Phosphorus

Flows to Swedish Food Chain, Fertilizer Value, Effect on Mycorrhiza and Environmental Impact of Reuse

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Cover: Mining of phosphorus in Morocco
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Abstract

Phosphorus is an essential element for plants, animals and humans and is also a scarce resource as a raw material for fertilizer production. The flows of phosphorus to and from Swedish agriculture and food chain was investigated with a material flow analysis (MFA). The fertilizer value of recycled phosphorus in chemically precipitated sewage sludge, biological sludge, mineral fertilizer and ash was investigated in a three-year field experiment. The impact of different phosphorus fertilizers on arbuscular mycorrhiza (AM) was studied in field and pot experiments, and the environmental impact of different phosphorus fertilizers was evaluated by Life Cycle Assessment (LCA).

The results showed that Sweden has a positive phosphorus balance, but that phosphorus is concentrated on farms with more than one animal unit per hectare. Furthermore, the calculations were made on recent production and if Sweden increases the level of self-sufficiency, more phosphorus would be needed in the system.

In a long-term perspective, the form of phosphorus fertilizer used seems to be less important than maintaining soil phosphorus content to ensure phosphorus delivery to the crop when conditions in terms of moisture, temperature *etc*. are optimal. Easily soluble phosphorus in mineral fertilizer can be essential in a short-term perspective under dry conditions, or on phosphorus-deficient soils.

This thesis shows that with balanced phosphorus fertilization, it is possible to achieve good yield without compromising the possibilities for AM fungi to colonize roots.

Phosphorus recovery from ash is costly in terms of both energy and carbon dioxide emissions. The most efficient option for sewage sludge is to use it directly on farmland, but associated cadmium addition to soils can be a problem. The cadmium imports with food and feed to Sweden are currently unknown. Nitrogen is a valuable nutrient and is energy-demanding to replace, so fertilization systems that can keep nitrogen in usable form for production are preferable.

It was concluded that the energy and carbon dioxide bound during photosynthesis should be assigned to agricultural production and not the product user, *e.g*. a sludge incineration facility. Production of bioenergy should include all products that contain energy bound by photosynthesis, not only products used for fuel and heating.

Keywords: Arbuscular mycorrhiza, cadmium, fertilizer, Life Cycle Assessment (LCA), manure, Material Flow Analysis (MFA), phosphorus, sewage sludge.

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Dedication

To everyone that is dependent on phosphorus

Det visslar en bondtrygg stare, det skymtar en räv över mon,
det hoppar en jagad hare - jag trampar en mask med skon.
Jag blev väckt av liv som larmar- jag har vaknat i vårens armar,
och fast hungrig jag strängat min lyra bland alarnas droppande blom,
är jag rusig av våren yra, där jag går i min fattigdom…

Dan Andersson,

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 Linderholm, K., Mattsson J.E. & Tillman, A.M. (2012). Phosphorus flows to and from Swedish agriculture and food. *AMBIO* (Accepted 26 April 2012).

II Linderholm, K., Palm, O. & Persson, P. (2003). Plant availability of phosphorus in ash, calcium phosphate and different types of sewage sludge. *Vatten* 59 (3), 161-167. (The article is based on the licentiate thesis Linderholm, K. (1997). Fosforns växttillgänglighet i olika typer av slam, handelsgödsel samt aska. *VA-Forsk rapport* 1997-6. ISBN: 91-88392-23-6).

III Linderholm, K., Caspersen, S. & Palm, O. (2012). Effect of sewage sludge and mineral fertilizer on arbuscular mycorrhiza. (*Manuscript*).

IV Linderholm, K., Tillman, A.M. & Mattsson J.E. (2012). Life cycle assessment of phosphorus alternatives for Swedish agriculture. *Resources, Conservation and Recycling* 66 (September), 27-39.

The contribution of Kersti Linderholm to the papers included in this thesis was as follows:

I Major part of the collection and evaluation of data and writing of the manuscript.

II Major part of the planning, maintenance of field experiment, sampling, evaluation and writing of the manuscript. The analyses were performed by commercial laboratories. Statistical analyses were carried out by Birgitta Vegerfors, SLU.

III Major part of the planning, maintenance of pot and field experiments, sampling, evaluation and writing of the manuscript. The analyses of nutrients *etc*. were performed by commercial laboratories. The analysis on mycorrhizal colonization was performed by Ola Palm and Monica Kling at Swedish University of Agricultural Sciences (SLU) in Experiment 1. Iver Jakobsen at Risö National Laboratory, Denmark, conducted the analysis on the field samples in Experiment 2 and Sadhna Ahlström at SLU carried out the quantification on samples from the pots in experiment 2. Statistical analyses were carried out by Birgitta Vegerfors and Jan-Eric Englund, SLU.

IV Major part of the collection of data, calculations and writing of the manuscript.

Abbreviations

AM	Arbuscular mycorrhiza
AU	Animal unit
ATP	Adenosine triphosphate
Cd	Cadmium
CO_2-equiv	CO_2 equivalents
DM	Dry matter
FU	Functional unit
GHG	Greenhouse gas
LCA	Life cycle assessment
LD	Livestock density
MFA	Material Flow Analysis
MJ	Megajoule = 0.28 kWh
P	Phosphorus P = 0.436* P_2O_5
P_2O_5	Phosphorus pentoxide = P/0.436
P-AL	Phosphorus soluble in ammonium lactic buffer
P-HCl	Phosphorus soluble in hydrochloric acid
Struvite	$MgNH_4PO_4$•$6H_2O$, magnesium ammonium phosphate
ton	metric ton = tonnes = Mg (10^6 g)
TSP	Triple superphosphate
WWTP	Wastewater treatment plant

1 Introduction

Phosphorus is an essential element for plants, animals and humans. In animals and humans most of the phosphorus is found in bones and teeth, but phosphorus is also a part of many enzymes, the energy carrier in cells, ATP, and is an important part of nucleic acid, which carries the genes. For plants, phosphorus is irreplaceable in the functions of photosynthesis, nutrient transport and energy transfer.

Phosphorus is also an essential nutrient for life in water. Losses of phosphorus from land or via the sewage system can cause eutrophication of water, giving rise to oxygen deficiency in aquatic environments (Carpenter, 2008). Material rich in phosphorus settles to the bottom, but it takes millions of years before the phosphorus in waters forms sediment worth mining as a phosphorus source (Filippelli, 2011; Nathan *et al.*, 1996). It is therefore worthwhile keeping phosphorus on land as long as possible.

Soils contain phosphorus in different amounts depending on their parent material origin, age and prior fertilization. For this reason, the soil can to some extent deliver phosphorus to the cultivated crop (Horst *et al.*, 2001). The actual amounts delivered depend on the content of nutrients in the soil, the effectiveness of the crop root system in locating and taking up nutrients and factors that affect mineralization in soil, *e.g.* climate, weather, moisture, microbiological activity *etc.*, but the delivery is most commonly suboptimal.

Historically, a common way to grow food on the Scandinavian Peninsula was to burn forest and use the available nutrients in the soil for a couple of years. This is called slash-and-burn agriculture and is still used in some parts of the world (Varma, 2003). In pace with the increasing number of mouths to feed over the centuries, there was a need for more efficient agriculture and it became necessary to supply the soil with the nutrients required. It is possible for some plants to fix nitrogen from the atmosphere by biological means, but

Figure 1. Symptoms of phosphorus deficiency in barley in plant on left.
(Photo: Søren Holm. With permission from Yara Danmark Gødning A/S).

phosphorus and potassium have to be supplied as fertilizer, if lacking (Figure 1).

Phosphorus is an element and cannot disappear, but it can become dispersed and thereby difficult to reuse. The reserves of phosphorus rock worth mining are limited, although it is not known exactly how limited (Driver *et al.*, 1999). A time horizon of 50-100 years has been suggested most frequently (Cordell *et al.*, 2009). In 2010, the International Fertilizer Development Center published a report stating that world resources of phosphorus had been underestimated (van Kauwenbergh, 2010). However, even if the phosphorus resources that can be used as fertilizer will last for some more decades, they are ultimately limited and utilization of these phosphorus reserves also carries an environmental cost.

In the beginning of 1990s, recycling and reuse was on the political agenda. This was also the time when a pioneer flow balance of nutrients for Swedish agriculture and the food chain was drawn up. At that time, the only input included in the flow analysis was mineral fertilizer (Pettersson, 1992). As production of food in Sweden is decreasing, with increasing imports of food and feed, the import of phosphorus in such food and feed can be significant. Today, Sweden is a net importer of food and agricultural products. The deficit has increased every year since 1998, except for 2000 and 2010 (Swedish Board of Agriculture, 2011b).

The fertilizer value of recycled phosphorus in chemically precipitated sewage sludge has been discussed in many studies. The results vary and some authors have concluded from experimental studies that plant availability of phosphorus in sewage sludge is low (Krogstad *et al.*, 2005; Larsen & Damgaard-Larsen 1981; de Haan, 1980), while others have concluded the opposite (*e.g.* Gestring & Jarrell, 1982). The reasons for these diverging conclusions could be that high doses of phosphorus have been used (Krogstad *et al.*, 2005) or that other nutrients such as nitrogen or liming effects have not been compensated for (Gestring & Jarrell, 1982). Rather many experiments have been conducted without plants (Udeigwe *et al.*, 2011; Smith *et al.*, 2002; Maguire *et al.*, 2001), which means that mainly the chemical system in soil is examined in those studies.

Most experiments concerning phosphorus availability in sludge are short-term pot experiments running for weeks or months, although phosphorus nutrition is a long-term question. Very few studies concerning phosphorus availability in sludge have been carried out as field experiments over several years.

The phosphorus cycle in soil is driven by chemical reactions but also by microbes. One well-studied group of important microbes concerning phosphorus delivery to plants is the arbuscular mycorrhiza (AM) fungi. AM is a symbiosis that occurs between fungi and plant roots in most cultivated agricultural and horticultural crops. The AM acts like an enlarged root system and helps the plant to assimilate nutrients. AM is known to be particularly active in the transportation of phosphorus to the plant roots (Smith & Read, 2008; Marschner & Dell, 1994). Microbiological activity in the soil can be affected by unwanted substances in fertilizers such as heavy metals (Witter *et al.*, 2000; Frostegård *et al.*, 1996). AM activity can also be affected by the phosphorus status in soil (Smith & Read, 2008). Although AM has been relatively well studied, studies in relation to sludge fertilization are lacking and especially concerning good quality sludge, *i.e.* with a high content of nutrients and low contamination by hazardous substances.

Even if Swedish sludge is of good quality as a fertilizer, with a low content of heavy metals and other unwanted substances, the discussion about using this sludge as a fertilizer continues. In this debate, different techniques for separating the phosphorus from the wastewater streams have been suggested (Cirkulation, 2009). The topic of separating phosphorus is not new, as international conferences on phosphorus recovery and recycling were held in 1998, 2001, 2004 in Europe and 2009 in Canada (Ashley *et al.*, 2009). Apart from the option of recycling phosphorus and other nutrients as sludge on agricultural land after sanitization, there is a wide range of techniques available for separating phosphorus in sewage streams or from sewage sludge.

Sludge as a waste has been studied previously in Life Cycle Assessment (LCA) (Lederer & Rechberger, 2010; Lundin *et al*., 2000; Tillman *et al*., 1998). Utilization of phosphorus in sludge for different purposes, *e.g*. land reclamation, golf courses and agriculture, has also been investigated (Johansson *et al*., 2008). Furthermore, recycling of phosphorus from small-scale wastewater systems has been studied in a LCA perspective by Tidåker *et al*. (2007). However, to the best of my knowledge, no previous study has examined the supply of phosphorus to agriculture on a large scale.

1.1 Objectives

The overall aim of this thesis work was to study possibilities for more sustainable management of phosphorus as a fertilizer in Swedish agriculture. Specific objectives of Papers I-IV were to:

- Carry out an updated phosphorus flow analysis on a national level for Swedish agriculture and food chain (Paper I).
- Determine the value of different types of sludge and ash as phosphorus sources for a cereal crop in a longer term perspective (Paper II).
- Evaluate the impact of different phosphorus fertilizers (sewage sludge and mineral-P) on AM colonization in Swedish soil in a longer term perspective; and determine whether precipitation chemicals used for treatment of wastewater or impurities in sludge affect mycorrhizal colonization (Paper III).
- Assess the environmental impact of different ways of supplying Swedish agriculture with phosphorus fertilizer of an acceptable quality as regards cadmium content and other impurities (Paper IV).

2 Materials and Methods

The overarching issue examined in this thesis is the sustainable supply of phosphorus as an essential nutrient in production of biomass on arable land. This issue was examined from different perspectives in Papers I-IV and consequently different methods were needed. The methods used in Papers I-IV are described briefly below. For full details of all methods and experiments, see the respective papers.

2.1 Flows of phosphorus to and within the Swedish food chain (Paper I)

Flows of phosphorus in the Swedish food chain, including agriculture, were quantified on a yearly basis by material flow analysis (MFA), sometimes called substance flow analysis (SFA), according to a methodology described by Brunner & Rechberger (2004). The system studied was food production and food consumption in Sweden, including the food chain and related waste and wastewater treatment.

Phosphorus inputs to Swedish agriculture comprise phosphorus in all supplies used in agriculture, *e.g.* fertilizers, feed minerals and imported animal feedstuffs. In addition, atmospheric deposition of phosphorus makes a direct input to arable land. Imports of food and commodities for food processing from other countries were also accounted for as inputs to the system studied and sources to wastewater other than human excreta, *e.g.* detergents. Phosphorus leaving the system was taken to be agricultural products exported to other countries, leakage from arable land and phosphorus in waste and wastewater that not is reused as feed or fertilizer in agriculture. Phosphorus in products and wastes used or recycled within agriculture (*e.g.* feed, manure) does not pass outside the system boundaries, and thus does not affect the national balance.

Most activities in the studied system have a rapid turnover of phosphorus, so the changes in phosphorus stocks in these were not investigated. However, agricultural soil represents a significant stock of phosphorus with a rather slow turnover, so the data obtained in Paper I were used to calculate the phosphorus balance in arable land, *i.e*. the change in phosphorus stock in cultivated soil per year.

Statistics for the period 2008-2010 were used where available. For flows that fluctuate annually, calculations were based on an average for 2-3 years. When official data were lacking, information was collected from scientific papers, databases, authorities or companies. Because of the recent Swedish regulation on phosphates in certain detergents, the data were corrected to reflect the current situation. The calculations were based on the total acreage of arable land in Sweden, including pasture, in 2009. One reason for including pasture was that significant proportions of the phosphorus flow go via milk and meat. An uncertainty analysis was carried out according to a method presented by Hedbrant & Sörme (2001). Uncertainty factors were estimated for data of different origins and the uncertainty interval was calculated by multiplying and dividing the data by the factors.

2.2 Long-term and short-term fertilizer value (Paper II)

A three-year field experiment was set up in mid-western Sweden. The soil was a silty clay loam (Linderholm, 1997). The phosphorus status of the soil was average according to the Swedish soil classification system (Sibbesen, 1983).

The treatment plot size was 8 x 40 m^2 and the treatments were carried out in three blocks. In addition to a control, the fertilizers tested were:

- Sludge precipitated with iron chloride
- Sludge precipitated with aluminium-based chemicals
- Sludge precipitated with slaked lime
- Biological sludge from the pulp and paper industry
- Mineral P fertilizer
- Ash from co-incineration of biological sludge mainly with wood, but also some coal.

All treatments except the control were fertilized in the spring of the first experimental year with 45 kg phosphorus per hectare in the form of the different fertilizers used. This amount was chosen to cover the three-year study period, as a normal cereal crop in the area removes about 15 kg of phosphorus per year and hectare in the grain. No phosphorus was added to any treatment in the second and third year.

The objective was to study phosphorus only, so all treatments, including the control, received nitrogen and potassium in all three years in amounts to meet crop requirements of these nutrients. A liming sub-treatment was also applied, with duplicate limed and unlimed plots of each fertilizer treatment and the control. The grain yield in kg DM/ha was statistically analyzed as a row-column design in three blocks. In each block the columns consisted of the different treatments (fertilizers) and the two rows of liming and no liming (SAS, 2000).

2.3 Impact of sewage sludge on arbuscular mycorrhiza (AM) (Paper III)

Pot and field experiments were set up to study the impact on arbuscular mycorrhiza of using different types of phosphorus fertilizers.

Experiment 1

Soil was sampled from one block in the field experiment described in section 2.2 above and Paper II. The soil was sampled in the autumn one year after the experiment, which was 3.5 years after fertilizing with the different types of P fertilizers. The soil was stored in an outbuilding, in plastic bags, until spring when the experiments were set up. Soil samples from the control and from the mineral P fertilizer, iron-precipitated sludge and biological sludge treatments were used in this pot experiment. The only phosphorus fertilizer added was mineral P fertilizer to some of the treatments, as shown in Table 1. All treatments, including the control, received nitrogen and potassium in equal amounts (Table 1).

Table 1. *Overview of treatments 1998 in pot experiment 1 (Paper III). Treatments in field experiment 1994-1996: All treatment soils except the control received 45 kg P as different types of fertilizers in 1994.*

Treatment in field:	In pots:	4 weeks (4w)			10 weeks (10w)		
		N	P*	K	N	P*	K
	Code:	kg/ha			kg/ha		
Control	Control	120	0	90	360	0	270
"	C+MinP	120	15	90	360	15	270
Iron precipitated sludge	SS	120	0	90	360	0	270
"	SS+MinP	120	15	90	360	15	270
Biological sludge	BS	120	0	90	360	0	270
Mineral P fertilizer	MinPf	120	0	90	360	0	270
"	MinPf+MinP	120	15	90	360	15	270

* The P was supplied as mineral P fertilizer

The crop grown in the pot experiment was Westerwold ryegrass (*Lolium multiflorum*, cv. Barspectra). There were two sets of pots, one where the roots were harvested after 4 weeks and one where the roots were harvested after 10 weeks. The shoots were harvested once in the 4-week pots and three times in the 10-week pots. All pots were randomly placed in a greenhouse during the normal growing period in Sweden (May-September). After each harvest of shoots, the pots were randomly repositioned in the greenhouse. The greenhouse had no artificial heating and the pots were irrigated with tap water.

Experiment 2 in pot and field

An experiment was set up on a field that had been managed according to the rules for organic farming for at least 10 years. After the three-year field trial, soil was collected from all plots and used in a pot experiment.

Table 2. *Overview of field and pot treatments in experiment 2 (Paper III)*

Treatments in field experiment	Field experiment†									Pot experiment		
	2000			2001			2002			2003		
	N	P	K	N	P	K	N	P	K	N	P	K
	kg/ha			kg/ha			kg/ha			kg/ha		
Control	0	0	0	0	0	0	0	0	0	0	0	0
Mineral fertilizer (NK)	60	0	45	60	0	45	60	0	45	80	0	60
Sewage sludge (SS)	110*	94 *	6*	60	0	45	60	0	45	0	0	0

†1990-1999: Organic farming with bovine animals. No input of fertilizers except via fodder.
* From sludge application. For N, estimated ammonia emission is subtracted.

The experimental field was situated in mid-western Sweden. The soil was silt with pH (H_2O) 5.7 (SS, 1994). Phosphorus status in terms of more easily soluble phosphorus (P-AL) was average in the Swedish soil classification system (Sibbesen, 1983).

Only one treatment, the sewage sludge, received additional phosphorus. The treatments in the first year were: Control (no fertilizer), mineral fertilizer (60 kg N and 45 kg K/ha) and iron-precipitated sewage sludge at 4 ton DM/ha, which gave 94 kg P/ha and 110 kg Tot-N/ha (Table 2).

In the second and third year the control was unfertilized and the two other treatments received 60 kg N/ha and 45 kg K/ha as mineral fertilizer. No phosphorus fertilizer was added in any treatment in the second and third year. No harvesting was carried out in the first year. The aboveground ley material was harvested in the second and third year, and then roots were harvested in August of the third year.

Soil was sampled from all plots after the field experiment and used in a pot experiment with Westerwold ryegrass. In the pot experiment no phosphorus fertilizer was added, but one treatment received nitrogen and potassium in the form of mineral fertilizer as shown in Table 2.

Crop cultivation and sampling were the same as for the 10-week pots in experiment 1, with the exception that the shoots were harvested twice.

Analysis

Shoots were dried, weighed and analyzed in commercial laboratories. The phosphorus concentration in the shoots was analyzed by ICP-AES determination after microwave-assisted acid digestion (Nordic Committee on Food Analysis NMKL 161).

Roots were washed and weighed and then sub-samples were taken, cleaned and stained with tryptan blue (Giovannetti & Mosse, 1980). This made it possible to study the fungal occupation of the host tissue. The remaining roots were dried at 80 °C for 48 h and weighed. Total and mycorrhizal root lengths were measured using microscopy and a line-intersect method (Tennant, 1975; Newman, 1966).

Statistics

For observations from more than one harvest, the data were analyzed using analysis of variance with a split-plot design. Data with no repeated observations were statistically analyzed using analysis of variance for a completely randomized design (SAS, 2000).

2.4 Environmental impact of different phosphorus fertilizers (Paper IV)

Life Cycle Assessment (LCA) was used to assess the environmental impact of different ways to supply Swedish agriculture with phosphorus. LCA is a well-established method for conducting a holistic environmental evaluation and avoiding sub-optimization. It is possible to handle a large amount of data and compare the environmental impact of different options. LCA as a method is regulated in ISO standards (ISO, 2006a, 2006b) and described in textbooks (Baumann & Tillman, 2004).

The options for phosphorus fertilization of arable land studied here were mining of virgin phosphorus; reuse of phosphorus from wastewater through sewage sludge and struvite precipitation in WWTP; and recovery of phosphorus from incinerated sludge (Figure 2). Data were collected from operations and plants working on a commercial basis, official statistics, authorities and scientific publications.

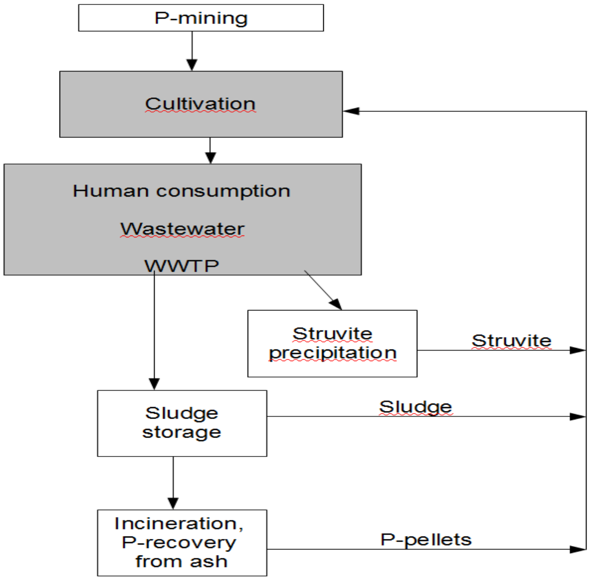

Figure 2. Options for phosphorus fertilization studied in Paper IV: Mining of virgin phosphorus; reuse of the phosphorus in sewage water though sewage sludge and struvite precipitation; and P recovery from incinerated sludge. Processes in the grey boxes were not included in the LCA calculations.

The functional unit (FU) was chosen to be 11 kg phosphorus (25.2 kg P_2O_5). The reason was that the harvested crop on average removed 11 kg phosphorus per hectare in 2007 (Statistics Sweden, 2011b). Swedish environmental monitoring data show that on average, the soils in Sweden have a satisfactory content of phosphorus (Eriksson *et al.*, 2010). Since the Swedish Board of Agriculture recommends balanced phosphorus fertilization, *i.e.* only replacing harvested phosphorus, on soils with satisfactory phosphorus status (Swedish Board of Agriculture, 2011a), 11 kg per hectare would be enough to support recent crop production in Sweden with phosphorus.

The resources studied were energy and phosphorus. Global warming and eutrophication by phosphorus were studied as ecological effects. Energy was considered as primary energy. Global warming was assessed in terms of CO_2-eq. using data taken from the literature. Cadmium added to soil was calculated as an indicator of the risk of cadmium contamination in food grown on arable land. Release of phosphorus to waters was used as an indicator of eutrophication. It should be noted that release of phosphorus and nitrogen to waters from cultivation and wastewater treatment was outside the scope of this study.

The study examined use of phosphorus fertilizer from the different sources, including its spreading on farmland. An assumption in the study was that in a longer-term perspective, all added phosphorus was of the same value for the crop (Linderholm, 1997). The geographical boundary for phosphorus requirements was taken as arable land in Sweden, concerning transport, techniques, legislation and fertilization strategy.

To make the options comparable, the fertilizer value of nitrogen in sludge and struvite was taken into account. The sludge and struvite options were credited with the avoided production of an equivalent amount of nitrogen fertilizer that was assumed to be used by the crop (40 % of Tot-N).

3 Summary of results and discussion

3.1 Flows of phosphorus to Swedish agriculture and food chain (Paper I)

Phosphorus imports in mineral fertilizer peaked in 1973-1974 at a level corresponding to 24 kg/ha arable land (Statistics Sweden, 1995). In recent decades the use of phosphorus mineral fertilizer has declined and for the study period average imports of mineral phosphorus fertilizer were equivalent to 3-4 kg P/ha (Statistics Sweden, 2011c). Besides mineral fertilizer, meat-and-bone meal is imported as a phosphorus nutrient source in organic farming.

Most roughage and grain required for animal production are produced inside Sweden, but many concentrates and minerals are imported. To calculate the phosphorus imports in feedstuffs, data sources from the Swedish Board of Agriculture and Statistics Sweden were used. Animal feed minerals are produced in Sweden by one company. The raw material, phosphoric acid, is imported. About 2700 ton phosphorus per year are used as animal feed minerals or feed phosphates (S.-O. Malmqvist, pers. comm. 2011).

Information about the volume of food imports and exports (Statistics Sweden, 2011a) was combined with information on DM content and phosphorus content from databases and the literature. Separate calculations were made for fish, since much imported fish is salmon from Norway, which is not consumed in Sweden but exported to a third country (Swedish Board of Agriculture, 2011b).

Most of the phosphorus in waste from food processing industries such as breweries, slaughterhouses, dairies, sugar mills, *etc*. is used either as a fertilizer or feed, including pet food (Wivstad *et al*., 2009; P.-J. Lööf, pers. comm. 2011). Pet food is defined as a flow exiting the system, as pet manure does not end up on farmland or in wastewater.

Fallen animals and risk material from slaughterhouses are treated and used as a fuel called Biomal, which leaves the system since the phosphorus ends up in ash. Sweden has one large-scale producer of grain-based fuel ethanol. A by-product of ethanol production is a protein feed (Agrodrank). In 2008-2009, about 40-45% of the Agrodrank produced was exported (B. Beckman, pers. comm. 2011).

About 1.2 million people in Sweden have private sewage systems and 290 000 summer cottages are not connected to municipal WWTP (Swedish EPA, 2012a). A minor proportion of the sewage is collected and ends up in a WWTP but most (80-90%) is infiltrated into soil, with a possible risk of polluting surface waters or groundwater (Steineck *et al.*, 2000). Of the phosphorus entering WWTP, 95% ends up in sewage sludge. In 2008, 72% of the sludge produced in WWTP according to the statistics fulfilled the legal requirements for use on agricultural land, but only 26% was used in this way (Statistics Sweden, 2010).

Manure is circulated in the agricultural system and therefore does not affect the phosphorus balance for Sweden. However, the figures are interesting for comparison with other phosphorus flows and in further calculations. The amount of phosphorus in manure was 25 080 ton in 2009, including 7440 ton phosphorus left on grazing land by animals (Statistics Sweden, 2011b), which means that 17 640 ton phosphorus in manure were spread actively on fields by machinery.

The results showed a net input of phosphorus to Sweden of about 12 600 ton, corresponding to 4.1 kg P/hectare (Figure 3).

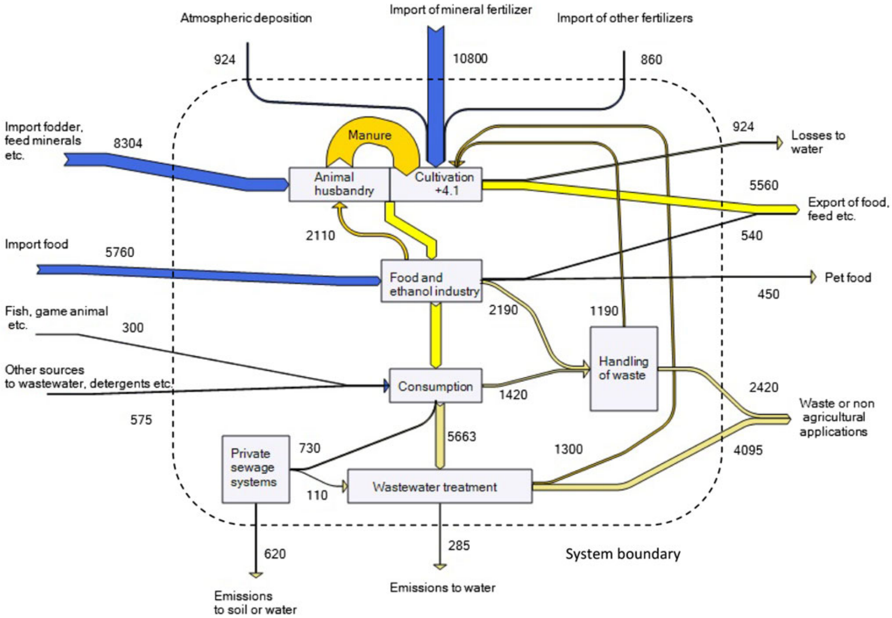

Figure 3. Quantified flows in tons of phosphorus in Swedish agriculture and the food chain. *Boxes* The processes of cultivation, consumption, treatment *etc*. Stock change in soil is shown in kg/ha. The width of arrows corresponds to the size of the flow of phosphorus. When a corresponding value is lacking for an arrow, the flow is only indirectly calculated from other results (Paper I).

The area of arable land needing above-balance phosphorus fertilization is less than the area of phosphorus-rich land (Eriksson *et al*., 2010). If available phosphorus sources were used, in theory no imports of mineral fertilizer would be needed.

The calculations were based on Sweden as a whole and an important condition is that the phosphorus is used optimally in the whole country. However, as shown in Figure 4, the surplus phosphorus is concentrated via manure on farms with more than 1 animal unit/ha. Farms with few or no animals have a negative phosphorus balance and sell off more phosphorus from their land than they replace with fertilizers.

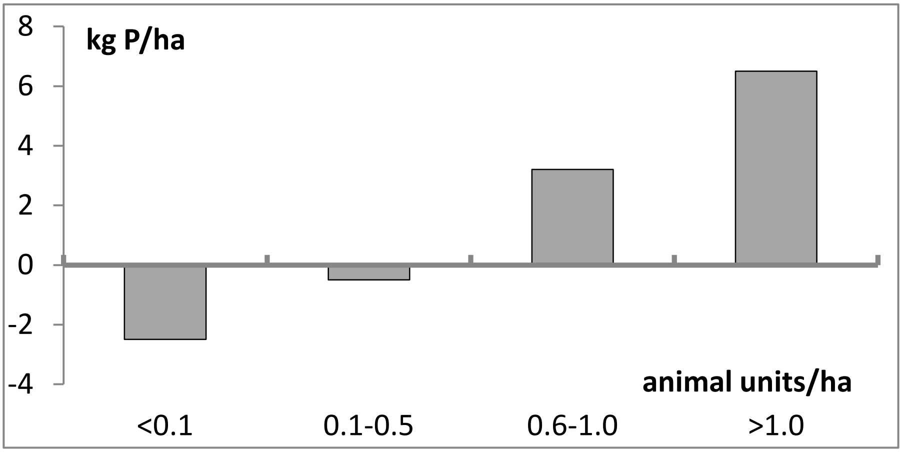

Figure 4. Phosphorus balance on farms with different animal density ranges (based on data from Statistics Sweden, 2011b).

This unbalanced use of phosphorus is the result of the change towards larger animal farms. Organic farms are no exception. Recent statistics from the Swedish Board of Agriculture show that organic farms with milk or meat production have more animals than the corresponding conventional farmers (Swedish Board of Agriculture, 2012b). However, as far back as 1988, Sweden regulated the permissible livestock density (LD) on farms on the basis of manure phosphorus content and not only nitrogen content (LSFS, 1988). The result is that Sweden has among the lowest net inputs of phosphorus to farmland in Europe.

3.2 Short-term and long-term fertilizer value (Paper II)

The first experimental year, when the phosphorus was added, was very dry and there were visible differences in the crop between the treatments. Mineral P fertilizer, together with the ash treatment, gave significantly better crop growth than the other treatments. The treatments with sludge and the control showed symptoms of lack of phosphorus.

The mean yield in the first year was very low, below 1900 kg dry matter (DM) per hectare of oats in the unlimed plots and nearly 2 200 kg DM grain/ha in the limed plots.

The second year of the experiment also had relatively low precipitation but the mean yield was normal, 3170 kg DM barley grain per ha. This was higher than the average yield in this part of Sweden, 2570 kg DM/ha (Swedish Board of Agriculture, 2012a). There were no significant differences in grain yield between the different treatments.

Two years after phosphorus fertilization, in the third year of the experiment, the mean yield of grain was very high, 5038 kg DM/ha. There were no significant differences in grain yield between the different treatments or the control.

Under dry weather conditions, it is apparently important for the plants to have access to easily soluble phosphorus in the soil, since the soil has problems in supplying sufficient phosphorus from mineralization of the organic and inorganic pool in these conditions. It is difficult to explain why the ash almost gave the same yield as mineral P in the first year, but it could be due to the liming effect of ash.

The most astonishing result of this field experiment was that the chosen 'average' field, lying in a typical grassland area with poor conditions for cereal production, gave an extremely high yield overall (on average over 5000 kg DM grain/ha) in the third year of the experiment. A soil with average phosphorus content can apparently deliver sufficient phosphorus to give a very high yield of grain in a year with optimal moisture and temperature, without any additional phosphorus being added.

In a long-term perspective, the form of phosphorus fertilizer used seems to be less important than maintaining the phosphorus content in the soil to guarantee phosphorus delivery to the crop when conditions such as moisture, temperature *etc*. are optimal. Although a three-year field experiment is too short for a full evaluation of fertilizers, the results indicate that in the long run, crop supply of phosphorus is more dependent on a correct comprehensive evaluation of soil type-crop-fertilizer than on the chemical form of the phosphorus in the fertilizer at the time of spreading.

3.3 Effect of sewage sludge and mineral fertilizer on AM (Paper III)

Experiment 1, pots

Four years after application of different phosphorus fertilizers as described in section 2.2 and in Paper II, the soil was used in a pot experiment (Paper III). Mycorrhizal colonization was found to be established in all treatments except that which had received both mineral P in the field in the first experimental year and in the subsequent pot experiment (MinPf+MinP) (Figure 5).

After 10 weeks, root length and colonized root length were significantly longer in the soil that had received sewage sludge in the field experiment (SS). Soil that received 45 kg mineral P in the first year in the field experiment or 15 kg mineral P in the pot experiment gave significantly low positive intersections and colonized root length in the pot experiment compared with the other treatments.

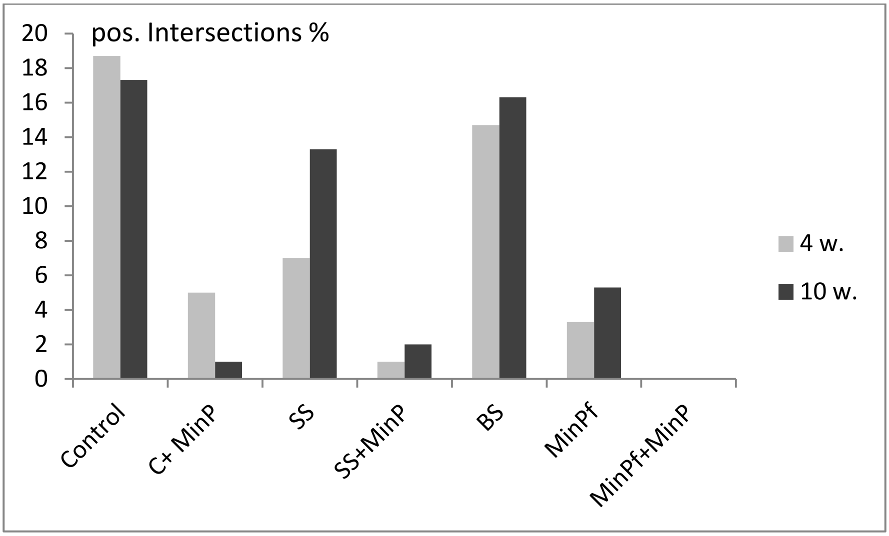

Figure 5. Percentage (%) of positive intersections in experiment 1 after 4 and 10 weeks. The treatment with mineral fertilizer in both field and pot experiments had 0% positive intersections. MinPf = 45 kg mineral P fertilizer/ha in field experiment; MinP = 15 kg mineral P fertilizer/ha in pot experiment.

Experiment 2, field and pot

There were no significant differences between the treatments regarding colonization by mycorrhiza in the field or pot experiment. In the pot experiment, which used soil from the field experiment, the DM yield was significantly higher in the only treatment that received mineral fertilizer (NK).

The results showed that it is possible to have good yield without compromising the possibilities for AM fungi to colonize the root (Figure 6). It was found that inadvertent addition of impurities or precipitation chemicals with quality-assured sludge in moderate amounts had little or no effect on the establishment of AM, measured as root colonization. Furthermore, 45 kg P/ha added in the form of sewage sludge did not disturb the establishment of AM, whereas mineral P added as a storage supply (45 kg P/ha) four years earlier decreased the AM dramatically.

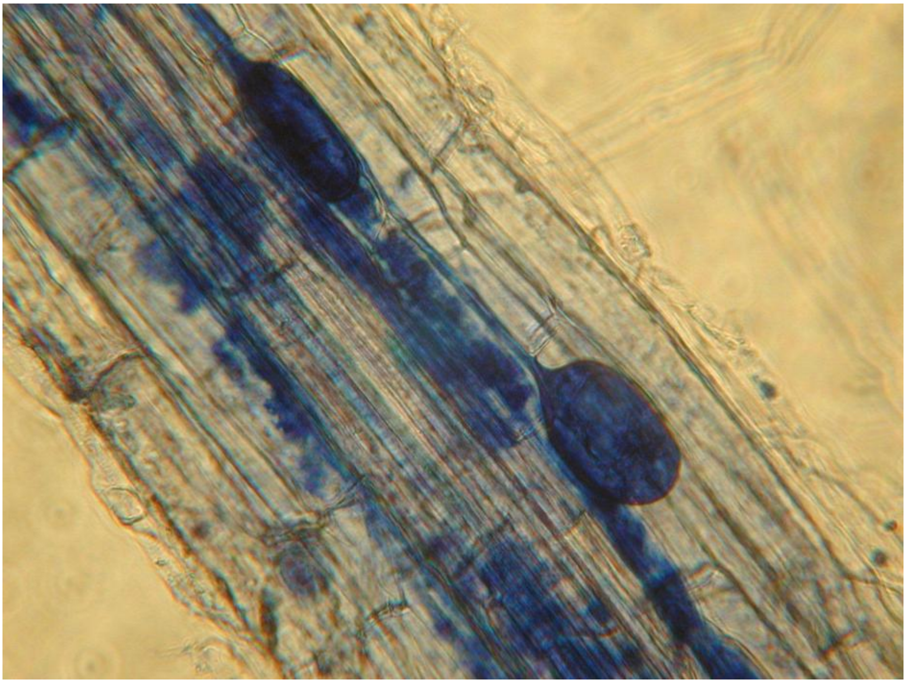

Figure 6. AM fungal storage structures (vesicles) in the root of plants from Experiment 2 (pots). (Photo: Bharadwaj and Ahlström, SLU).

3.4 Environmental impact of supply and reuse of phosphorus (Paper IV)

Sewage sludge as a phosphorus fertilizer used less energy and gave lower emissions of greenhouse gases than any of the other options studied. The negative value for sludge is due to the crediting for the content of nitrogen in sludge (Figures 7 and 8). Even if sludge and struvite only contain a small amount of nitrogen, all of which can be assumed to be used by the crop, this nitrogen is significant in a study such as this.

Nitrogen is a valuable nutrient and is energy-demanding to replace, so sewage sludge and struvite that can keep some nitrogen in usable form for production are preferable. A disadvantage with incineration is that nitrogen is lost to the air and energy is required to re-capture this nitrogen in mineral fertilizer or through nitrogen fixation by legumes.

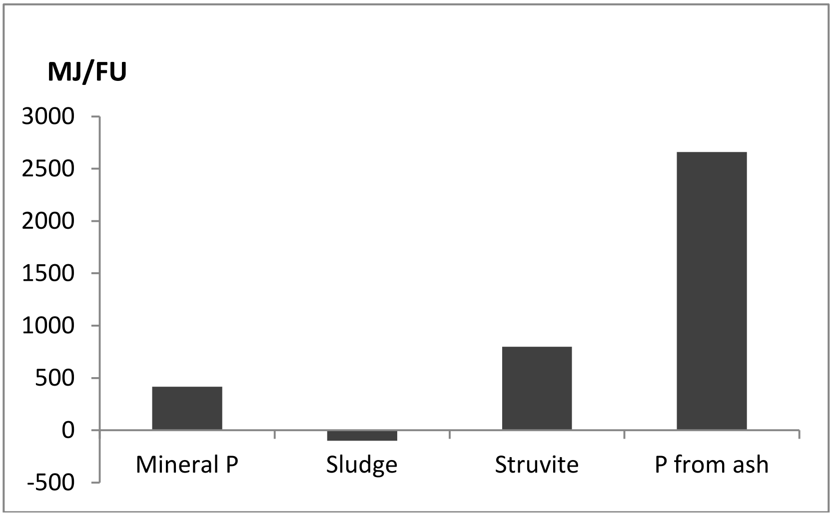

Figure 7. Energy use in the production of 1 FU (11 kg P) for the different sources of phosphorus studied in Paper IV. The negative value for sludge is due to the crediting for the content of nitrogen in sludge.

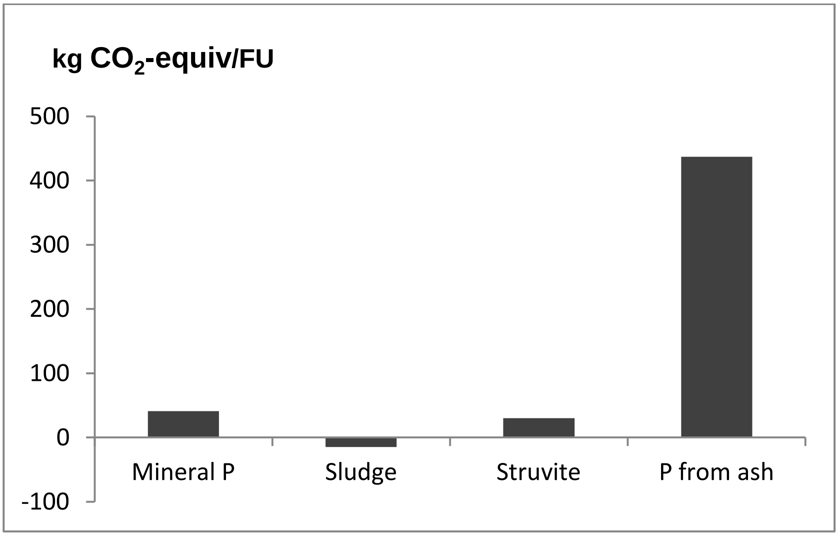

Figure 8. Emissions of greenhouse gases in the production of 1 FU (11 kg P) for the different sources of phosphorus studied in Paper IV. The negative value for sludge is due to the crediting for the content of nitrogen in sludge.

Phosphorus recovery from incinerated sludge was the most energy-demanding option and gave most emissions of greenhouse gases. Figures 9 and 10 provide a more detailed picture of the results for the recycled phosphorus options.

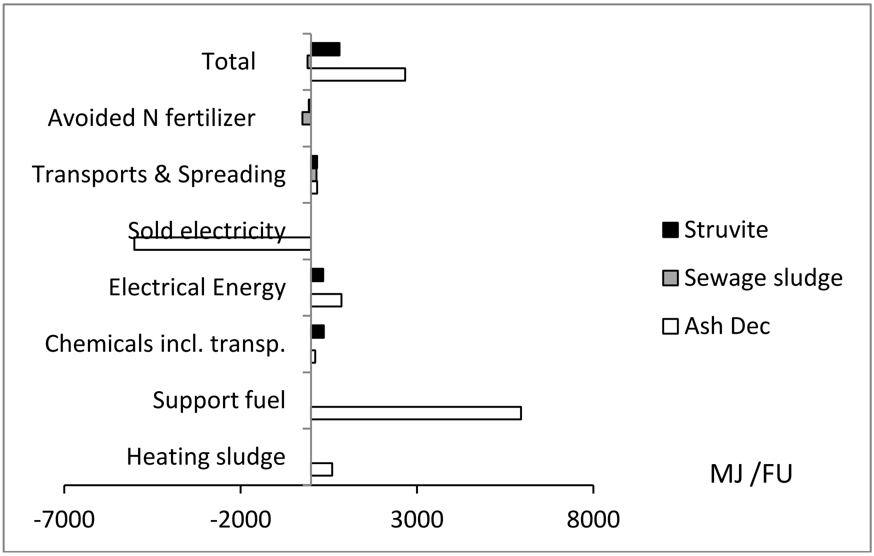

Figure 9. Energy demand and energy recovery for recovered phosphorus (MJ/FU) including primary energy.

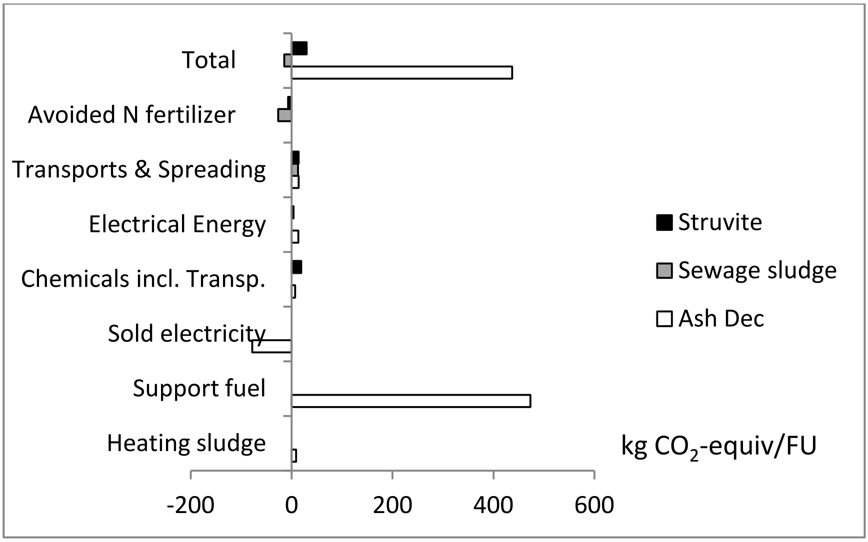

Figure 10. Emissions of CO_2-equiv and avoided emissions for recovered phosphorus in kg CO_2-equiv/ FU, including emissions from production.

The only option that used virgin phosphorus was the process with mineral fertilizer. For 1 FU in mineral fertilizer, 11 kg P (25.2 kg P_2O_5) are required. Fertilizing with sewage sludge, struvite and P from ash are all different ways to reuse phosphorus that has already been a part of food or feed.

Mineral fertilizer and sewage sludge contained most cadmium, 62 and 314 mg per FU, respectively. This is in the same range as cattle slurry, which contains about 187 mg per FU (calculated from Steineck *et al*., 1999). However, atmospheric deposition of cadmium is still the main source of cadmium to arable land, supplying about 0.4 g Cd/ha and year (KEMI, 2011; Eriksson, 2009), which can be expressed as 400 mg Cd/FU.

Struvite and phosphorus recovered from ash contained less than 60 and 4.4 mg cadmium per FU, respectively. The phosphorus fertilizer from incinerated ash adds almost no cadmium to the soil. In this case the cadmium normally ends up in the fly ash. Analyses on cadmium in struvite are very few, but struvite also seems to have a low content of cadmium, in the same range as mineral fertilizer. In the case of struvite, the cadmium probably ends up in the sewage sludge that is produced in addition to the struvite. Looking only at the cadmium contribution to soils, the alternative of burning the sludge is the best option. However, it should be borne in mind that flue gas cleaning and the environmental costs for the fly ash were not included in this study due to lack of information. Cadmium is an important issue in the flue gas and ends up in the fly ash.

A very difficult question is how to account for the solar energy and carbon dioxide (CO_2) bound in the field and released later, for example in the incineration of sludge. There are two options for this:

1. Credit arable land for the binding of solar energy and CO_2. In this option, the emissions of CO_2 are counted when released in incineration and energy from incineration of sludge is not included.
2. Do not credit arable land. In this option the CO_2 emissions from incinerating the sludge are not counted and the energy released is credited to the incineration.

Both options have problems relating to lack of data, but option 2 is probably the most common way of accounting in LCA. It was also the method used in this study, since the LCA concerned a secondary product from the field, sewage sludge, and calculation of losses of carbon dioxide and use of energy in human consumption would be difficult. This means that the energy released in the incineration was credited to the process whereby phosphorus was extracted from ash and counted as avoided energy, and the carbon dioxide emissions in the incineration were not included. Figures 11 and 12 show the LCA results (‘Ash 1’) and the situation with the solar energy and the binding of carbon dioxide in the field credited to the field and not the incineration of the sludge (‘Ash 2’).

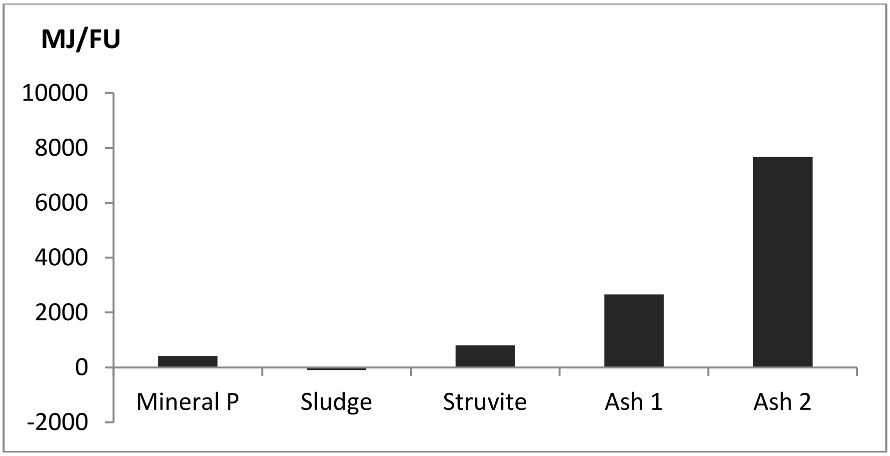

Figure 11. Results from the LCA study (Ash 1) with an additional option 'Ash 2', which shows energy consumption if energy bound in biomass is assigned to arable land and not incineration.

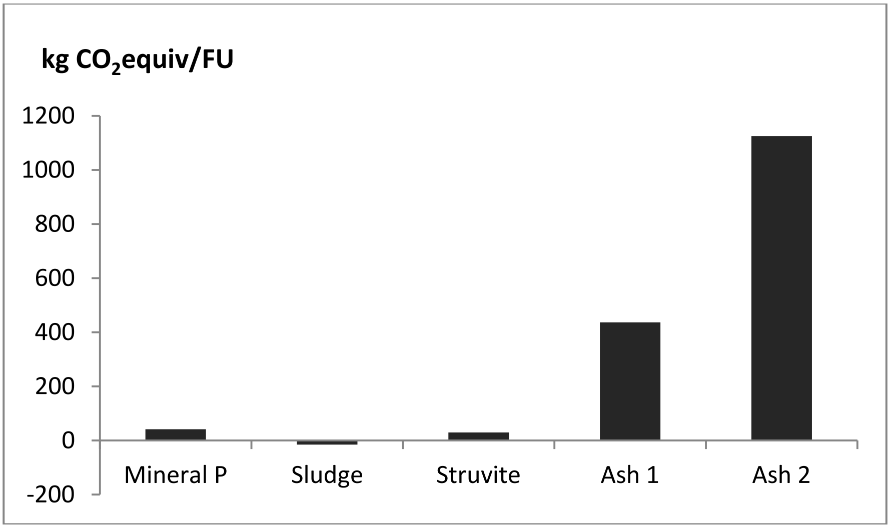

Figure 12. Results from the LCA study (Ash 1) with an additional option 'Ash 2', which shows emissions of CO_2-equiv/FU if the incineration is charged for the emissions.

Production of 1 FU in the form of mineral phosphorus fertilizer released around 5 x 10^{-6} kg P to water. Production of 1 FU in the form of recycled phosphorus released no phosphorus to water within the boundaries of the system studied here, since emissions of phosphorus from WWTP were not included in this LCA. However, from the WWTP included in the statistics, 313 ton P are lost to recipient waters (Statistics Sweden, 2010). In 2008, these WWTP gave more than 0.5 million FU in sludge (6000 ton P). This means that 1 FU from Swedish WWTP releases 0.54 kg phosphorus to water. Struvite precipitation as practised in Edmonton, Alberta, Canada, would increase phosphorus emissions to water. However, Swedish legislation does not allow this, so the assumption here was that chemical precipitation would be adjusted in the WWTP to avoid an increase in eutrophication. The figure 0.54 kg phosphorus is then the same for all recycled phosphorus options studied.

When 1 FU is used on farmland, in addition to 2 kg phosphorus from other fertilizer sources, the emissions are 0.3 kg phosphorus (Swedish Board of Agriculture, 2008). Moreover, there is probably a difference for blue-green algae between the emissions of phosphorus from arable land and those from wastewater treatment and private sewage systems (Krogstad & Løvstad, 1991). The phosphorus from wastewater is mainly in phosphate form, while the phosphorus from arable land is mainly in particulate form and not potentially bioavailable (Filippelli, 2011).

This means that 1 FU as recycled phosphorus emits more phosphorus to waters during production than during cultivation (Figure 13).

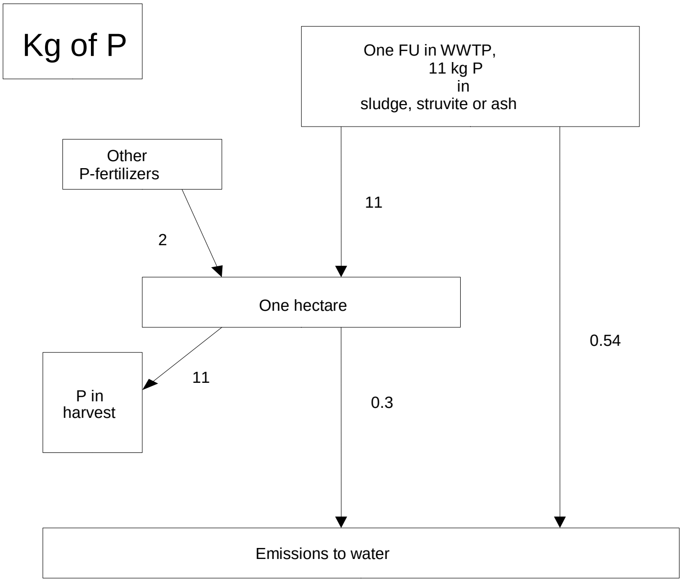

Figure 13. Flowchart showing phosphorus emissions to water from arable land, which contributes 0.3 kg P to water. The emissions from 1 FU when produced in WWTP are almost double, 0.54 kg P (Paper IV).

However, regardless of the use of phosphorus in wastewater, the emissions of phosphorus to waters will still be the same from WWTPs.

4 General discussion and conclusions

4.1 Phosphorus

The results presented in this thesis show that under recent agricultural production in Sweden, with considerable imports of food and feed, crop access to phosphorus is sufficient even without import of mineral fertilizer. However, there is a need for more optimal use of phosphorus in organic fertilizers, especially manure. Sweden can be self-sufficient in staple foods and animal feed if agricultural land is used optimally (Larsson, 2004), but this is not the case at present. If Sweden were to decide on a higher degree of self-sufficiency, there would be a need for more phosphorus than shown in this study.

This finding indicates that successful phosphorus fertilization is a question of maintaining the soil phosphorus status so it can supply the crop requirements when conditions are optimal. Phosphorus supply is a question that must be considered on a long-term basis.

4.2 Cadmium

Under conditions with no imports of food and feed, there would in principle be a situation where only the cadmium from arable soil was returned with the waste products from human and animal digestion. However, two sources of cadmium may give a net input to Swedish arable land; imported food and feed, and atmospheric deposition.

Today Sweden imports a great deal of food and feed, which is probably grown with fertilizers with higher cadmium levels or more contaminated sludge, since this is the typical situation in the world. Despite the great concern about cadmium in Swedish fertilizers and sludge, almost no regular analyses are made on imported vegetables, meat, *etc*. (A. Jansson, pers comm. 2011; Å.

Kjellgren, pers comm. 2011). This means that the amount of cadmium imported with food products is unknown. In contrast, for sewage sludge, which is not eaten but used as a fertilizer, a minimum of 2660 analyses are performed every year (A. Finnson, pers comm. 2011). It is perplexing that the controls and legislation about cadmium in fertilizers, including sludge, are stricter than those for the food we eat.

Atmospheric deposition of cadmium is the main source of cadmium to arable land in Sweden and is estimated to be 0.35-0.45 g/ha (KEMI, 2011; Eriksson, 2009), *i.e.* 1200 kg per year. This is seven-fold higher than the 170 kg cadmium per year in all sewage sludge from Swedish WWTP (Statistics Sweden, 2010).

Another important factor is cadmium to waters. A clean sludge benefits the environment by giving less chemicals and metals in the water leaving the WWTP (Palmquist, 2004). No WWTP can eliminate all metals and chemicals in the incoming water. Using sludge as a fertilizer highlights the importance of receiving wastewater to the WWTP that contains as few unwanted substances as possible.

4.3 Manure

The total quantity of manure in Sweden contains 25 000 ton phosphorus (Statistics Sweden, 2011b), almost in the same range as total imports of phosphorus to Swedish farmland (28 000 ton) according to the results in Paper I. To get a balance on phosphorus at field level and not only on country level, the phosphorus in manure needs to be used more optimally. This can be done by transporting manure. Other ways to solve the phosphorus problem are to encourage animal farmers not to expand too much according to available land or to recover the phosphorus in manure by technical methods and re-distribute it to farmers without animals (IWA, 2009).

However, it should be borne in mind that Sweden was early in regulating the animal density on farms to the phosphorus content in manure (LSFS, 1988), which raises the question of what the situation is like in other countries.

4.4 Reflections about photosynthesis in LCA

For the agronomist, it seems somewhat illogical that the solar energy and carbon dioxide bound in the field by photosynthesis are not credited to agriculture. If the boundary for LCA calculations were to be set between production in agriculture and use of the products, the results would be different.

The term production of bioenergy is normally only used for products that give energy which can be used as fuel or heat and very seldom the bioenergy in food or feed. Wheat produced in agriculture can be used for ethanol production (energy for cars) or bread (energy for people). In LCA calculations the production of food, *e.g*. wheat, is a burden, but when the same wheat is used to produce energy for cars the result is positive and counted as a community benefit. Products such as straw can be incinerated and give energy without the emissions of carbon dioxide being counted. Straw can also be used as a building material and the carbon is then bound for a long time. In both cases, the binding of carbon dioxide from the atmosphere could be credited to the field, whatever the final use of the product.

The Swedish Board of Agriculture is working on a project to make farmers more energy-efficient (Swedish Board of Agriculture, 2011c). The project is mapping the use of energy on farms with the aim of reducing the amount used. According to this way of thinking, it is not permissible to use more nitrogen and get higher yield, even if this gives net production of energy and binding of carbon dioxide.

The same thinking is found in Sweden's report under the United Nations Framework Convention on Climate Change and the Kyoto Protocol (Swedish EPA 2012b). The summary of the report contains the following sentence: *The most important reasons for the reduced emissions are reduced livestock keeping and reduced application of N fertilisers in agriculture*. In other words, less production gives less emissions. However, the energy balance when using N fertilizers is around 7, which means that the energy output is seven times the energy input (Odling i Balans, 2000). Energy binding with the help of solar energy and photosynthesis also means binding of carbon dioxide. It would be more justifiable if the net balance (or surplus) of carbon dioxide were counted, and not only the emissions.

In contrast, the forest sector, which besides agriculture and aquaculture is the only sector that binds solar energy and carbon dioxide via photosynthesis, is considered a sink for carbon dioxide. The question is why food for humans is expected to lie inside the system boundaries of agriculture, but not the emissions from the supplies used to produce the food. And why is it different if the farmer sells grain to ethanol production or to a bakery?

DeCicco (2012) suggested the use of annual basis carbon (ABC) accounting, which does not automatically credit biogenic carbon. He concluded that the method was well suited for addressing biofuels, which involve uncapped sectors (agriculture and forestry). He also concluded that these kind of calculations would lead to emissions-limiting decisions and would enhance the prospects for major technology and systems changes that are truly climate friendly.

As long as the bioenergy consumed by humans is not counted in the same way as bioenergy for cars, heating *etc.*, there will be difficulties in valuing agricultural production in a climate change perspective.

4.5 Conclusions

- Sweden has a positive balance of phosphorus, but the phosphorus is concentrated on farms with more than one animal unit per hectare. Even if no mineral fertilizer at all were imported, the phosphorus balance would be positive for Sweden as a whole, due to imports of phosphorus in food and feed.
- Easily soluble phosphorus is of the utmost importance for the plant in a short-term perspective under dry conditions. In a long-term perspective, the form of phosphorus fertilizer used seems to be less important than a soil in balance between moisture, air, temperature and fertilizer. It is important to maintain the phosphorus content in the soil to guarantee phosphorus delivery to the crop in the future.
- Addition of 45 kg phosphorus per hectare in an easily soluble form on a soil with a moderate content of phosphorus had a negative effect on establishment of mycorrhiza according to analyses four years later. Sewage sludge, quality assured and used as a phosphorus fertilizer in moderate doses, did not have any negative effects on mycorrhizal colonization.
- Phosphorus recovery from ash is costly in terms of both energy and emissions of carbon dioxide. Using the sludge directly on farmland is the most efficient option in terms of energy and emissions of carbon dioxide, but the cadmium contribution to soils can be a problem. The cadmium imports with food to Sweden are unknown.
- Nitrogen is a valuable nutrient and is energy-demanding to replace, so systems that can keep the nitrogen in usable form for production are preferable.
- Life Cycle Assessment calculations need to take into account the binding of solar energy and carbon dioxide in the field. The solar energy and carbon dioxide bound during photosynthesis should be assigned to agricultural production and not the user of the products, *e.g.* sludge incineration. Assessments on the production of bioenergy should include all products that contain energy bound by photosynthesis, not only products used for fuel and heating.

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Svensk Sammanfattning

Fosfor är helt livsnödvändigt för växter, människor och djur och är oersättligt vid odling. De brytvärda tillgångarna av fosfor är begränsade, särskilt de med låg halt kadmium vilket är den fosforråvara som Sverige efterfrågar.

I denna avhandling studerades förutsättningar för en uthållig försörjning med fosfor till svenskt jordbruk. Fosforflöden till svenskt jordbruk och matkedja undersöktes med hjälp av en materialflödesanalys (MFA). Värdet av mineralgödsel, olika typer av slam, samt aska som fosforgödselmedel undersöktes i ett tre år långt fältförsök. Växten samarbetar ofta med svamp för att bättre komma åt fosfor i jorden. Det samarbetet kallas mykorrhiza. Hur mykorrhizan påverkas av olika fosforgödselmedel studerades i fält- och kärlförsök. Slutligen gjordes en livscykelanalys (LCA) över olika sätt att förse svenskt jordbruk med fosforgödselmedel för att bedöma de olika gödselmedlens miljöpåverkan och energiåtgång.

Flödesanalysen av fosfor visade att Sverige har en positiv balans av fosfor, men att fosforn koncentreras på gårdar med mer än en djurenhet per hektar. Beräkningarna gjordes på nuvarande produktion i Sverige vilket innebär att om Sverige höjer sin självförsörjningsgrad av livsmedel så behövs mer fosfor till systemet.

Studien av fosforns växttillgänglighet visade att tillgången till lättlöslig mineralgödsel kan vara avgörande för acceptabel skörd vid torra förhållanden eller på jordar med lägre fosforhalt. Då förhållandena är optimala för grödan kan stora skördar erhållas även utan årlig fosforgödsling på jordar med normalt eller högre fosforinnehåll.

Samarbetet med mykorrhizasvampen påverkades inte av gödsling med kvalitetssäkrat slam, dvs. näringsrikt slam med låg grad av oönskade ämnen. Däremot minskade mykorrhizan dramatiskt i den behandling som hade fått en förrådsgiva av fosfor (45 kg/ha) med mineralgödsel fyra år tidigare.

Livscykelanalysen visade att utvinning av fosfor från aska är dyrbart för miljön både räknat i energi och utsläpp av växthusgaser. Den mest miljövänliga metoden, räknat som energiåtgång och utsläpp av växthusgaser, är att använda avloppsslam direkt på åkermark, men kadmiumtillförseln kan bli ett problem. Mycket av kadmiumet i slammet kommer med födan av vilken en stor andel är importerad. Därför är det anmärkningsvärt att det görs mycket få analyser av kadmium på importerade livsmedel. Trots att avloppsslam bara innehåller en mindre mängd kväve som kommer till nytta i odlingen, så ger detta kväve starkt utslag i LCA-beräkningar. Det är därför en nackdel att göra sig av med kvävet genom förbränning av slam för att kunna utvinna fosfor ur askan.

I beräkningarna tillämpades den accepterade metoden att inte räkna koldioxidutsläppen vid förbränning av slam. En slutsats av LCA-studien är att det är mer rättvisande att tillskriva åkern och jordbruket bindningen av solenergi och koldioxid och inte den som använder produkten (förbrännaren). Termen bioenergi borde även omfatta bioenergi i livsmedel. Jordbrukaren kan välja att sälja sitt vete till kvarnen eller etanolfabriken. Samhällsnyttan borde anses vara lika stor (eller större) att producera mat som energi till bilar.