeriet 2010). These are here used as a proxy to show the intensity of livestock production.

Region		Population		Area		Agricultural area		Livestock units	
		1000	*% of total*	*1000 ha*	*% of total*	*1000 ha*	*% of total*	*1000*	*% of total*
A	Sjælland Hovedstaden	2,532	45%	983	23%	558	22%	3,208	11%
B	Syddanmark Midtjylland	2,468	44%	2,533	59%	1,537.9	60%	24,438	65%
C	Nordjylland	580	10%	791	18%	470.8	18%	9,145	24%
DENMARK TOTAL		5,580	100%	4,308	100%	2,566.8	100%	36,792	100%

The MFA model, as set up in STAN 2.5, consists of 166 flows and 50 processes including three regional subsystems each for *agriculture & forestry*, and *waste management*, and several “virtual” processes for layout (routing of flows) purposes.

On the country level, the system encompasses the five main processes *industry & trade*, *consumption*, *wastewater treatment*, *agriculture & forestry*, and *waste management*. On the regional level, the *agriculture & forestry* process was furthermore divided into three regional subsystems with an *agricultural soil* process (to which P is applied and from which it is taken up by crops, as well as lost), *crop production*, *animal production*, and *biogas production*. The latter was included in agriculture (as opposed to waste management) since it comprises mainly farm-scale and joint (centralised) biogas plants receiving mainly manure, with these flows still relatively minor. The regional *waste management* subsystems contain the processes *sludge treatment*, *incineration*, *composting*, and *landfills & construction*, as well as the "virtual" processes *input organic waste* and *input solid waste* for routing P flows from these waste streams to different treatment processes. The input flow of household waste (called MSW in the figure) includes household waste and comparable waste from businesses and services (e.g. shops, offices, restaurants), excluding bulky waste. It was split into a biowaste and non-biowaste residual fraction for merely illustrative purposes to visualise the potential of P recovery from the former. The household biowaste fraction, as defined in the EU Waste Framework Directive (European Parliament & European Council 2008) consists mainly of garden waste, and kitchen vegetable and animal food waste; in this study, garden waste is the main part of a separate flow generically called organic waste, since it is collected separately and treated mainly by composting.

4.1.2 P flows & recovery potentials

The STAN diagrams in Figures 1-3 show the P flows in Denmark on the country scale (Figure 1), and regional scale for agriculture (Figure 2) and waste management (Figure 3). All values reported are values after data reconciliation in STAN (cf. section 3.2). A complete list of flow values, stock changes, and uncertainty ranges, as entered and after the data reconciliation step, can be found in the supplementary information for Klinglmair et al. **I**.

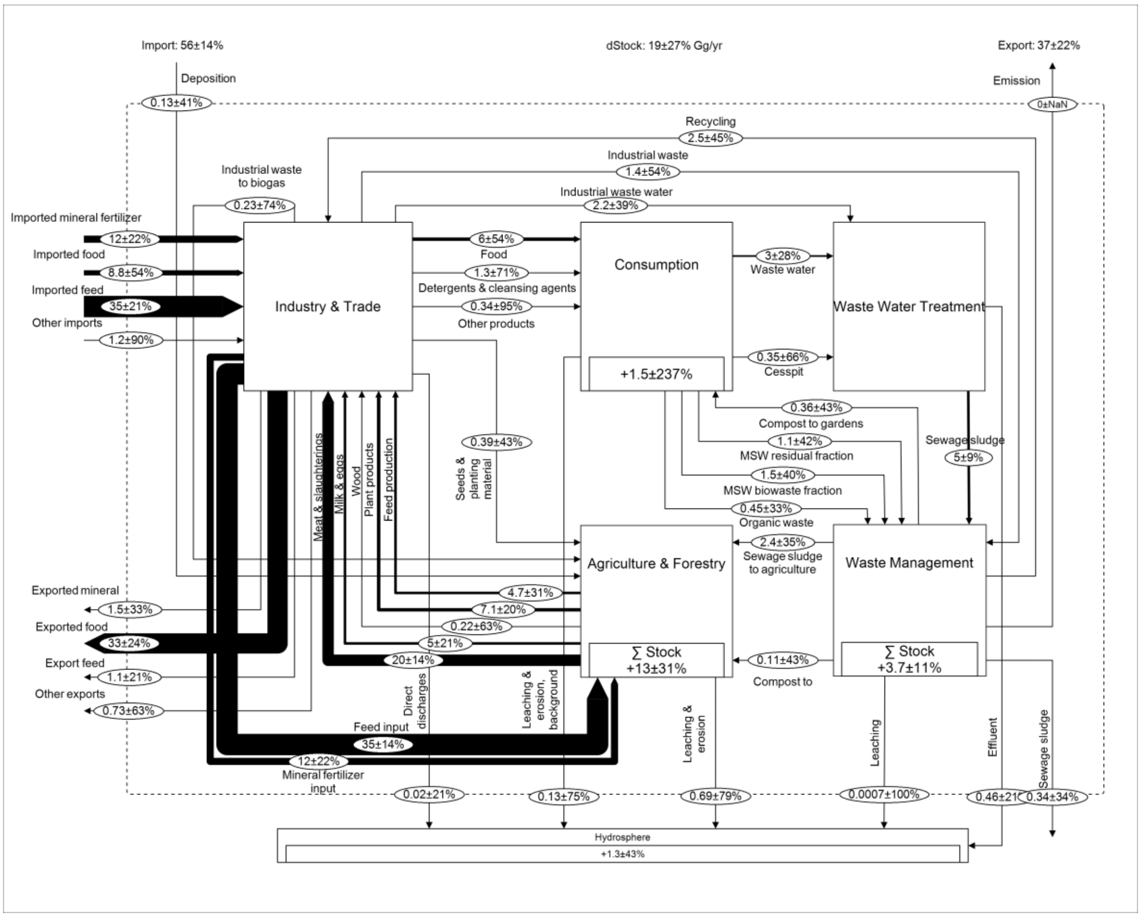

Figure 1. Diagram of P flows [Gg yr^{-1}] in Denmark in the year 2011. Arrows show substance flows of P, with arrow width corresponding to flow size. The system boundary is shown as a dashed line. Flow values and relative uncertainties in oval shapes. Where applicable, stock changes (and relative uncertainties) shown in boxes inside the process symbols. Diagram shows reconciled values.

As can be expected for a European country without phosphate rock deposits, the overall P inflow to Denmark, in the form of mineral fertiliser, animal feed, food products, and assorted minor inflows of detergents and other chemicals, clearly outweighed P outflows (Figure 1). Total P inflows amounted to 56 Gg yr^{-1}, with outflows of 37 Gg yr^{-1}; animal feedstuffs (35 Gg yr^{-1}) and mineral fertiliser (12 Gg yr^{-1}) were the largest inflows, while the largest P outflow was in food products (33 Gg yr^{-1}). The amounts of P remaining in the system (18.2 Gg yr^{-1}) were found to accumulate mainly in agricultural soils (68% of total P accumulation, or 13 Gg yr^{-1}), in landfills in the waste management sector and construction via cement kilns (20% of total P accumulation, or 3.7 Gg yr^{-1}), and in consumption (8% of total annual P accumulation or 1.5 Gg yr $^{-1}$). The latter was likely due to home composting (0.36 Gg yr $^{-1}$) and fertiliser use on private and public non-agricultural lands; pet excreta also added to this accumulation. The amount of compost applied to agricultural land was found to me quite minor (0.11 Gg yr $^{-1}$), amounting to roughly one third of compost applied to gardens or public lands (0.36 Gg yr $^{-1}$). The organic waste flow shown was predominantly garden waste. The flow of household waste (MSW in the figure) was split into two flows (residual fraction and biowaste fraction) for visualisation; the biowaste fraction is the collectable biowaste (vegetable/animal food waste) generally not collected separately in Denmark at present (see section 4.1.1).

Within the country, the largest flows (by an order of magnitude) occurred within the agri-food system, i.e. between *agriculture* and *trade & industry*. The inflow of animal feed to agriculture (35 Gg yr $^{-1}$) surpassed the amount of mineral fertiliser (12 Gg yr $^{-1}$) here; in the opposite direction, export of food products, including both plant & animal products, amounted to 32 Gg yr^{-1}. Intensive animal husbandry, furthermore, meant that a total P flow of 23 Gg yr $^{-1}$ of animal manure was a main contributor to P accumulation in agricultural soils.

The total re-use of P in the system equalled 5.4 Gg yr^{-1}, which is approximately 10% of total P import flows (56 Gg yr $^{-1}$), counting current use of compost (0.47 Gg yr^{-1}), fish meal and slaughterhouse waste (in the recycling flow; 2.5 Gg yr^{-1}), and recovery from sewage sludge (2.4 Gg yr^{-1}).

Agricultural P flows were found to differ considerably between regions (Figure 2). These differences resulted from a crop production historically being concentrated in the east of the country (Zealand, Funen, and south-eastern Jutland), while livestock production has become more concentrated in the western and north-western part (south-western and northern Jutland) in the last 30-40 years, with specialisation in farming having driven this differentiation (Rubæk et al. 2013). The results of the MFA reflected this distinction. In region A (Figure 2a), mineral fertiliser was the largest inflow at 2.9 Gg yr $^{-1}$ (5.2 kg ha^{-1} yr $^{-1}$ or 44% of total P inflows) whereas animal feed clearly dominated inflows in region C (Figure 2c) at 8.9 Gg yr $^{-1}$ (19 kg ha^{-1} yr $^{-1}$). Agricultural output, as a consequence, had the highest proportion of plant products (and thus P removal through plant P uptake) in region A, while regions C and B produced a larger share of animal products (meat, eggs, and dairy). Concomitant with these differences in input and output (i.e. product) flows, the agricultural soil P budgets in the MFA were found to be distinctly positive in regions B and C (6.5 and 6.6 kg ha^{-1} yr^{-1}, or 3.1 Gg yr^{-1} and 10 Gg yr^{-1}, respectively), while region A showed a slight P deficit (-0.9 kg ha^{-1} yr-1 or -0.5 Gg yr^{-1}).

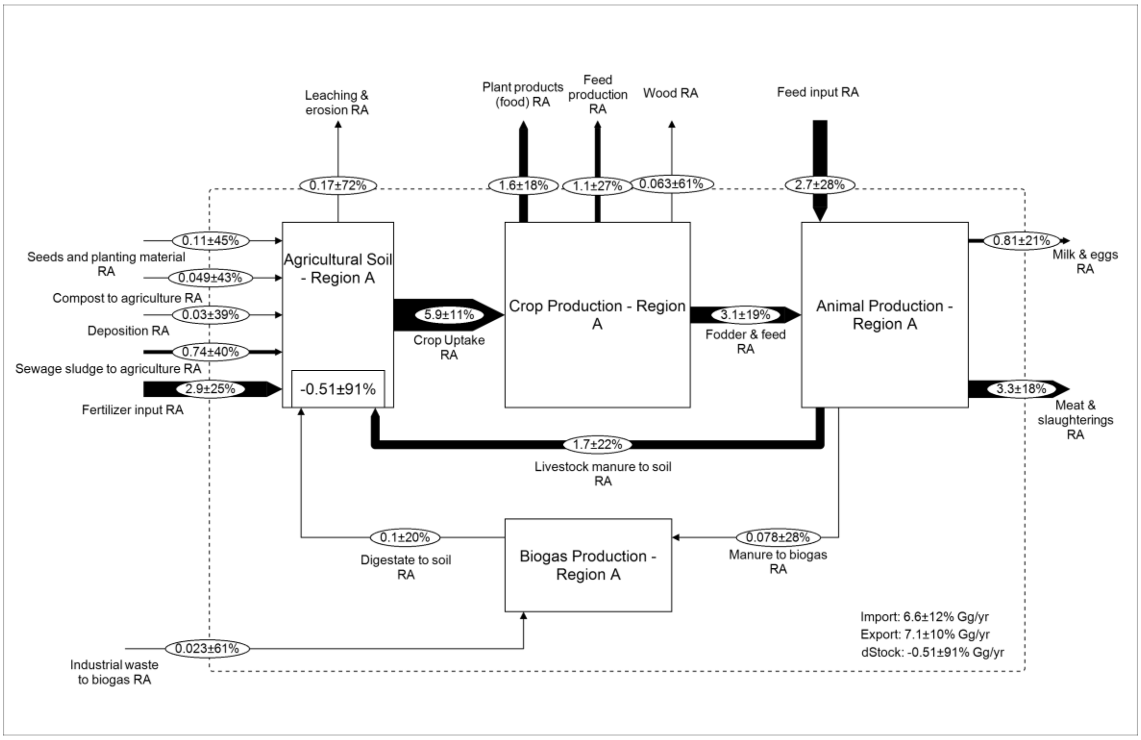

Figure 2a.

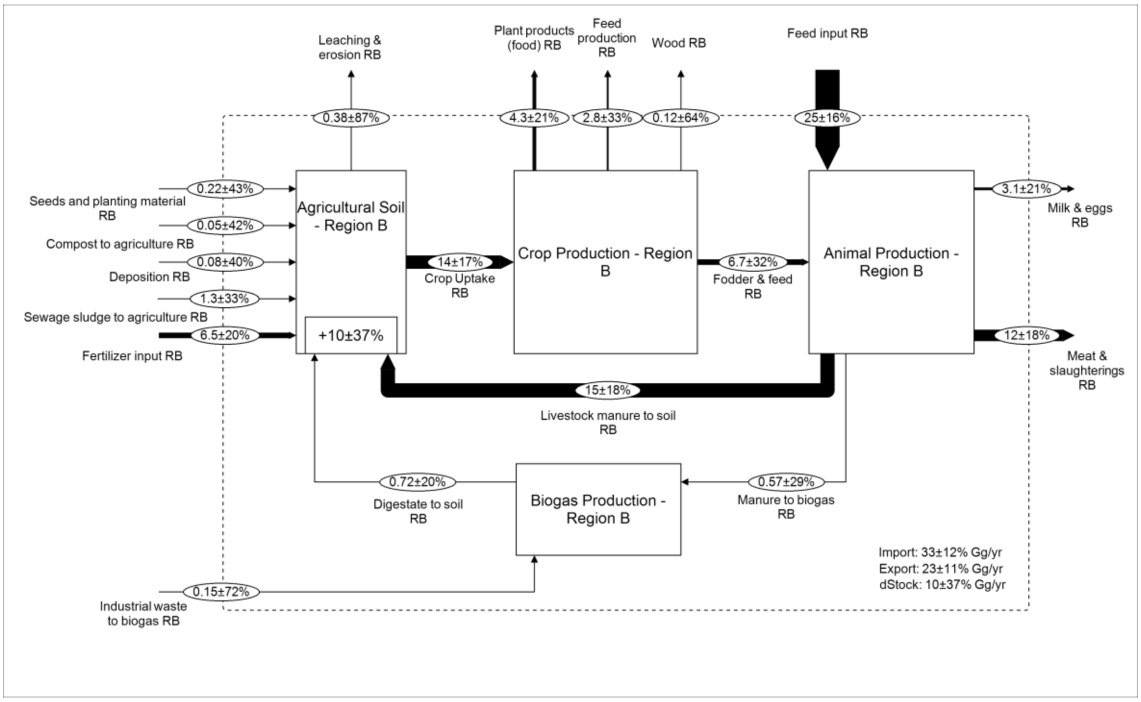

Figure 2b.

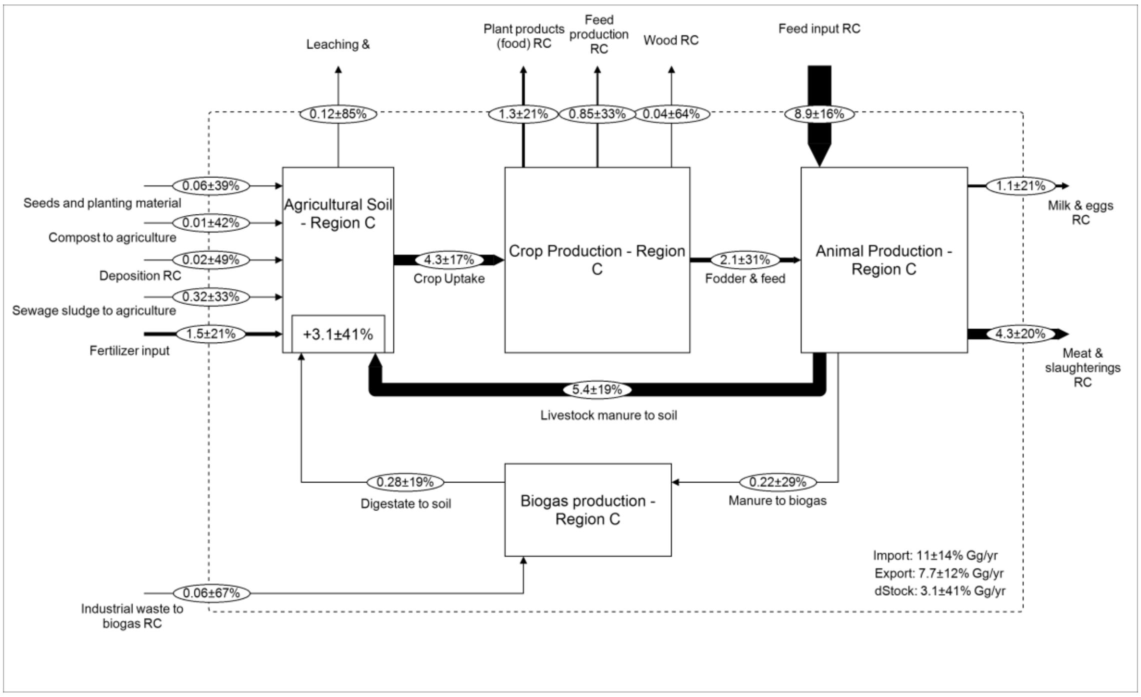

Figure 2c.

Figure 2. Regional-scale P flows [Gg yr^{-1}] for agriculture & forestry. Regions A, B, and C abbreviated as RA, RB, RC in flow names. Diagrams show reconciled values. Klinglmair et al. **II** shows a complete list of reconciled flow values and stock changes in absolute, per-hectare, and per-capita terms.

Regional differences in the P budgets found in the waste management subsystems (Figure 3) were due mainly to differences in population; industrial density also played a minor role. In waste management, the flows holding potential for P recovery were of particular interest; this applied, especially, to two flows: sewage sludge, and household biowaste as defined in section 4.1.1. Sewage sludge accounted for the largest inflow into the waste management subsystem, with approximately 0.8 kg cap^{-1} yr^{-1} in each region, or 2 Gg yr^{-1} in region A and B (with similar population sizes) and 0.5 Gg yr^{-1} in region C. In the case of the household waste, the MSW stream was divided into two flows for illustrative purposes to visualise the P potential in household biowaste. With the exception of garden waste (see the separate flows in Figure 1 and 3), this fraction was generally not collected separately in Denmark. The inflow of P in this fraction was, however, considerable (1.5 Gg yr^{-1}), amounting to roughly one third of the P inflows in sewage sludge (5 Gg yr^{-1}). The P contained in this fraction could be considered lost to P recovery (e.g. by digestion or composting). P in the waste management subsystem accumulated in incineration slag and ash, which was either landfilled or used in construction (i.e. in cement kilns) and thus permanently lost for recovery. The P flow in the residual fraction mainly stemmed from the P content in paper, cardboard, processed wood, and natural textiles. The recycling flow shown in Figure 3 was mostly composed of slaughterhouse waste (fish meal, processed meat and bone meal) for rendering or partly used as animal feed in the fur industry (Fødevarestyrelsen 2014).

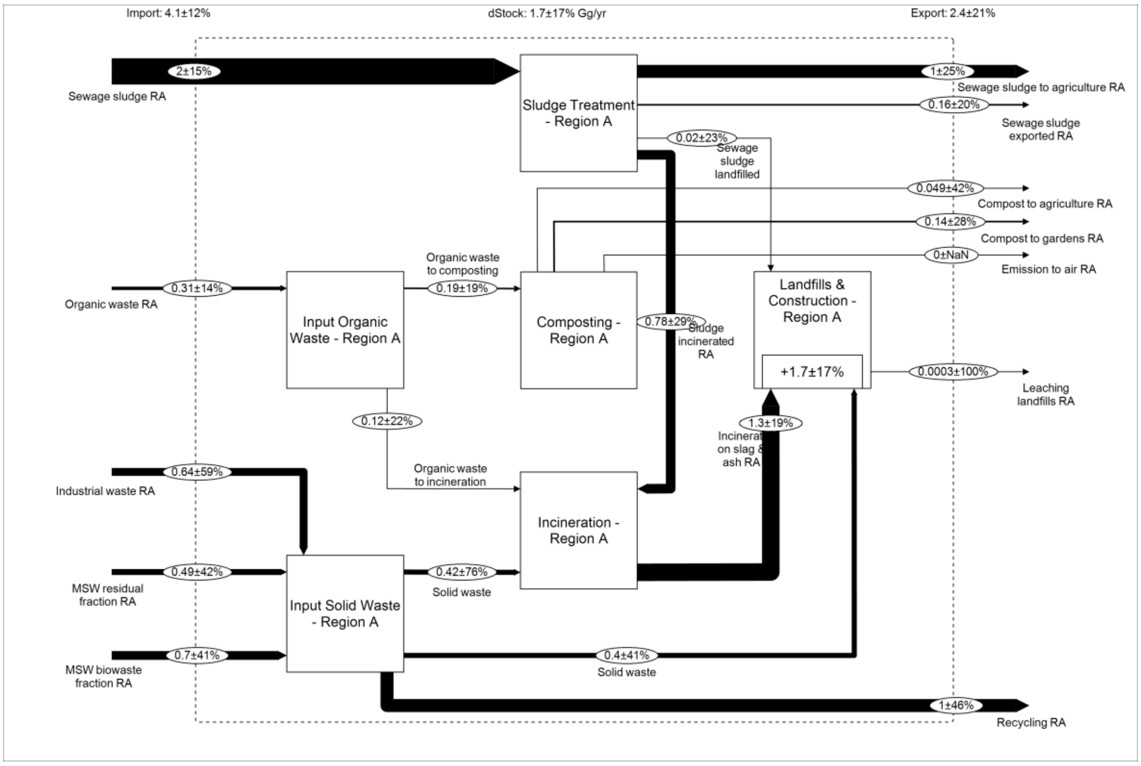

Figure 3a.

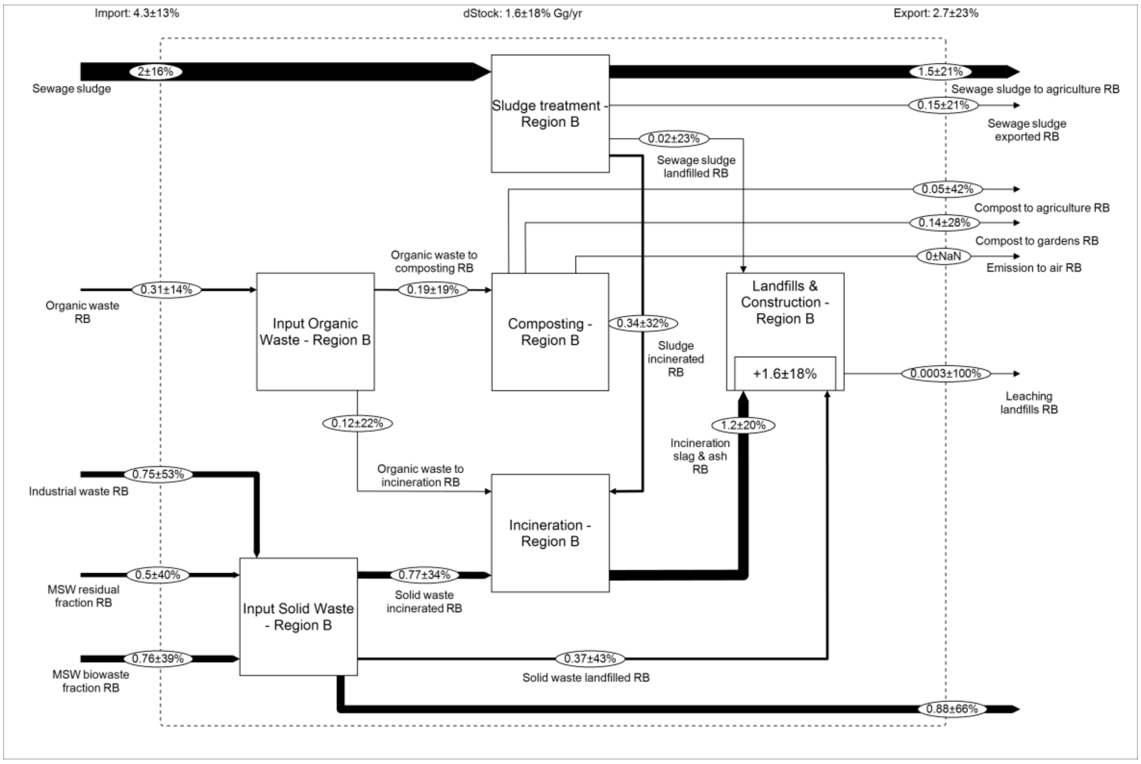
Import: 4.3±13%
dStock: 1.6±18% Gg/yr
Export: 2.7±23%
2±16%
Sewage sludge
Sludge treatment - Region B
1.5±21%
Sewage sludge to agriculture RB
0.15±21%
Sewage sludge exported RB
0.02±23%
Sewage sludge landfilled RB
0.05±42%
Compost to agriculture RB
0.14±28%
Compost to gardens RB
0±NaN
Emission to air RB
Organic waste to composting RB
0.31±14%
Organic waste RB
Input Organic Waste - Region B
0.19±19%
Composting - Region B
0.34±32%
Sludge incinerated RB
Landfills & Construction - Region B
+1.6±18%
0.0003±100%
Leaching landfills RB
0.12±22%
1.2±20%
Incineration slag & ash RB
Organic waste to incineration RB
0.75±53%
Industrial waste RB
Incineration - Region B
0.5±40%
MSW residual fraction RB
0.77±34%
Solid waste incinerated RB
Input Solid Waste - Region B
0.76±39%
MSW biowaste fraction RB
0.37±43%
Solid waste landfilled RB
0.88±66%

Figure 3b.

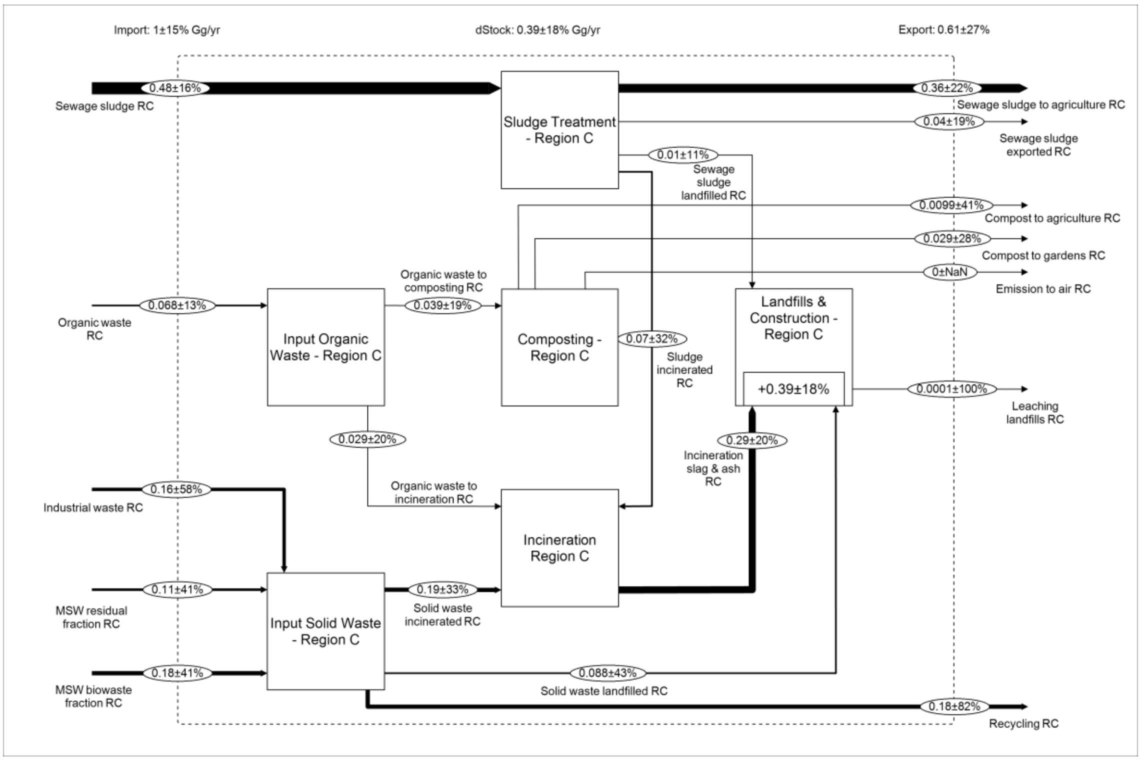

Figure 3c.

Figure 3. Regional-scale P flows [Gg yr^{-1}] for waste management. Regions A, B, and C abbreviated as RA, RB, RC in flow names. Diagrams show reconciled values. A complete list of reconciled flow values and stock changes in absolute and per-capita terms is shown in Klinglmair et al. **I**.

Counting the ratio of outputs of recovered P from compost and sewage sludge, to inputs of sludge and separately collected organic waste, the recovery rate of P in the waste system for use on agricultural land was 47% in region A and 67% in regions B and C; this ratio was lower in region A due to sludge incineration being concentrated in the Copenhagen area. It is to be noted, however, that this is a theoretical upper limit set by total amounts of P; a recycling rate of 100% is thermodynamically impossible, and 100% sorting a practical impossibility. Moreover, a 1:1 substitution between fertilisers is unlikely due to differences in P bio-availability to plants. Most recovered P comes from sewage sludge at present. About 48% of the P in sludge were applied to land; the total quantity of P in sewage sludge produced (5 Gg P yr^{-1}) corresponded to 42% of the mineral fertiliser P imported in 2011. The household biowaste fraction in MSW, which holds about 60% of the P in MSW but is currently not collected (and thus made recoverable) separately in Denmark, was found to hold a total theoretical recovery potential of 1.6

Gg P yr^{-1} corresponding to approx. 14% of mineral fertiliser P imported in 2011. The total unrecovered quantities of P (in sludge and biowaste) thus amounted to 4.2 Gg yr^{-1}, or approximately 35% of P in mineral fertilisers imported.

The most important hindrance to further closing the P loop on the country scale in Denmark is distribution; as Figures 2 and 3 make clear, the differences in regional P households were considerable. In the east of the country (region A), the agricultural system is dominated by crop production with a slight overall P deficit in agricultural soils, and the concomitantly large amounts of P applied in the form of mineral fertilisers. Region C, in contrast, showed the largest surplus of P in agriculture, with livestock production resulting in the highest relative inflow of animal feed, and output of manure at rates exceeding plant removal of P. In the instance of region A, the country's largest urban agglomeration (Copenhagen) and largest relative amounts of sewage sludge produced coinciding with high fertiliser application rates suggested a high potential of substitution through increased recovery of P from sludge. Sludge incineration is likewise concentrated in region A; for volume reduction and ease of transport, the separate storage of sludge mono-incineration ash has been suggested as an option for later treatment and nutrient recovery (e.g. Lederer & Rechberger 2010; Miljøstyrelsen 2013). For the case of manure, distribution (transport) is a more crucial issue due to the amounts (see Figure 2) and long transport distances involved; at present, the output stays quite local (Rubæk et al. 2013; Sørensen et al. 2013). Biogas production did not yet contribute at a significant scale to redistribution of recovered P (see Figure 2), but provided the implementation of the right technologies (e.g. mechanical separation) in conjunction with biogas plants this may play a more important part in a relocation of manure P.

4.1.3 The Danish P budget in a European context

Ott & Rechberger (2012) established a detailed P balance for the EU-15. Aiming at better comparability of national P budgets across the EU, while more recently van Dijk et al. (2015) presented P balances for the EU-27 and its member states. Going from the supra-national or continental to the country scale, the Danish situation offers itself to several comparisons with recent research.

The overall ratio of P out- to inflows in Denmark (0.66, implying accumulation and losses of 34%) was close to the figure for the EU-27 at 0.61 (van Dijk et al. 2015; with the base year 2005). In other respects, too, Denmark can be said to resemble the typical European situation. It was found to be a typical case in its dependence on primary P imports of mineral fertiliser and animal feed; especially in the relative predominance of feed imports, Denmark closely resembled the Swiss case (Binder et al. 2009; Lamprecht et al. 2011). Utilisation rates of sewage sludge (or derived products) on land (48%) were comparable to France (48%; Senthilkumar et al. 2014), while lower than in the UK (71%; Cooper & Carliell-Marquet 2013), although claims of over 70% utilisation rate for Denmark also exist (Miljøstyrelsen 2013). Conversely, these utilisation rates point to as-yet unused quantities of P to be recovered. The French case (Senthilkumar et al. 2014) is instructive here as it highlights the amount of P in household biowaste (vegetable and animal food waste): 43% of P in MSW were recovered from this fraction. The corresponding Danish amount, currently not separately collected and thus lost to P recovery, held a theoretical potential of 60% of the total P in MSW (while a realistic number will be lower due to incomplete separation and collection). These observations are mirrored on the European scale: the largest losses in waste flows (liquid and solid) were found by van Dijk et al. (2015) to be in the form of sewage sludge and food waste in the EU-27.

4.2 Material flow analysis: the influence of modelling choices

4.2.1 System layout and definitions

The two MFA studies compared (Klinglmair et al. **I**; Zoboli et al. 2015) in Klinglmair et al. **II** share a similar geographical scope, in that they established P balances for European countries of comparable size. They differed in focus, since the Austrian case did not aim to integrate regional subdivisions, but established a 22-year timeline of the system to show variation in the P budgets from 1990 to 2011. For the purpose of this comparison, the year 2011 was considered in both studies. While the Danish model comprised 50 processes and a total of 166 flows (including regional subdivisions), with 12 “virtual” processes to split or aggregate flows for visualisation, the Austrian model comprised 56 processes and 122 flows (10 “virtual” processes). Both systems were overdetermined; more variables (flows and stock changes) were known than would be necessary to solve the balance equations, with 7 unknowns and 41 independent balance equations in the Danish case and 16 unknowns (44 balance equations) in the Austrian case. Unknown flows were calculated in STAN as part of the data reconciliation step.

The main processes—agriculture, soils, forestry, crop production, wastewater treatment, waste management, and industry and trade—and flows were found to conform to each other as could be expected. The hydrosphere was the only exception, as in the Austrian case it was placed inside the system boundary due to an interest in the influence of anthropogenic P emissions on the load dynamics in the river Danube; in the Danish case, such emissions were merely counted as losses. Slight differences in the process definitions were identified with respect to the location of sub-processes in either industry or agriculture, with some ambiguity in both systems regarding slaughterings. The individual instances are reported in Klinglmair et al. **II**.

The layout of an MFA system is closely linked to the datasets used or available. Figure 4 shows a side-by-side overview of the types of data sources used; in both cases, it can be seen that technical reports and statistical offices made up approximately 80% of the data sources. A closer comparison of sources (see the supplementary data for Klinglmair et al. **II**) showed that differences in data availability could largely be excluded as a reason for the differences in the system layouts (with the exception of minor flows in subsystems; see Klinglmair et al. **II**). If differences in system layout are not clearly justified by an MFA's goal and scope definition, and given comparable data availability, such differences in subsystems can be reasonably attributed to the MFA practitioner's judgement. The layout of both studies reflected what may be called an existing (or at least emerging) consensus on system boundaries, processes and flows to be included in regional P balances (cf. Seyhan 2009; Scholz et al. 2014; van Dijk et al. 2015), also due to the same key processes and flows in human P use.

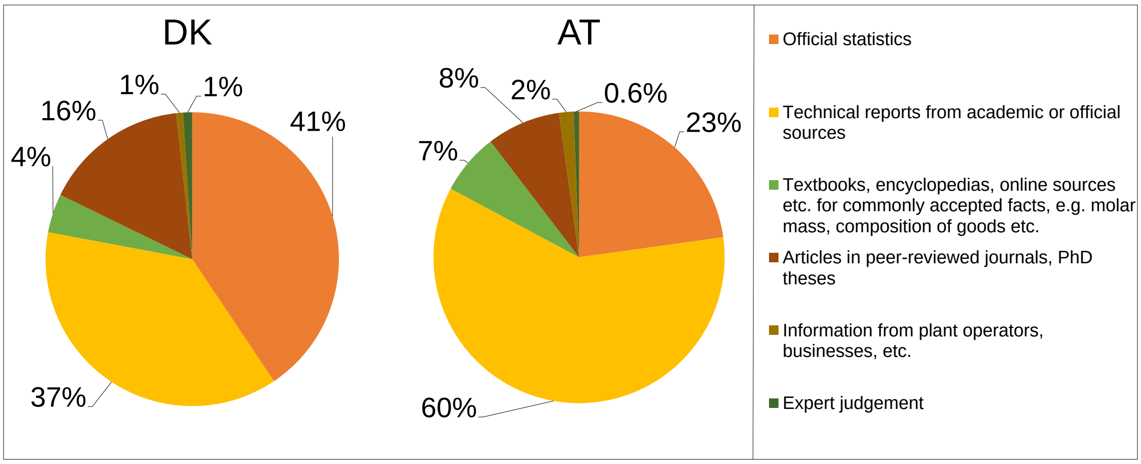

Figure 4. Data sources used in the MFA systems for Denmark (DK) and Austria (AT) compared, counting each reference for each flow (DK: 399 citations; AT: 500 citations total). A complete list of flows, flow values, and references is provided in the supplementary information for Klinglmair et al. **II**.

An additional aspect worth a mention is the spatial resolution of an MFA. An increased spatial resolution such as in the Danish case, multiplying regional-level of flows by the number of regions considered, engenders higher requirements on data consistency. Although the Austrian model exhibited a higher number of types of flows, this multiplication of regional flows in the Danish case led to higher overall number of connected flows, resulting in a higher risk of data conflicts.

4.2.2 Uncertainty assessment

There exists considerable freedom for the MFA practitioner to choose the uncertainty assessment procedure for a given MFA study. In the two cases compared in Klinglmair et al. **II**, different approaches were chosen. Klinglmair et al. **I** drew upon the concept data characterisation proposed by Hedbrant & Sörme (2001) and its adaptation by Laner et al. (2015b), in which each data source is categorized and linked to an uncertainty level, based on which a symmetric uncertainty range (as CV) for input in STAN is calculated (see section 3.2 of this thesis). The uncertainty characterisation in the Austrian study was based on an adaptation of a so-called "pedigree matrix" proposed by Weidema & Wesnæs (1996) for assessing data quality on life cycle inventory (LCI) data. The approach assigns to each datum a discrete score based on a set of quality indicators; the uncertainty interval (CV) is based on the combination of the various indicator scores for each datum. In both studies, the uncertainty range of a P flow value is the combination of the uncertainties of the mass flow and its P concentration. Figure 5 shows the relative uncertainties for each flow before and after data reconciliation in both systems. It is to be noted that data reconciliation in STAN always results in a reduction of uncertainty throughout the system. Where the figure shows an increase in relative uncertainty, this is due to a decrease in the reconciled flow value. The uncertainty assessment method used (section 3.2) in the Danish case (with one overall indicator each for the uncertainty of P concentration and mass flow) led to the "stepwise" appearance of the entered intervals, whereas in the Austrian study, the combination of several indicators (with the uncertainty for P concentration and mass flow each based on several different scores) resulted in a "smoother" line i.e. a greater number of different uncertainty intervals.

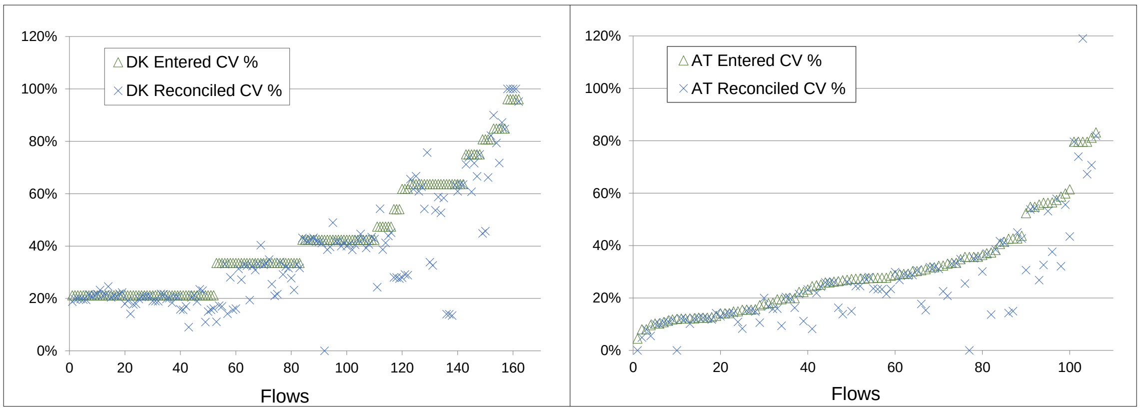

Figure 5. Relative uncertainty (coefficient of variation, CV) for the MFAs of Denmark (DK) and Austria (AT), as entered in STAN and after data reconciliation. Each point on the x-axis represents a flow, with flows numbered and sorted from lowest to highest relative uncertainty as entered. Flows calculated in STAN (i.e. no flow values entered) are not shown here.

An examination of the underlying data material in both studies revealed a similar composition of data sources used (Figure 4), yet Figure 5 highlights the results of the different approaches to uncertainty characterisation. In both cases, the approach chosen followed a well-defined procedure, yet the choice in assigning scores or levels, and associated uncertainty ranges, to data sources remains, to some degree, based on the modeller's best judgement. This revealed a difficulty in comparing data quality between MFA studies, e.g. for the user drawing upon an MFA as data source itself. Flows of the same composition, based on the same data source or type of data source, may be found in several studies, yet their associated uncertainty intervals may imply large widely different data qualities. In the case study at hand, the majority of flows (66%) were assigned relative standard deviations above 30% in the Danish MFA (with none below 20%), while in the Austrian MFA, the relative majority of flows (52%) showed relative standard deviations below 30% (see also Figure 5 in Klinglmair et al. **III**). In short, the study by Klinglmair et al. (**I**) exhibited less optimism in its judgement of data quality. Comparing uncertainty characterisation between the two models exemplified that—lacking a consensus on characterising uncertainty in MFA—uncertainty estimates are not directly comparable between different MFAs, regardless of the similarity of the data material used. Rather, these uncertainties can reasonably only be evaluated relative to the uncertainties of other flows found in the same model.

4.2.3 Data consistency and data reconciliation

Apart from choice of method (the effect of which has been shown at the example in Klinglmair et al. **II**, and section 4.2.2. of this thesis) characterisation of uncertainties in an MFA reflects the belief of the MFA practitioner in the quality of the input data. An overly optimistic estimation of data quality, i.e. assigning low uncertainties to flow values, can have a detrimental effect on the mass balancing in the model. In such a case, the extent of data reconciliation will be overly strong in some cases and result in the reconciled value lying outside the uncertainty range entered in STAN. The reconciled value then lies too far to the side of the normal distribution, becoming highly improbable.

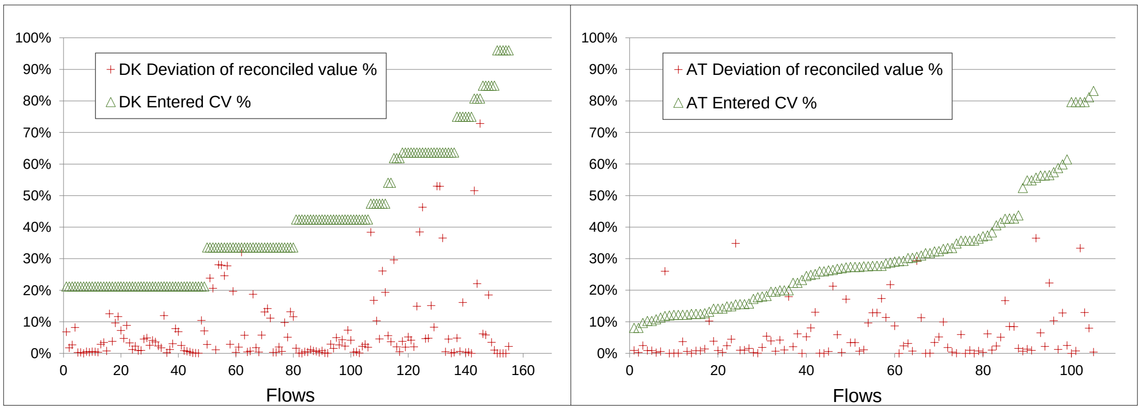

Figure 6. Uncertainty ranges (CV, in %) as entered in STAN, and relative deviation of reconciled flow values from entered flow values (in % of the entered flow value). Each point on the x-axis represents a flow, with flows numbered and sorted from lowest to highest relative uncertainty as entered. Flows calculated in STAN (i.e. no flow values entered) are not shown here.

Figure 6 compares the relative uncertainties to the deviations in flow values resulting from data reconciliation. If the deviation of the reconciled flow value from the entered value is too high, reconciliation was too strong. In such a case, the uncertainty estimate by the modeller was either overly optimistic, or, especially if this issue is widespread or persistent even with more pessimistic uncertainty estimates, this necessitates a critical evaluation of the model equations (the system layout) or the data material used. If, conversely, the modeller's uncertainty estimates are much larger than the changes in flow values resulting from data reconciliation, such a pessimistic assessment of uncertainty may lead to an unnecessary loss of model precision.

The extent of data reconciliation can, furthermore, serve as an indicator for the consistency of an MFA model, i.e. the fit of the data given the constraints of the mass balance equations and the defined uncertainty ranges. Figure 4 shows the mean relative deviation of reconciled flow values from the values entered in STAN on a per-process basis. The average extent to which reconciled data deviate from entered data provides a useful measure to identify the weak spots in an MFA model, and to assess the quality and data of the system. The mean of all deviations gives a measure of the extent of data reconciliation or 'fit' of the model and data (Eq. 1; see also Zoboli et al. 2015), with $\overline{D}$ being the mean relative deviation of the reconciled data from the entered values, m the total number of flows, x_n the entered value for flow n, and $\hat{x}_n$ the reconciled value for flow n.

$$\overline{D} = \frac{\sum_{n=1}^{m} \left| \frac{x_n - \hat{x}_n}{x_n} \right| * 100}{m} \qquad (1)$$

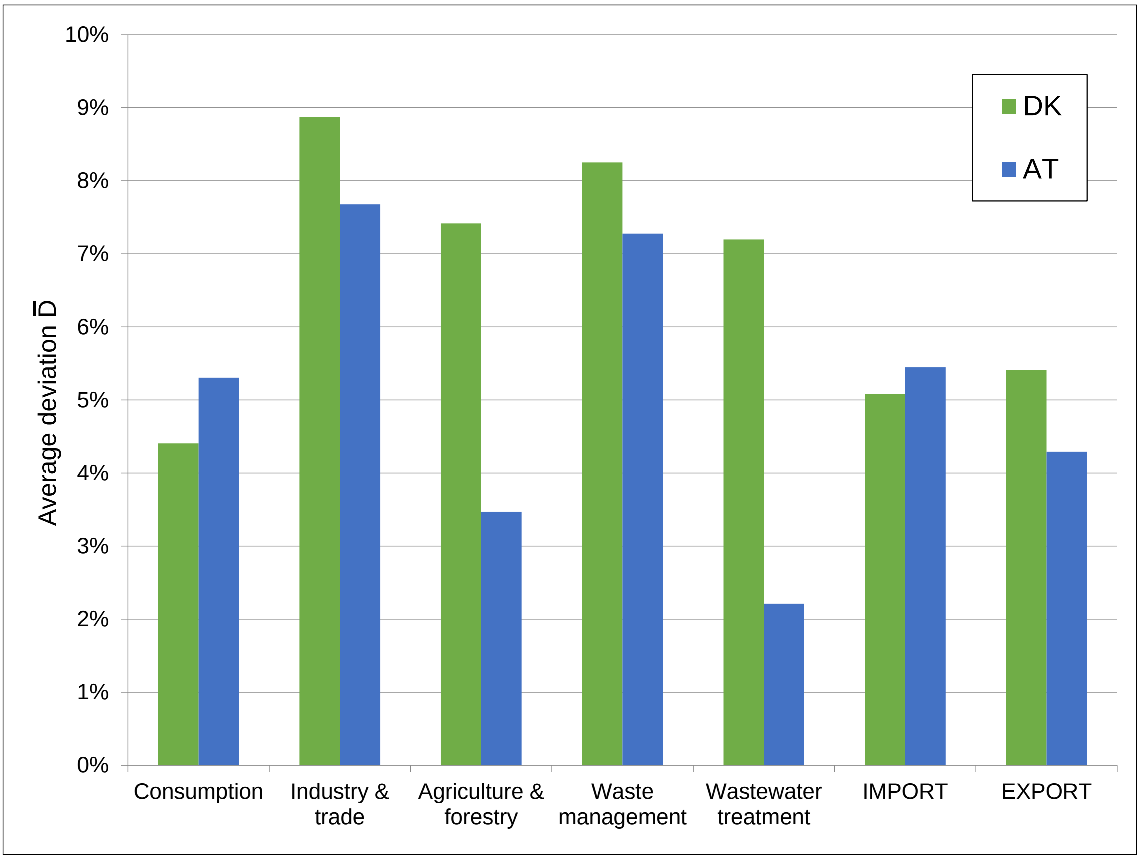

Figure 7. Average relative deviation $\overline{D}$ of reconciled flow values from entered flow values. Each column represents all flows connected to, or belonging to a subsystem in, a process.

In the Danish model, the average deviation of reconciled from entered flow values was 8.6% over all flows, with data reconciliation weakest (i.e. changes in reconciled values smallest) around the *consumption* process (4.4% average deviation), and strongest around the *industry & trade* process (8.9%). The Austrian model could be said to be more consistent overall, with an average deviation of reconciled flows of 4.9%, with reconciliation weakest for flows connected to and contained in *wastewater treatment* (2.2%), and strongest for *industry & trade* (7.7%). This difference could be partly attributed to a higher degree of over determination in the Danish MFA, and thus a higher probability of data conflicts. In the case of the Danish model, specifying those national-level flows, which were aggregates of several regional flows, and were based on the same data sources, as unknowns (and calculating them in STAN), would have resulted in higher but more realistic uncertainties given the available information. It is to be noted, however, that generally it is not advisable to forego entering additional available data into an MFA model to avoid data conflicts.

To avoid both on overly optimistic and pessimistic estimation of uncertainties in MFA, and the resulting misrepresentation of model precision in the outcomes, an iterative approach to uncertainty characterisation (see also Laner et al. 2014) is advisable; i.e., it is simply the responsibility of the modeller to iterate and check the uncertainties entered against the constraints imposed by the balance equations (as shown in Figure 6) to ensure that flow values remain within specified ranges.

4.3 Optimization of P resource flows in Denmark

4.3.1 Challenges

The MFA carried out in Klinglmair et al. (**I**) identified a set of flows holding additional potential for recovering P and possibly substituting significant amounts of otherwise imported mineral P. While the application of manure was quite local due to the bulk of the material flow, the unused quantities of sewage sludge and the currently lost (due to practically no separate collection) flow of household biowaste held the most salient potentials. In the adaptation of the MFA, no transport of manure or raw sewage sludge was assumed; only composted organic household waste and P recovered from sludge mono-incineration ash were assumed to be transported.

However, the utility of these flows as fertilisers depends on the availability of the contained P to plants. Phosphate ions are taken up from the soil solution by plants, while a part of the P applied becomes adsorbed to soil solids, becoming part of the plant-available P pool as soluble P is removed by plant uptake. Applied P can remain in the soil for years and gradually add P to the soil solution (e.g. Syers et al.

2008; Ylivainio & Turtola 2013). Other influences on soil P availability are e.g. soil pH, soil type, climate, and pest control. Moreover, the differences in P flows between the Danish regions identified in the MFA strongly suggested transport of at least part of the recovered P between regions for the most efficient application.

In this work, a strongly simplified approach was taken, using typical or average values for Europe (see Klinglmair et al. **III**), and 3 annual time steps.

4.3.2 Improvement potentials by redistribution of material flows

The MFA model from Klinglmair et al. **I** was adapted as shown (for one region) in Figure 8. An MFA diagram showing the national-level layer, linking the regional subsystems, can be found in Klinglmair et al. **III**. As the figure shows, processes and flows not pertaining directly to P use in the agri-food system and its recovery in waste management and wastewater treatment were omitted and other flows (e.g. animal products, comprising meat & slaughterings, milk, and eggs) combined. In total, the adapted model comprised 28 processes and 75 flows including all 3 regional subsystems. Exchanges between regions were routed through a common pool on the national-level layer. A complete set of MFA diagrams, showing all regions and time steps, is provided in the supplementary material for Klinglmair et al. **III**.

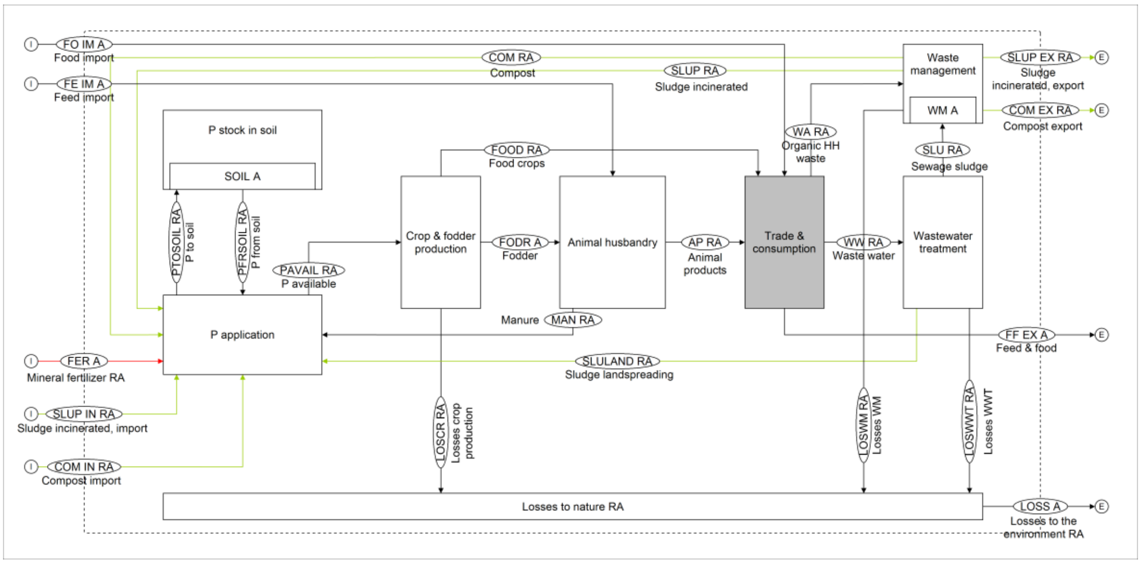

Figure 8. Qualitative representation of MFA model (in STAN) on the regional level, also forming the superstructure of the optimization model (flows within the subsystem for region A shown here). The decision variables concern the activity levels of the processes (boxes), with only the activity level for trade and consumption (grey) given. Flows subject to change by the model are marked green. Imports of mineral fertiliser (to be minimised) shown in red.

Figure 9 shows the decline of demand for mineral fertiliser application over time as a result of soil P from earlier additions becoming gradually plant available over time. Likewise, net additions to soil stocks gradually declined or remained stable across regions and time steps. Within the time steps shown, total mineral P addition decreased from 10.2 Gg yr^{-1} after 1 year to 2.2 Gg yr^{-1}. Without including composted organic household waste in the model but still including ash from sewage sludge incineration, the corresponding value rises to 8.2 Gg yr^{-1}. A test of continuing the timeline to 10 years resulted in an eventual evening out of both net additions to soil stocks, and a stabilization of mineral P imports at 1-2 Gg yr^{-1} (see Supplementary Information S4 in Klinglmair et al. **III**) The static MFA in Klinglmair et al. **I** showed a maximum potential amount of P in unused sewage sludge and organic household waste to substitute approximately 35% of mineral fertiliser. Adapting the MFA to account for the dynamic aspects of transport and P plant availability over time, however, suggested an eventual substitution of about 82% considering a time period of three years.

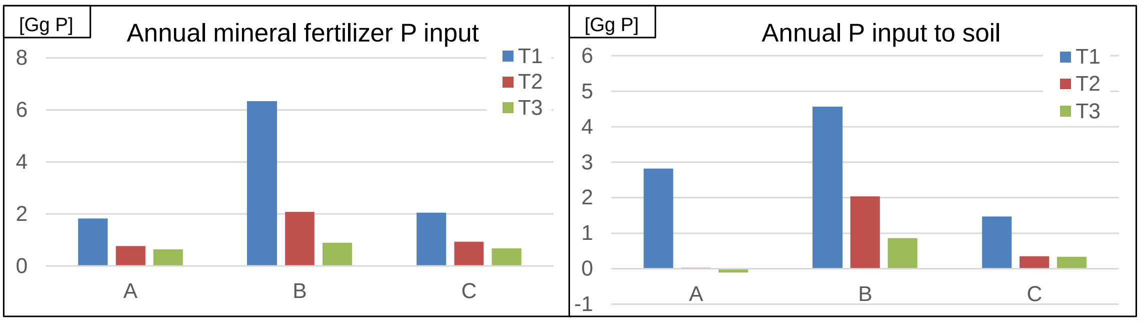

Figure 9 Annual mineral fertiliser input [Gg P] and net annual P addition to soil stocks [Gg P] per time step (T1 to 3) and region (A to C).

In Figure 10, the import, export, and local use and size of the recovered P flows (sludge and compost, respectively), as determined via the optimization model, are shown (imports to, amounts generated within, exports from, and local re-use (application on land) within a region). In the case of sludge, only P recovered from incineration ash was allowed to be transported; for local re-use, sludge generated within the region could be either incinerated or applied directly on land in the region. All sewage sludge generated was potentially available for recovery in the model. As the figure shows, region B (Mid-Jutland and South Denmark combined) exhibited the greatest use of recovered P (as shown by imports of recovered P and local re-use); this is due to the region's large overall population size and agricultural area. Region C, in contrast, was a net exporter of recovered P, while region A showed a more balanced picture of in- and outflows.

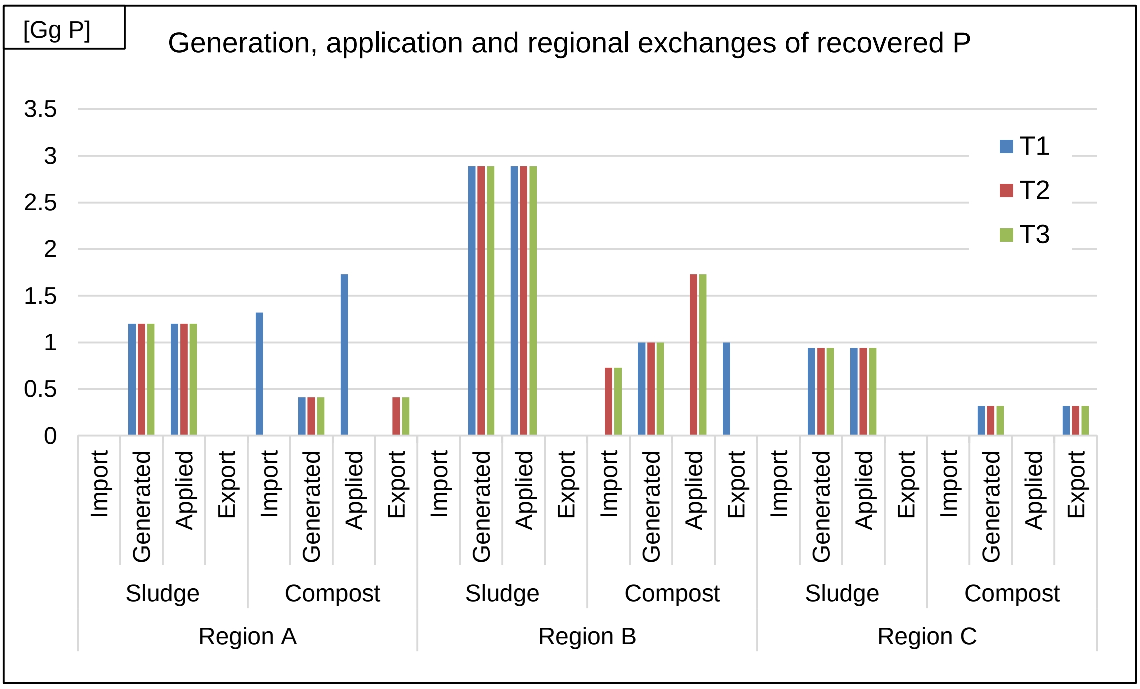

Figure 10. Generation and application of P recovered within regions, and inter-regional exchanges of recovered P, considering P recovery from direct sewage sludge land application or incineration of sludge and land application, and land application of recovered P via composted organic household waste, as determined in the optimisation model. Zero values indicate no import, export, or local re-use of P recovered within the region, respectively.

Apart from the flows of transportable recovered-P products, the model also allowed for direct application of manure and sewage sludge locally, i.e. within the respective region. The model considered all sewage sludge generated, including the amounts of sludge already used, and amounts currently not applied on land. The highest imports, of 1.5 Gg P over the entire timeline, and a local use of 10.7 Gg P from composting and direct application of sludge, took place in region B. In contrast, the high local amounts of manure in region C resulted in a low regional demand and high potential exports of recovered P. If, as in Figure 10, the additional P flow from composted organic household waste is included in the model, sludge is shown to stay within the region to be applied directly, with imbalances (challenges in most efficient P application) in the system met by comparatively minor exchanges of P recovered from composted organic household waste. The exchange and local re-use of incinerated sludge becomes desirable in the model if compost is not an option (see Klinglmair et al. **III**). The Supplementary Information S3 in Klinglmair et al. **III** shows all flows, regions, and time steps as MFA diagrams, including losses to the environment and via accumulation in waste management.

If compost was not included in the model, the relevant amounts of P were assumed to be lost as incineration residues to the waste management process. Otherwise, near-complete utilization of household biowaste was assumed. Other losses to the environment (from agriculture and wastewater treatment) were determined by the transfer coefficients based on the MFA in Klinglmair et al. **I**. Since not all the processes of that MFA were included in the adapted MFA in Klinglmair et al. **III**, the lower losses of 0.8 Gg yr^{-1} (compared to 1.3 Gg yr^{-1} in Klinglmair et al. **I**) shown by the optimization model are plausible. As becomes clear from this optimization model, and the results of the MFA shown in section 4.1.2, transport of P-containing wastes or products from P recovery play a prominent role in improving P management across Denmark. Due to the sizes of the material (bulk) flows involved, manure was not considered transportable in this optimization model; in fact, at present, manure is applied locally (see section 4.1.2) and further handling and volume reduction through e.g. anaerobic digestion in biogas plants, concerns negligible amounts at present, compared to the overall amounts of manure. As regards the transport of sludge incineration product, P "banks" have been suggested in several publications (Miljøstyrelsen 2013; Lamprecht et al. 2011; Egle, Rechberger, et al. 2014) to allow for storage of mono-incineration ash and later treatment for P recovery. Since the flows in the optimization model were not uniform in their direction and location, such buffers for intermediate storage, of temporarily unneeded quantities of material suggest themselves in the case of P management demonstrated here.

5 Conclusions and outlook

5.1 Conclusions

The integration of a comprehensive country-scale MFA of anthropogenic P flows and stocks in Denmark with a regional consideration in the agriculture and waste management subsystems showed that an end to the reliance on primary P imports is unlikely. Yet the study points to significant improvement potentials in closing the P cycle in the country. A resolution higher than the country scale was shown to enable more meaningful statements about the Danish P households. This work could quantify the most relevant flows of those quantities of secondary P (recoverable P fertiliser sources) on a regional, sub-national basis, linking them to the complete P household of the Danish anthroposphere. Significant amounts of P, although potentially recoverable, are currently lost in the waste management system (unused sewage sludge, household biowaste) or in agriculture (by manure application surpassing the crop demand resulting in accumulation of P in soils). The currently unused quantities of P in organic household waste not collected separately, and sewage sludge currently not applied on land, correspond to approximately 35% of annual mineral P imports in fertilisers. To a large extent, increasing the resource efficiency for P is a transport, and policy, problem. In addition to transport, however, the utility of recovered P in its ability to substitute mineral fertiliser is a crucial element. Adapting the static MFA "snapshot" to reflect these dynamic elements, it was demonstrated how P build-up in soil and gradual uptake by plants, and transport of certain recovered P products between regions, changed the picture of the static MFA, resulting in a gradual decrease of mineral P imports and net additions to the soil P stocks, without reaching zero in either case; an eventual potential for substituting more than 80% of mineral P imports was shown. At the country scale, MFA forms a useful quantitative groundwork for any resource conservation effort or policy, and may be drawn upon as a source for information by a reader in the scientific or policy fields. In the work presented in this thesis, a comparison of two MFA studies of similar P MFAs (for Denmark and Austria) showed the influence of an MFA practitioner's choices on model outcomes. It was demonstrated that the degree of arbitrariness in designing an MFA, while not influencing the main conclusions to be drawn, is especially relevant regarding the judgement and reporting of data sources and their quality but hinders the direct comparison of uncertainties between studies. The assessment of uncertainties in country-scale MFA is to some extent arbitrary; it reflects the practitioner's belief in his model's consistency and quality. The necessary extent of reconciliation of data conflicts, moreover, provides a measure of an MFA's consistency as a proxy for quality. Metadata matter; transparency by the MFA practitioner is a necessity for MFA to serve as a trustworthy source of information.

5.2 Outlook

The findings in this thesis, and the experience gathered leading up to its completion, suggest several issues lending themselves to future research work.

Without question, increasing the spatial resolution of MFAs of resource flows and stocks can greatly increase the informative value of such studies, for P as well as for other substances. In this study, the imbalances in the resource household for P, even in a geographically relatively homogeneous country such as Denmark, were clearly shown at the example of three distinct Danish regions. Further increasing the resolution e.g. to the communal/municipal level can yield further insight and open possibilities to integrate resource management on the country scale with measures taken by local authorities or waste companies. GIS-based approaches could be useful to establish cadastres of resource stocks, such as P-containing wastes.

Inventorying a defined set of critical resources according to standardised templates, along with regular updates of key figures, could provide a useful database in which to follow the quantities, pathways, and accumulation or depletion or resources in a given area (e.g. a country). Closely related to this topic, the heterogeneity of the data material drawn upon in country-scale resource balances. Absent a clear consensus at the time this thesis was written, a transparent, reproducible, and widely accepted approach to appraise the quality of MFA data continues to be a topic for research.

Lastly, and specifically connected to the topic of P management, the value of various products of P recovery from waste or manure streams as substitutes for primary P imports, i.e. detailed knowledge of their behaviour in various soils and availability to plants, remains a topic of ongoing research beyond the scope of this thesis. Likewise, the economics of a more comprehensive recovery and efficient redistribution of P, and the technologies involved, is an area of research crucial to future P management schemes.

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The Department of Environmental Engineering (DTU Environment) conducts science-based engineering research within six sections: Water Resources Engineering, Water Technology, Urban Water Systems, Residual Resource Engineering, Environmental Chemistry and Atmospheric Environment.

The department dates back to 1865, when Ludvig August Colding, the founder of the department, gave the first lecture on sanitary engineering as response to the cholera epidemics in Copenhagen in the late 1800s.

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