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Regional Substance Flow Analysis for Assessment of Long-term Phosphorus Accumulation in Soil

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Master in Industrial Ecology
Submission date: June 2013
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Marina Zabrodina

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Sammendrag

Fosfor er en ikke-fornybar ressurs som er avgjørende for matproduksjon. Samtidig, kan fosfor forårsake miljøproblemer siden overskudd av fosfor i landbruksjord fører til eutrofiering. For rasjonell fosforforvaltning trenger man pålitelige estimater av fosforlager og fosforstrømmer og forståelsen til fosfordynamikk i matjord. Studier med Materialstrømsanalyse av fosfor som inkluderer fosforlager i jord er få og gir ikke en pålitelig innsikt i jordfosforlager og deres bidrag til planteproduksjon fordi de analyserer først og fremst de globale og nasjonale fosforstrømmene.

Denne studien hadde som mål å vurdere akkumulering av fosfor i matjord over en lengre periode ved hjelp av en regional fosforstrømanalyse. Tre norske regioner med kontrasterende landbruksproduksjonssystemer ble valgt: Akershus med dominerende kornproduksjon, Rogaland med husdyr produksjon og Sør-Trøndelag med blandet jordbruk. Fosforstrømmer og jordbalanse ble beregnet for 1950-2011 på årsbasis og sammenlignet med estimater av tilgjengelig fosfor i jord basert på P-AL analysedata. Resultatene av P-AL analyse gjenspeiler ikke fosforakkumulering i jord over langtidsperioden. Dette kan bety at en stor del av fosfor akkumuleres i et utilgjengelig lager for planter. Sammenligning av gjennomsnittstall over tre år for 1997-1999 og 2009-2011 på tvers av de tre regionene viste påvirkning av landbruksproduksjonssystem på fosforsstrømmer og fosforsbalanse. Den kornproduserende regionen Akershus ble funnet svært avhengig av mineralgjødselimport, mens region med husdyrproduksjon Rogaland akkumulerte store mengder fosfor fra husdyrgjødsel, men var avhengig av kraftfôrimport. Fosforbalansen i jord ble funnet positiv i alle tre regioner over hele tidsperioden. I den kornproduserende regionen Akershus gikk fosforbalansen i jord ned siden 1970-tallet, mens i husdyrproduserende regionen Rogaland økte den siden 1990-tallet og er nå høyest.

Høy avhengighet av mineralgjødsel- og kraftfôrimport med samtidig fosfor akkumulering i jord skaper en ubalansert situasjon med hensyn til fosfor. Fra et systemperspektiv er resirkulering av fosfor fra områder med høy husdyr tetthet til kornproduserende områder nødvendig for å oppnå en bedre grad av bærekraftighet.

Abstracts

Phosphorus is a non-renewable resource that is essential for food production. At the same time, phosphorus may cause environmental problems because excess phosphorus in agricultural soil often leads to eutrophication. For rational and sound phosphorus management in order to mitigate resource scarcity and eutrophication problems, reliable estimates of phosphorus pools and flows and the understanding of phosphorus soil dynamics are needed. Studies in Material Flow Analysis that consider soil phosphorus stocks are few and do not allow a reliable insight in soil phosphorus pools and their contribution to plant production as they mainly analyze global and national phosphorus flows.

This study aimed to assess long-term soil phosphorus accumulation in Norway by analyzing the regional phosphorus flows. Three Norwegian regions with contrasting agricultural production systems were chosen: Akershus with dominating cereal production, Rogaland with high livestock density and Sør-Trøndelag with mixed agriculture. Phosphorus flows and soil budget were quantified on a yearly basis from 1950 to 2011 and compared with estimates of available soil phosphorus based on the ammonium lactate soil phosphorus test. It was shown that the results of soil phosphorus test did not reflect soil phosphorus accumulation over years; this may indicate the accumulation of a large part of phosphorus in the unavailable pool. Comparison of the three-year average for 1997-1999 and 2009-2011 across the three regions showed the influence of agricultural production system on phosphorus flows and budget. The crop producing region Akershus was found highly dependent on mineral fertilizer import, while the region with high livestock density Rogaland accumulated large amount of phosphorus in manure but depended on input of feed concentrates. The phosphorus soil budget was found positive in all three regions over the entire time period. In the mainly crop producing region Akershus the phosphorus soil budget was decreasing since 1970s while it is increasing in the animal producing region Rogaland since 1990s.

High dependence on the inputs of mineral fertilizer and feed concentrates and simultaneous phosphorus accumulation in soil create an unbalanced situation with respect to phosphorus. From the system perspective, the recycling of phosphorus from the areas with high livestock density to crop-producing areas is necessary in order to achieve a better degree of sustainability.

Contents

List of Tables

List of Figures

List of Abbreviations

ADP	Adenosine diphosphate
ATP	Adenosine triphosphate
DNA	Deoxyribonucleic Acid
JOVA	Soil and water monitoring in agriculture (Jord- og vannovervåking i landbruket – *Norwegian*)
GMO	Genetically modified organism
MFA	Material flow analysis
NAPI	Net anthropogenic phosphorus input
P	Phosphorus
P-AL	Phosphorus – ammonium lactate test
RNA	Ribonucleic acid
SFA	Substance flow analysis
UMB	Norwegian University of Life Sciences (Universitetet for Miljø og Biovitenskap – *Norwegian*)

1. Introduction

1.1. The importance of phosphorus

Phosphorus is an essential element for all forms of life. As a component of nucleic acids, cell membrane phospholipids, ADP and ATP that supply cell processes with energy, and a number of enzymes and co-enzymes, phosphorus has no substitute and is therefore critical ingredient in primary production. Plant production is often phosphorus-limited, and this makes the availability of phosphorus in soils extremely important for all terrestrial ecosystems as well as agriculture.

The phosphorus cycle is not looped like other element cycles, e.g. carbon or nitrogen. To a great extend it is one-way flow from phosphate rocks to soil and then to lakes and oceans. At present terrestrial phosphorus cycle is dominated by human activities, especially agriculture (Oelker and Valsami-Jones, 2008; Shen et al., 2011). Human activity more then doubled global phosphorus mobilization compared to natural phosphorus flow due to weathering (Tilman et al., 1999; Smil, 2000; Bouwman et al., 2009). In the past phosphorus was recycled within farming systems, but in the 20th century food production became geographically separated from food consumption, as well as crop production from livestock production (Bateman et al., 2011). Urbanization, segregation of mixed-farming systems and “Sanitation Revolution” in 19th – early 20th centuries led to the change from phosphorus recycling society to phosphorus put-through society (Ashley et al., 2011; Schröder et al., 2011).

Food production accounts for 90% of global phosphorus consumption (Cordell et al., 2009; Neset and Cordell, 2011), 79% of phosphorus is used as fertilizer, 11% – for animal feed and food additives (Johnston and Steen, 2000; Bøen and Grønlund, 2008). 50-60% of all phosphorus supply comes from phosphate rocks (Smil, 2000).

Phosphorus is not a renewable resource. Easily accessible phosphate rock deposits are limited (Elser and Bennett, 2011). With today’s production rates these reserves are expected to be exhausted in 50-150 years (Smil, 2000; Cordell et al., 2009; Schröder et al., 2011), or, according to different forecasts, from 30-40 to 300-400 years (Cordell et al., 2012). Peak production is expected around year 2030 (Elser and Bennett, 2011). Potential phosphorus scarcity poses a danger for global food security. The problem can be worsened by increased growing of bio-energy crops. In addition, phosphate rock reserves are unevenly distributed: they are found mainly in Morocco, China and the US; while Western Europe and India are totally dependent on import (Cordell et al., 2009). However, currently the possible exhaustion of phosphate reserves is not the main issue, and restriction in fertilizer use and other measures designed to achieve more efficient phosphorus utilization are motivated by the negative impact of excessive phosphorus use on the environment rather then by resource scarcity (Schröder et al., 2010).

1.2. Eutrophication

Excess phosphorus may represent a serious environmental problem. Soil over-fertilization with phosphorus causes water pollution. Crops do not absorb all fertilizer applied to the soil, and excess fertilizer run-off causes increased phosphorus levels in surface water bodies (Carefoot et al., 2003). Elevated phosphate concentration in surface water (especially lakes) stimulates the growth of microalgae and cyanobacteria, and often causes eutrophication. Increased phosphorus input accelerates eutrophication of freshwater (Sharpley, 1994a, b), which leads to reduction of biodiversity and wildlife population due to oxygen shortage and degradation of drinking water quality. This causes increase of drinking water treatment costs and decrease of recreational benefits.

The phytoplankton growth had been shown to be proportional to phosphorus concentration in water (Schindler, 1977; 2012). The phosphorus flow to water is caused mostly by human activities like crop cultivation, animal husbandry, human excretion, household and industrial waste (Leeben et al., 2008; Han et al., 2013). Human activity increased global phosphorus flux from land to water 3-fold (Hovarth et al., 2002). Point source phosphorus inputs to water were greatly reduced during the last decades by improved wastewater treatment and detergent reformulation, while non-point sources, mainly nutrient leakage through soil erosion and run-off from agricultural land, especially in areas of intensive crop and livestock production, are of growing importance (Sharpley and Rekolainen, 1997; Sharpley, 1999; Gentry et al., 2007; Cordell et al., 2009; Elser and Bennett, 2011). A decrease of phosphorus input from point sources left agriculture the main contributor of phosphorus to many lakes in Norway (Bechmann et al., 2005a). Eutrophication due to excessive phosphorus discharge from agricultural soil is one of the most important environmental problems in some North American and European regions (Delgado and Torrent, 2001).

1.3. Phosphorus in soil

Soil is the principal reservoir of phosphorus accessible to life in terrestrial systems (Hesterberg, 2011). Filippelli (2002) estimated that 98% of phosphorus in a global soil / biota system is in soil. Plants take up phosphorus in orthophosphate form, mostly in the form of $H_2PO_4^-$, and less HPO_4^{2-} (Syers et al., 2008). Plants can also acquire phosphorus from organic sources through symbiotic Mycorrizae fungi (Schachtman et al., 1998).

Due to the highly reactive nature of phosphorus, only a small fraction of total phosphorus in soil is available for plant uptake; phosphorus readily binds to soil particles or other compounds and becomes biologically unavailable. The chemical reactions of phosphorus in soil are described in the Appendix 1. Thus, especially in agricultural areas with excessive fertilizer use, phosphorus is accumulating in soil in a form unavailable for plants. However, several case studies of phosphorus recovery and use efficiency in various soil types in different parts of the world have shown evidence that phosphorus applied in the form of fertilizer and manure is not irreversibly fixed in soil (Syers et al., 2008).

Three soil phosphorus pools are usually defined (Figure 1). (1) **Dissolved phosphorus** contains mostly dissolved orthophosphates: $H_2PO_4^-$ in acidic conditions ($pH<7$) and HPO_4^{2-} in alkaline conditions. A little amount of dissolved organic phosphorus may also be present (Hansen, 2002). (2) **Loosely bound / active / labile phosphorus** is in dynamic equilibrium with the soil solution and can be released relatively easy. Inorganic phosphorus in this pool exists in relatively soluble minerals or at soil ion-exchange sites, organic phosphorus is from relatively fresh organic material that can be easily decomposed (Hansen, 2002). (3) **Fixed / tightly bound / stable phosphorus** refers to very insoluble inorganic compounds, like crystalline Al and Fe compounds or Ca compounds, and organic compounds resistant to mineralization. This pool is in equilibrium with the other pools, and there exists a slow conversion from “fixed” to “active”, but it is usually considered too slow to be important for agricultural production (Hansen, 2002). One more pool – very strongly bound phosphate minerals – is sometimes included (Syers et al., 2008).

For model purposes only two pools – “labile pool” and “non-labile (stable) pool” – are often defined (Vadas et al., 2006, Sattari et al., 2012). Kreuzeder (2011) modeled three pools: inorganic-solution-organic.

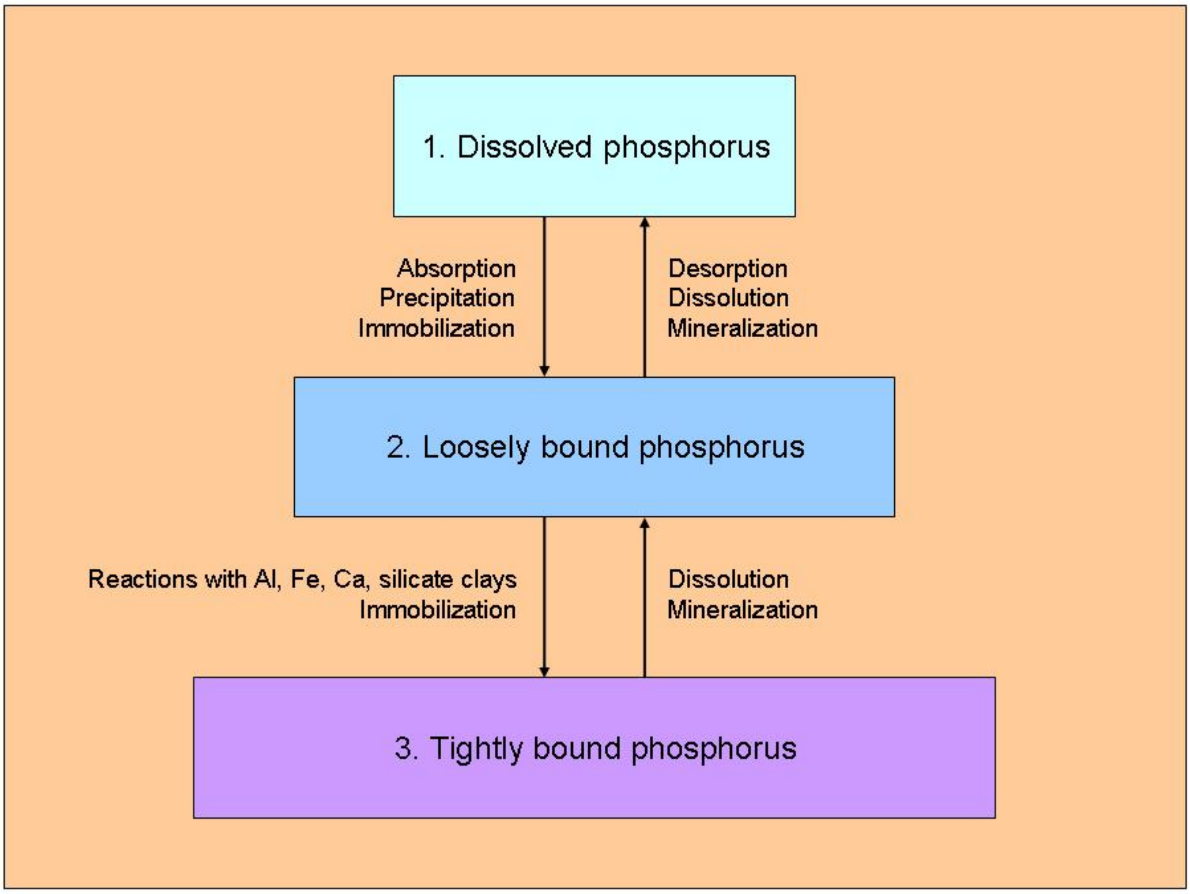

Figure 1. Soil phosphorus pools. Boxes represent soil phosphorus pools. "Dissolved" pool includes orthophosphates and little amount of dissolved organic phosphorus. "Loosely bound" pool includes inorganic phosphorus at soil ion exchange sites and easily decomposable organic phosphorus. "Tightly bound" includes insoluble organic compounds and recalcitrant organic compounds. Arrows represent conversion between pools.

1.4. Concepts of phosphorus dynamics in soil – the history of the question

Mid-nineteen century field experiments with fertilizer application in the United Kingdom showed that for achieving acceptable yield it was necessary to apply more phosphorus than was removed with harvested crops. It was observed that soil treated with phosphate fertilizer contained more soluble phosphorus than untreated. However, a part of the phosphorus budget could not be accounted for. As no downward transfer in the soil profile was found, it was concluded that part of the applied phosphorus was "fixed" in the topsoil. The process of phosphorus retention was attributed to calcium carbonate in calcareous soil and aluminium and iron oxides in acid soil (Syers et al., 2008).

In the mid-twentieth century it was suggested that phosphate ions were removed from the soil solution mainly by adsorption and, to less extend, by precipitation. However, it was concluded that adsorbed phosphorus was still available to plants (Syers et al., 2008). After 1950 the attention was paid mostly to phosphorus precipitation in soil. Low plant availability of phosphate fertilizer in many soil types was explained by rapid reaction with soil components. The fact that phosphorus would be adsorbed and absorbed on particulate matter was usually ignored (Syers et al., 2008).

In 1980s it was suggested that absorbed phosphorus could be released over time (Syers et al., 2008). Phosphorus output with crop yield can exceed amount of "available" phosphorus. In soil with high phosphorus content it may take many years to reduce soil test phosphorus to the level where crops respond to fertilizer application, as phosphorus is slowly released from "stable" to "available" pool (McCollum, 1991; Sharpley and Recolainen, 1997; Oehl et al., 2002; Dodd et al., 2012). In a long-term field experiment at Rotamshed (United Kingdom), increase in Olsen soil test phosphorus accounted for only 14% of the positive phosphorus budget during 45 years of fertilizer application, while decrease in Olsen soil test phosphorus accounted only for 36% of phosphorus removed with harvested crops during the following 73 years without phosphorus fertilizer application (Syers et al., 2008). So, it was finally understood that phosphorus "fixed" in soil could be recovered and taken up by plants over time (Syers et al., 2008).

In the most part of 20th century the emphasis was to determine the amount of nutrients necessary for optimum crop production, but last 20-30 years it turned to environmental impacts. In the future the main focus will be on producing economical yield with reduced nutrient input (Hochmuth, 2003).

1.5. Phosphorus management and fertilizer application

Concern about surface water quality motivated a number of measures in agricultural management in many North-American and European countries (Cordell et al., 2009), among them Norway (Ministry of the Environment, 1976; 1992; Bechmann et al., 2005a), in order to avoid over-fertilization. During the last two decades fertilizer use was decreased in Western Europe in general (Figure 2) and in Norway in particular (Figure 3). The decrease in mineral fertilizer use led to a decline in the soil phosphorus budget (Figure 4). The observed decline in phosphorus fertilizer use in industrialized countries became possible not only due to change in agricultural practices, but also due to soil phosphorus stock accumulation after many years of excessive fertilizer application.

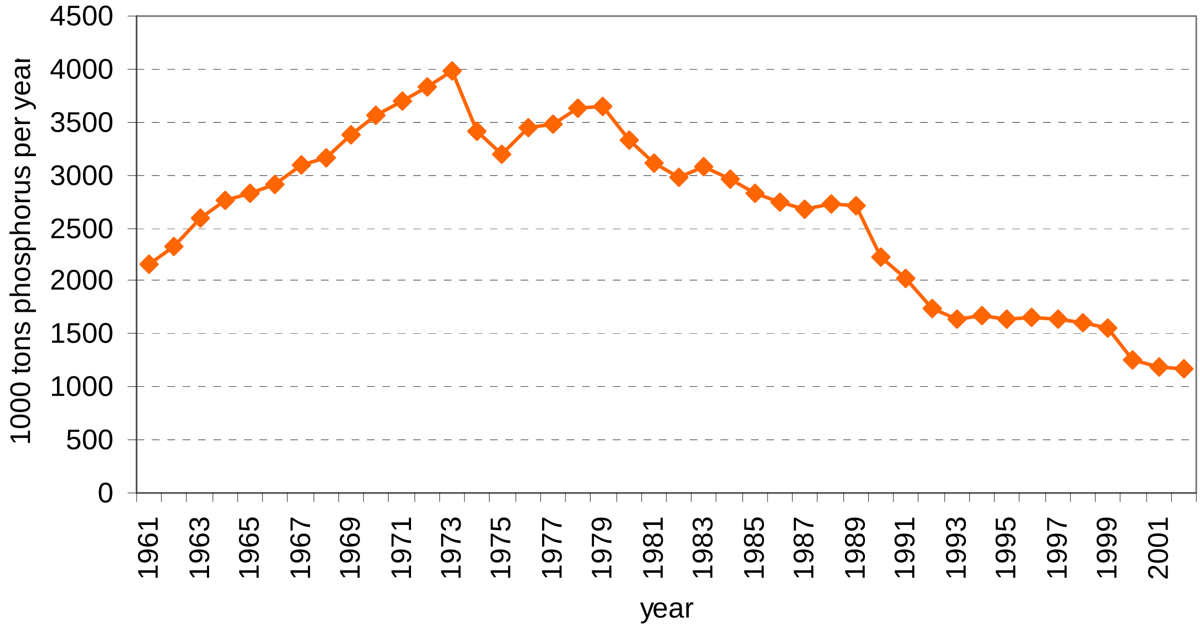

Figure 2. Phosphorus fertilizer use in Western Europe (data from FAOstat, 2013)

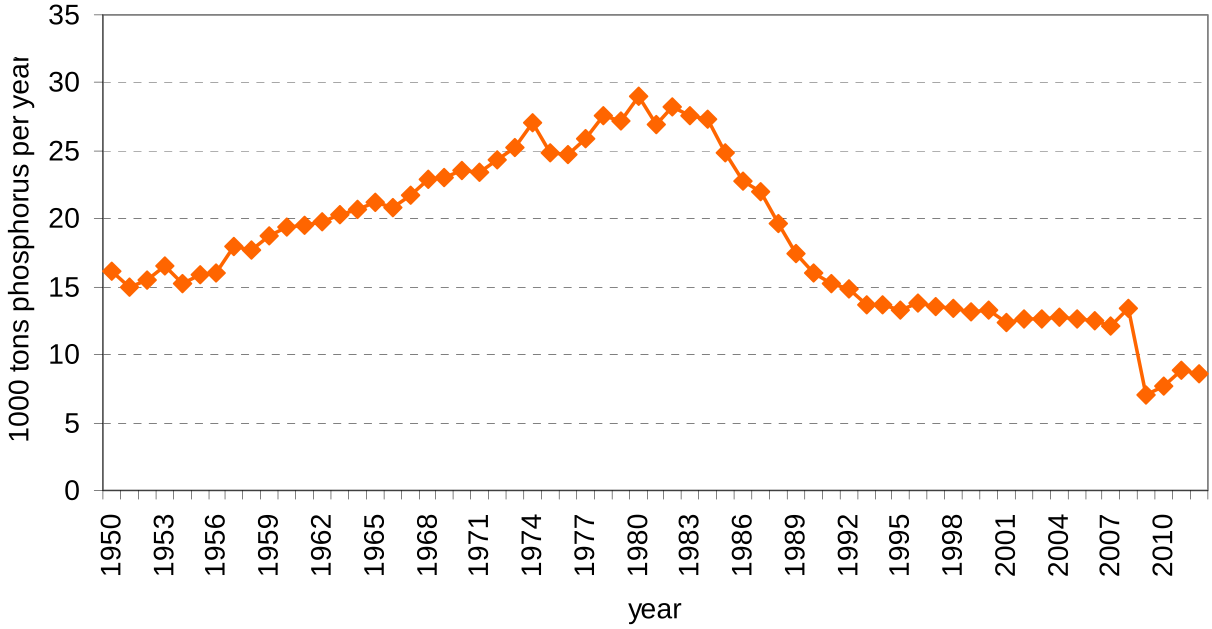

Figure 3. Phosphorus fertilizer use in Norway (data from Mattilsynet, 2012)

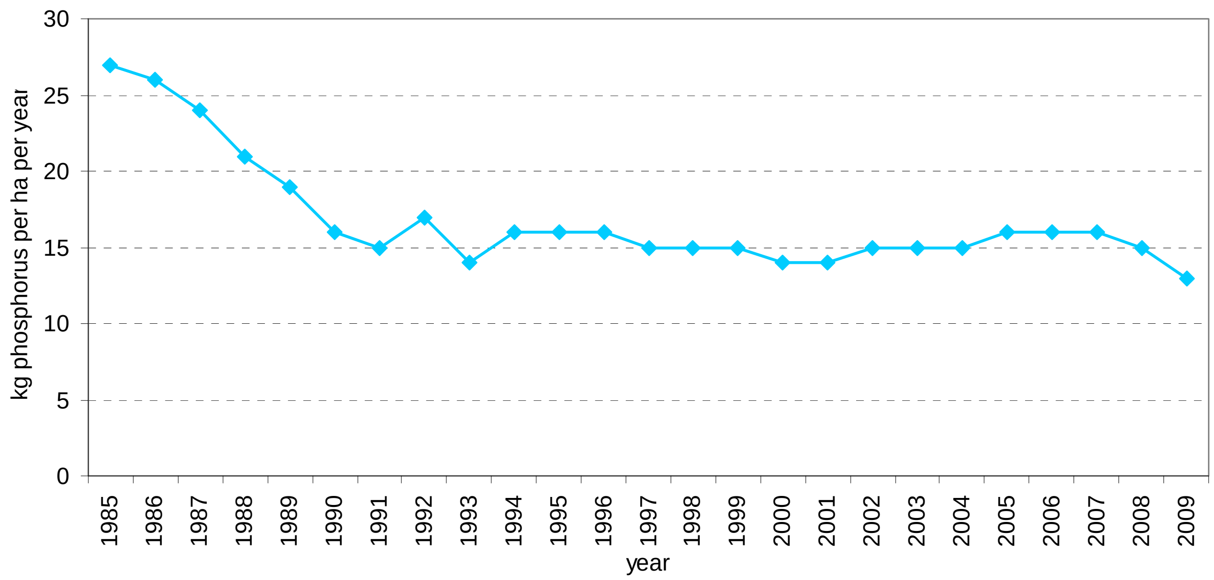

Figure 4. Soil phosphorus budget in Norway, kg phosphorus per ha per year (data from Eurostat, 2013)

Yield response to phosphorus fertilization is dependent on phosphorus content in soil and soil type. The main soil types, their properties and land use options are shown in the Table 2 (Appendix 2). After phosphorus content in soil reaches a certain level, crop yield ceases to respond to an increase in fertilization (McCollum, 1991; MacDonald et al., 2011). Due to long-term accumulation in European and North-American agricultural soil, phosphorus content is above this “critical level” and only small input is needed to replace phosphorus removed with harvest (Cordell et al., 2009; Neset and Cordell, 2012). New fertilizer recommendation with lower phosphorus doses are developed in many countries (Castoldi et al., 2009; Valkama et al., 2009; Litaor et al., 2013). Balanced fertilization strategy (addition of the same amount of phosphorus as removed by crops) was introduced in areas with medium-high phosphorus content in South-Eastern Norway as there was no need for a phosphorus surplus (Krogstad et al., 2008).

Fertilizer recommendations are based on phosphorus contents in soil. For practical agriculture a determination of the total amount of phosphorus in the soil is not meaningful. So, soil tests for plant-available phosphorus used in agriculture are designed not to determine the total phosphorus concentration in soil or even concentration of plant-available phosphorus, but to provide the index measurement of phosphorus that can be taken up by plants during growing season (Hansen et al., 2002). The important thing is the correlation between the phosphorus amount extracted by chemical extractant and phosphorus amount taken up by the plant. The extractant that shows the highest correlation is selected for the test (Watson and Mullen, 2007). Soil test phosphorus usually represents less than 5% of total soil phosphorus in unfertilized soils, and much higher percentage in the case of long-term fertilizer or manure application (Hansen et al., 2002). Several widely used soil phosphorus tests are shown in the Table 3 (Appendix 3). Different soil phosphorus tests were found to be well correlated with each other and with phosphorus tests used for environmental monitoring (Ebeling et al., 2003b).

Strong correlation was shown between the results of Mechlich and Bray-Kurtz (Ebeling et al., 2003b), Mechlich and P-AL (Bechmann, 2005b), and P-AL and Olsen methods (Mattson, 2008), though P-AL may be overestimated at high pH (Mattson, 2008). As chemical extractants in soil P tests simulate phosphorus availability, results of these tests can be used to estimate available / labile / active / loosely bound phosphorus soil pool.

1.6. Current research

For rational and sound phosphorus management, reasonable and reliable estimates of phosphorus pools and flows are needed. MFA studies that consider phosphorus stocks in soil or flows to and from soil are few, they differ in scope and methodology, and their results are sometimes contradictory.

Bouwman with colleagues (2009) analyzed global trends in phosphorus soil budgets over time. They calculated global phosphorus fluxes (fertilizer, manure, human waste, harvest and grazing, soil accumulation, erosion and leaching) for 1970, 2000 and 2050 for The Millennium Ecosystem Assessment scenarios (Bouwman et al., 2009). The global phosphorus input with inorganic fertilizer was estimated to be 8 Mt/yr, the one with manure 13 Mt/yr, soil accumulation (cropland + grassland) was 9 Mt/yr in 1970. In 2000 inputs and soil accumulation increased – 14 Mt/yr with inorganic fertilizer, 17 Mt/yr with manure, 12 Mt/yr accumulated in soil. Stocks were not considered in this study (Bouwman et al., 2009). Cordell with colleagues (2009) studied global phosphorus flows through the food system with respect to phosphorus peak, global food security and recycling options. The global fertilizer input was estimated to be 14 Mt/yr, manure 13 Mt/yr, considerably less than estimated by Bouwan et al. (2009). As a consequence, the soil accumulation (inputs less outputs) in “arable soil” was estimated 4.5 Mt/yr around year 2000 (Cordell et al., 2009), also much less than in (Bouwman et al., 2009). Erosion and run-off losses were 8 Mt/yr (Cordell et al., 2009). Liu with colleagues (2008) also analyzed global phosphorus flows for the year around 2000 and summarized the available information about global total and available phosphorus soil pools. Flows were calculated for global cropland (arable land). Inorganic fertilizer inputs was 13.8 Mt/yr, almost the same as in two studies discussed above, but the input from recycled plant residues, manure and human waste together was only 6.2 Mt/yr. Due to outputs in harvest (12.7 Mt/yr) and erosion/run-off losses from cropland (19.3 Mt/yr), and permanent pastures (17.2 Mt/yr) the resulting global budget is highly negative (10.5 Mt/yr net losses from global cropland) (Liu et al., 2008).

Studies on national phosphorus flows are difficult to compare. Suh and Yee (2011) estimated the life-cycle phosphorus use efficiency of the US food system and calculated P flows for 2007 using mass balance. They did not examine soil as such, but inputs to and outputs from "crop cultivation" process included "fertilizer use" (1810 Kt/yr), and "waste to soil" (672 Kt/yr). No erosion or run-off losses were assumed, and no soil stock considered (Suh and Yee, 2011).

Matsubae-Yokoyama with colleague (2009) analyzed Japanese phosphorus flows for the year 2002 and found that over 90% of fertilizer input to agriculture was accumulated in the soil, but soil stock was not quantified (Matsubae-Yokoyama et al., 2009). Over 80% of phosphorus extracted in China in 1984-2008 was "lost to natural water and soil", and agricultural soil stock was estimated 38.3 Mt (Ma et al., 2012). Yuan with colleagues (2011) assessed in- and outflows to and from agricultural soil and soil phosphorus stock in China in the year 2008. Soil stock was calculated as a sum of all inflows multiplied by 0.3 – "efficiency of soil sediment" (Yuan et al., 2011). Han et al. (2013) found upward trend in net anthropogenic phosphorus inputs (NAPI) from 1981 to 2009 in Mainland China. Inorganic fertilizer accounted for 57-84% of net anthropogenic phosphorus input (Han et al., 2013).

In the study of nutrient stocks and flows in the Finnish food production and consumption system in- and outflows to and from agricultural soil in 1995-1999 were estimated, as well as stock of plant available phosphorus, but not total phosphorus in soil. The phosphorus surplus (inputs less outputs including losses) was 12.7 kg P/ha-yr on agricultural land (Antikainen et al., 2005). Neset with colleagues (2008) analyzed phosphorus flows based on food production and consumption of an average inhabitant of Linköping (Sweden) in 1870, 1900, 1950 and 2000. There was found increase in chemical fertilizer input, in the flow from animal production and, as a consequence, in the flow reaching the consumer and then waste management system (Neset et al., 2008). Phosphorus flows in agriculture and food system in Sweden in 2008-2010 were studied with the main focus on overall system (Linderholm et al., 2012). Balance for agricultural soil was not analyzed, as "plant cultivation" and "animal husbandry" belonged to the same process. The net phosphorus inputs to soil in Sweden were estimated to be 4.1 kg/ha for all agricultural land (including pasture) (Linderholm et al., 2012). The study of phosphorus flows in France (Senthilkumar et al., 2012a; 2012b) showed that the national soil phosphorus budget was reduced from 18 kg/ha-yr in 1990 to 4 kg/ha-yr in 2006, mainly due to reduction in inorganic fertilizer application.

The analysis on the regional level in France demonstrated the dependence of phosphorus flows and budgets on agricultural production system: the region with dominating crop production was found heavily depending on fertilizer use, while the animal farming region accumulated phosphorus in soil (Senthilkumar et al., 2012b). The soil stocks were not calculated in this study due to difficulties in obtaining data on the total phosphorus content. Neither was resulting phosphorus soil accumulation discussed though soil phosphorus budget was calculated for 16 years (Senthilkumar et al., 2012b). However, the regional approach may be useful in assessment of phosphorus soil stocks especially when it is possible to analyze regions with contrasting production system.

1.7. Aim and scope of the thesis

A pre-study was conducted during the autumn 2012 term with the goal to assess phosphorus stocks in soil (Zabrodina, 2012). The phosphorus fluxes and stocks were calculated for US cropland for one year around 2008. The results indicated that phosphorus amounts in fertilizer and harvested crop were of same order of magnitude as soil phosphorus in available / labile / loosely bound pool. It may serve as a confirmation of the conversion from "tightly bound" pool to "labile / available" pool.

The pre-study has revealed the necessity of the use of less aggregated data, like regional-scale phosphorus flow data, for demonstrating phosphorus soil saturation and assessment of contribution of "fixed" phosphorus to plant nutrition. Another reason to use the less aggregated data is to avoid large uncertainties. In addition to cropland the analysis should include grassland, as inorganic fertilizer may be used for both. Further, to observe changes in phosphorus flows and stocks over time, the flows should be analyzed for the period of several decades.

The present study aimed to address the topic of phosphorus dynamics in soil using Material / Substance Flow Analysis by characterization of phosphorus stocks and fluxes in regions with different agricultural production systems and their development over time.

The specific research questions were:

1. What are the main phosphorus stocks and fluxes in Norwegian regions with contrasting agricultural production systems (Akershus, Sør-Trøndelag and Rogaland)?
2. What are the main changes in phosphorus flows and stocks in regions with different agricultural production systems?
3. How do the type of agricultural production system and fertilizer use during the last 60 years has influenced soil phosphorus accumulation in the study areas?
4. Can results of the P-AL soil phosphorus tests reflect phosphorus soil accumulation over time?
5. What are possible implications of the current situation and trends in soil phosphorus stocks and flows for phosphorus management in Norway?

2. Methods

2.1. System definition

2.1.1. The study area and timeframe

The system boundaries are set as agricultural system in a Norwegian county; one year (average for 1997-1999 and 2009-2011). Agricultural land (jordbruksareal – *Norwegian*) is defined as the land used for crops, cultivated pastures and permanent grassland (including fertilized pastures even if not completely cleared), as well as lawn and gardens (SSB, 1974; 1982). Uncultivated land (utmark – *Norwegian*) is outside the agricultural system.

Three Norwegian regions were chosen: (1) Akershus with dominating cereal production, (2) Rogaland with high livestock density and dominating grass production and (3) Sør-Trøndelag with mixed agriculture. In Akershus over 77% of agricultural land is dedicated to cultivation of cereals, and this county accounts for more than 20% of national cereal production, while Rogaland accounts for considerable part of the Norwegian livestock: 17% cattle, 28% pigs, over 21% sheep, and over 29% poultry; 94.5% of agricultural land in Rogaland are meadows for mowing and pasture (SSB, 2012). Sør-Trøndelag accounts for 5.3% of national cereal production, 9% of the Norwegian cattle livestock, 6.4% pigs and 5% poultry, having 23% of agricultural area used for cereals and 75% as meadows (SSB, 2012).

Phosphorus fluxes and budgets in agricultural system were independently calculated for three Norwegian counties on a yearly basis from 1996 to 2011. Three-year average values were calculated for the years 1996-1999 and 2009-2011. In addition, the phosphorus budget for soil was quantified for every year from 1950 to 1995, in order to estimate the lower border of total phosphorus accumulated in soil over last 60 years.

2.1.2. System design

Agricultural system includes four processes: (1) “Soil” (2) “Plant Production” (3) “Feed and Fodder Market”, and (4) “Animal Husbandry” (Figure 5).

The process “Soil” comprises the fluxes of phosphorus containing goods through soil and phosphorus soil stock. Inputs are phosphorus in (1) mineral fertilizer, (2) composted waste, (3) sewage sludge applied to agricultural land and (4) animal manure applied to agricultural land or dropped at pastures. Phosphorus inputs in seeds and atmospheric deposition are considered to be negligible. Outputs are (1) phosphorus taken up by plants, including harvested crops and grass consumed through grazing and (2) phosphorus lost due to soil erosion and leaching. Phosphorus in plant residue left on fields is assumed to be staying in the soil.

The process “Plant Production” includes one input – phosphorus taken up by plants: harvested crops (excluding plant residue left on fields) and grass consumed through grazing, and three outputs: (1) phosphorus in crop products exported from agricultural system and (2) phosphorus in fodder and (3) phosphorus consumed through grazing. No stock is assumed.

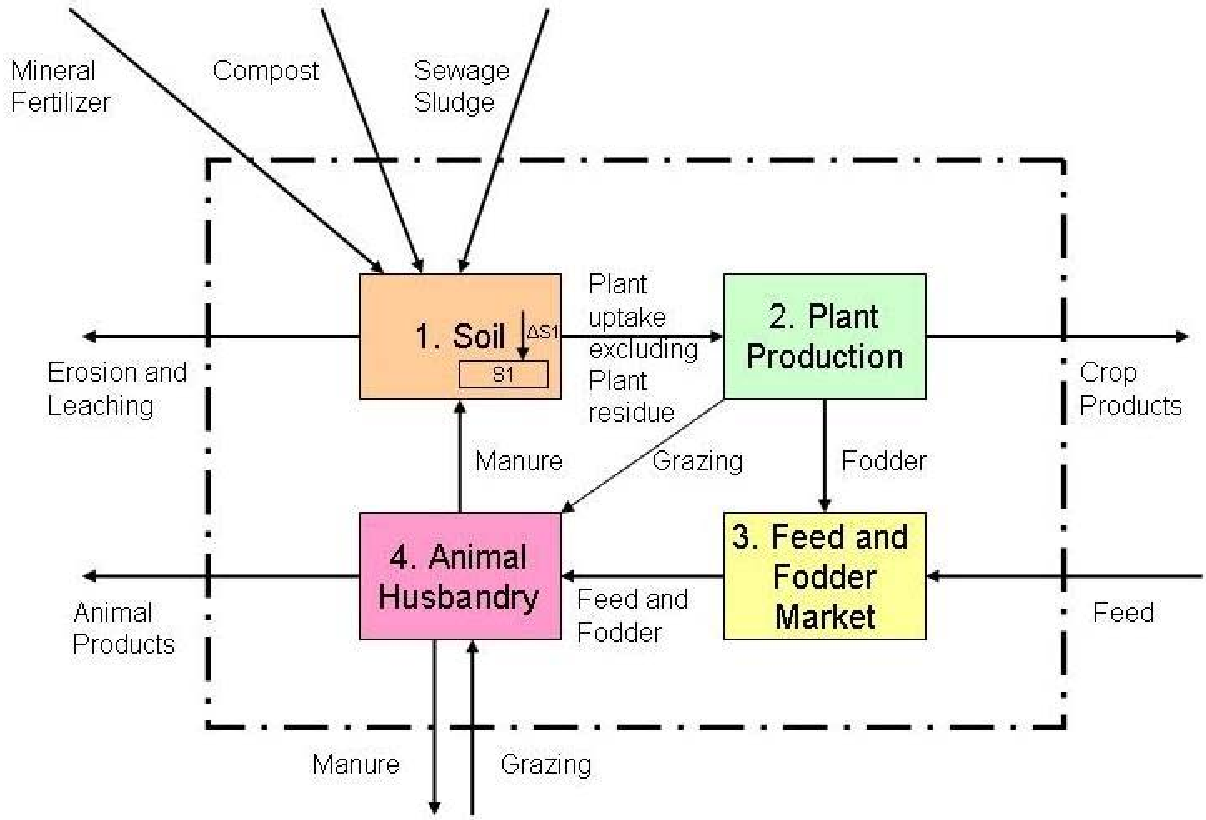

Figure 5. System for phosphorus flow analysis at the county scale (conceptual design). The boxes represent processes. The arrows represent phosphorus fluxes.

The process "Feed and Fodder Market" consists of inputs of phosphorus in (1) fodder and (2) imported feed, and output of phosphorus consumed in feed and fodder. No losses are assumed. No stock is assumed.

The process "Animals Husbandry" (Figure 6) includes horses, ruminants (cattle, sheep and goats), pigs and poultry (hens and chicken). Fur animals, rabbits and turkeys are considered to be negligible. The process comprises input of phosphorus consumed in (1) feed and fodder and (2) through grazing, both on cultivated (inside agricultural area) and uncultivated land (outside agricultural area), and outputs: (1) animal manure to "Soil", both inside and outside agricultural area, and (2) animal products: carcasses of slaughtered animals, milk and eggs; while wool and fur are considered to be negligible. Phosphorus stock change in the process "Animal Husbandry" was not accounted for.

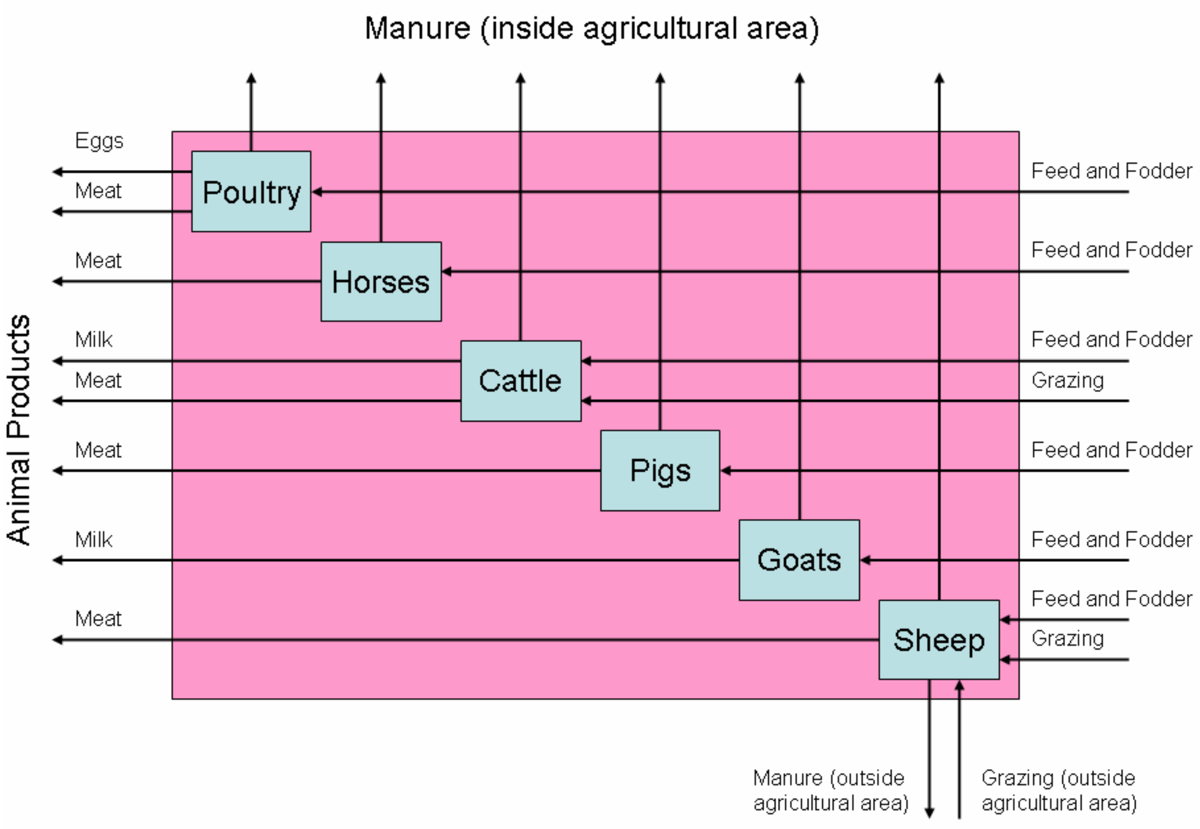

Figure 6. Sub-processes in the process "Animal Husbandry". The boxes represent sub-processes (animal groups). The arrows represent phosphorus fluxes.

2.2. Data sources and quantification methods

Data for agricultural area, crop yield and livestock of domestic animals are gathered from Statistics Norway (SSB, 1974; SSB, 1974-1996; SSB, 1982; SSB, 1995; Rognstad and Steinset, 2011; SSB, 2012; StatBank, 2013). Data for sales of mineral fertilizer were obtained from the Norwegian Food Authority (Mattilsynet, 2010; 2011; 2012; Bøen, 2013) and YARA (Nyhus, 2013). Data for sales of feed concentrates and raw products used for feed production were provided by the Norwegian Agricultural Authority (SLF) (Breen, 2013). Data for phosphorus concentration in crops, fodder and animal products (slaughtered animals, milk and eggs) are taken from scientific literature and the Norwegian food product tables (Antikainen et al., 2005; Matvaretabellen, 2013). Phosphorus concentration in mineral fertilizer and feed was calculated using data from the Norwegian Food Authority (Mattilsynet, 2010; 2011; 2012; Bøen, 2013) and the Norwegian Agricultural Authority (Breen, 2013) respectively. Nutritional requirements of domestic animals were found in Merck Veterinary Manual (Merck, 2013). Data on phosphorus excretion in animal manure are taken from UMB report (Karlengen et al., 2012) and a number of other sources (Tveitnes, 1993; Bolstad, 1994; Øgaard, 2008; Knutsen and Magnussen, 2011; Nesheim et al., 2011; Daugstad et al., 2012; Bioforsk, 2013; Lovdata, 2013).

MFA (SFA) was used to assess phosphorus fluxes and phosphorus stock in soil. Stocks and flows are expressed in kg elementary phosphorus per hectare agricultural land (for stocks) or kg elementary phosphorus per hectare agricultural land per year (for fluxes) unless otherwise noted.

2.3. Quantification of phosphorus fluxes

The parameters, variables and equation used for quantification of phosphorus fluxes and soil stock are shown in Table 4, Table 5 and Table 6 (Appendix 4). The analytical solution is shown in the Appendix 5.

2.3.1. Mineral fertilizer

The data of the phosphorus fertilizer amount sold in 1996-2011 were obtained from the Norwegian Food Authority (Mattilsynet, 2009; 2010; 2011; Bøen, 2013; Nyhus, 2013). Phosphorus concentrations in different fertilizer products calculated based on the data at the national level from the Norwegian Food Authority (Mattilsynet, 2009; 2010; 2011; Bøen, 2013; Nyhus, 2013) are shown in the Table 7 (Appendix 6). Phosphorus content in all fertilizer products sold in every county was calculated for every year from 1996 to 2011 (e.g. Table 8, Appendix 6 for 2011) and summed up to give phosphorus input with mineral fertilizer.

No county data for 1950-1995 were available. To estimate phosphorus input with mineral fertilizer for this period it was assumed that the share of every county in national fertilizer consumption was equal to their share in 1996-2011 and constant over time: 10.6% for Akershus, 3% for Rogaland and 6.4% for Sør-Trøndelag.

2.3.2. Waste composted

Phosphorus input with composted waste in 1996-2011 was calculated using the statistical data of waste sent to biological treatment (StatBank, 2013). According to Briseid et al. (2010), phosphorus content in food waste is 0.4% of dry matter, while dry matter content is 30-40%. Therefore, phosphorus content in organic waste sent to biological treatment was assumed 0.15%. For 1950-1995 this flow was not quantified due to data unavailability. However, it may be considered negligible due to low proportion of this flow in phosphorus input to soil quantified for 1996-2011.

2.3.3. Sewage sludge used in agriculture

To estimate phosphorus input by sewage sludge the data of phosphorus discharge by county were used for 2001-2011 (StatBank, 2013). For 1996-2000 discharges were assumed to be in the same range as later – 50 tons/year for Akershus, 30 tons/year for Rogaland, and 90 tons/year for Sør-Trøndelag. The average phosphorus removal in 2011 was 92.27% in Akershus, 52.08% in Rogaland, and 46.45% in Sør-Trøndelag (Berge and Mellem, 2012). Based on this, 90% for Akershus, 50% for Rogaland, and 45% for Sør-Trøndelag were assumed for 1996-2010. In Norway 43-64% of sewage sludge was used for soil fertilization or soil improvement during 2001-2011 (Berge and Mellem, 2012), but this may vary from county to county. Therefore it was assumed that 50% of collected sewage sludge was applied to agricultural land. For 1950-1995 this flow was not quantified as data of phosphorus discharge were not available.

2.3.4. Animal manure

Phosphorus excretion with manure was quantified for every year as a product of number of animals (SSB, 1974; SSB, 1974-1996; SSB, 1982; SSB, 1995; StatBank, 2013) and phosphorus excretion per animal or animal place per year (Karlengen et al., 2012; Bolstad, 1994) for all groups of animals except lambs. The comparison of data on phosphorus excretion is shown in the Table 9 (Appendix 7). The amount of lamb manure is calculated according to mass balance principle as a difference between nutritional requirement and phosphorus in slaughtered sheep. The average slaughter age for lambs in Norway is 160 days (Ringdal et al., 2012). Sheep grazing on uncultivated land is 3 months in average (Karlengen et al., 2012). Therefore, lambs are assumed to graze and excrete their manure 90 days outside agricultural land and 70 days inside agricultural area.

2.3.5. Losses with erosion and leaching

The JOVA (soil and water monitoring in agriculture) project estimated phosphorus losses due to erosion and leaching in 1992-2009 at a number of sites in different parts of Norway (Rød et al., 2009; Ulen et al., 2012). However, results for a specific site may be not representative for the county. As phosphorus losses were reported to be 0.3-2.6 kg phosphorus per ha per year (Ulen et al., 2007), or 0.35-1.86 kg/ha per year, ca.1±0.8 kg/ha per year in average in 2008/2009 (Rød et al., 2009; Ulen et al., 2012), phosphorus losses in the present study were assumed to be 1 kg/ha per year for all three counties throughout the whole analyzed period 1950-2011.

2.3.6. Animal Products

For 1996-2011 phosphorus in animal products as cow milk, eggs, and the whole carcasses of slaughtered horses, cattle, pigs, sheep and broilers / hens were calculated using statistical data of quantities of products (StatBank, 2013) and phosphorus concentrations in these products (Antikainen et al., 2005; Matvaretabellen, 2013). Internal flows, like milk fed directly to the calves that ends up in meat and manure, were not calculated separately. For 1950-1995 phosphorus output in animal products was not quantified, as the data on slaughtered animals were not available on the county level.

2.3.7. Feed concentrates

For 1996-2011 phosphorus input in feed concentrates was calculated using data of sales of feed by county from Norwegian Agricultural Authority (Breen, 2013) and average concentration in feed was estimated using data of raw products sold for feed production from Norwegian Agricultural Authority (Breen, 2013) as well as phosphorus content in the products (Matvaretabellen, 2013; Nutritiondata, 2013; Stein, 2013; Tangkanakul et al., 2005; Hertrampf and Piedad-Pascual, 2000). The calculation of concentration in feed concentrates in 2010 is shown in the Table 10 (Appendix 8). For 1950-1995 phosphorus input in feed concentrates was not quantified, as there were no available data on the county level.

2.3.8. Phosphorus consumed by domestic animals through feed concentrates, fodder and grazing.

Phosphorus intake by animals in 1996-2011 was quantified using mass balance principle as a sum of phosphorus excreted with animal manure and phosphorus in animal products.
No county data of animal production are available for 1950-1995, so phosphorus intake was estimated using nutrient requirements of domestic animals (Heje, 1974; 1992; 2000; Volden, 2011; Merck, 2013). Lambs are assumed to graze 90 days on the uncultivated land and 70 days on the cultivated land (see section 2.3.4). Milk consumed inside process “Animal Husbandry” is not accounted for (see section 2.3.6).

2.3.9. Plant uptake

Plant uptake is quantified as a sum of phosphorus in wheat, barley, oats, rye and triticale, potato, green fodder and silage, and hay harvested (SSB, 1974; SSB, 1974-1996; SSB, 1982; SSB, 1995; StatBank, 2013) and phosphorus taken up through grazing on the cultivated land. Plant residue is excluded as it is assumed to be left on fields. Grazing share in nutrition is found in statistical data (SSB, 1974; SSB, 1974-1996; SSB, 1982; SSB, 1995; StatBank, 2013; Tine, 2013) for cattle, assumed negligible for goats, as goats constitute a little proportion of animal stock even in Rogaland, and assumed 50% for sheep (4-5 months out of 9 months on the cultivated land).

2.3.10. Soil stock change

Soil stock change for the whole period from 1950 to 2011 was quantified using mass balance.

$$Soil\_phosphorus\_budget = \sum inputs - \sum outputs$$

Net accumulation was quantified as a sum of all soil stock changes during the whole period from 1950 to 2011.

$$Net\_accumulation = \sum_{1950}^{2011} Soil\_phosphorus\_budget$$

2.3.11. Fodder and crop products

Phosphorus fluxes from plant production to feed and fodder market (fodder) and export of crop products were quantified using mass balance for 1996-2011 (equations 3 and 2 respectively, Table 6, Appendix 4). It was not possible to quantify these fluxes for 1950-1995 due to absence of data on imported feed.

2.4. Available phosphorus soil stock

In Norway the content of plant available phosphorus in soil is estimated using P-AL (ammonium-lactate) method (Krogstad et al., 2008). The results of P-AL analysis were taken from (Krogstad, 1987) for 1960-1985 for Romerike (Akershus) and Jæren (Rogaland), and obtained from Bioforsk Jordsdatabanken for 1988-2011 for Akershus, Rogaland and Sør-Trøndelag (Grønlund, 2013).

To convert results of P-AL analysis (expressed in mg P per 100 g soil) to phosphorus soil stock (expressed in kg per ha), 20 cm depth and 1.2 t/m^3 soil bulk density were assumed due to following reasons. Phosphorus concentration in soil drops considerably with depth, and is very low at depths exceeding ca. 15 cm (Cole et al., 1977). This applies both to total P, Mehlich III P, and organic matter content (Curtis et al., 2010). Therefore 20 cm depth was chosen for soil phosphorus stock quantification. The numbers of soil bulk density from different data sources vary considerably: 1.1 t/m^3 for clay, 1.3 for silt loam, 1.4 for loam, 1.6 for sandy soil (Agriinfo, 2013), 1.2 t/m^3 for loose earth (Engineering toolbox, 2013), 965-1035 kg/m^3 for grassland soil (Stroia et al., 2007), 800 kg/m^3 for top 25 cm of agricultural land (Johnston and Steen, 2000; Liu et al., 2008), 1.2-1.3 t/m^3 for average Norwegian soil (Bleken, 2012; Krogstad, 2013). Here the soil bulk density is assumed to be 1.2 t/m^3. Then the top 20 cm will contain on average 2400 ton soil per ha.

2.5. Uncertainty propagation

The information used for modeling of phosphorus fluxes in the present study came from different sources and was of different quality. To account for differences in the information quality, the standard deviation was calculated for each of the parameters, based on the assumptions on relative errors. Most of statistical data (agricultural area, phosphorus fertilizer use, waste sent to biological treatment, phosphorus discharge to water, animal livestock, slaughtered animals, crop harvest, feed sold) were assumed relatively certain, as well as dry matter content, and phosphorus content in crops. For these parameters relative errors were set as 10%. Such parameters as phosphorus removal by waste water treatment, proportion of sewage sludge used in agriculture, phosphorus concentrations in animals, feed and waste and phosphorus excretion with manure, included a number of assumptions some aggregation. For these parameters the relative error was set as 20%. For the data of nutrient requirements that were not county-specific and not always country- and time-specific, relative errors were set as 30%. The most uncertain were lamb's nutrient requirements; for this 50% relative error was assumed. As Norwegian phosphorus erosion losses were 1±0.8 kg/ha per year (Rød et al., 2009; Ulen et al., 2012), 1 kg/ha-yr was used as a mean value with 80% relative error.

To incorporate the propagation of uncertainties into the calculated MFA results, the numerical approach (Monte-Carlo simulation) was used. The a priori values and uncertainties of phosphorus fluxes and soil stock were calculated based on the mean value and standard deviation of each parameter. The normal probability distribution was assumed and 2000 iterations were used.

3. Results

3.1. Phosphorus flow analysis for three Norwegian regions with different agricultural production systems

The phosphorus fluxes and soil stock change for three Norwegian counties are shown in the Figure 7. Average results for 1997-1999 show that in all three areas the main imports to regional agricultural systems were mineral fertilizer (18.8 kg/ha-yr in Akershus, 10.6 kg/ha-yr in Sør-Trøndelag, 4.6 kg/ha-yr in Rogaland) and feed concentrates (4.7 kg/ha-yr in Akershus, 10.6 kg/ha-yr in Sør-Trøndelag, 22.3 kg/ha-yr in Rogaland). The main exports were crop products (10.3 kg/ha-yr in Akershus, 2.7 kg/ha-yr in Sør-Trøndelag, 4.0 kg/ha-yr in Rogaland) and animal products (1.4 kg/ha-yr in Akershus, 3.7 kg/ha-yr in Sør-Trøndelag, 5.6 kg/ha-yr in Rogaland). For Rogaland, the fluxes of phosphorus consumed through grazing and excreted with manure outside agricultural land ("utmark") were also quite high (2.4 kg/ha-yr and 1.6 kg/ha-yr respectively). The most important phosphorus input to soil in addition to mineral fertilizer was animal manure (5.7 kg/ha-yr in Akershus, 18.6 kg/ha-yr in Sør-Trøndelag, 29.0 kg/ha-yr in Rogaland). Inputs with composted waste and sewage sludge can be considered negligible as they are both under 1 kg phosphorus per ha per year. The most important output from soil was plant uptake (12.5 kg/ha-yr in Akershus, 14.2 kg/ha-yr in Sør-Trøndelag, 15.6 kg/ha-yr in Rogaland). The main input to animal husbandry was feed and fodder (6.2 kg/ha-yr in Akershus, 18.0 kg/ha-yr in Sør-Trøndelag, 27.5 kg/ha-yr in Rogaland), grazing was also important for Sør-Trøndelag (4.1 kg/ha-yr) and Rogaland (6.3 kg/ha-yr).

There were noticeable differences in fluxes depending on the type of agricultural production system in the region. Akershus, a region with the dominant crop production, had the largest mineral fertilizer input and the smallest animal manure input to the soil. Feed concentrates input was the lowest, while the output of crop products was the highest in this county. Rogaland, the area with the highest livestock density, had the smallest mineral fertilizer input and largest animal manure input to the soil. The inputs with feed concentrates (22.3 kg/ha-yr) and through grazing (6.3 kg/ha-yr on the cultivated land + 2.4 kg/ha-yr on the uncultivated land) were higher then those both in Akershus (4.7 kg/ha-yr and 0.8 kg/ha-yr respectively) and Sør-Trøndelag (10.6 kg/ha-yr and 4.1 kg/ha-yr respectively). Over 80% of phosphorus taken up by plants in Akershus, less then 20% in Sør-Trøndelag and 25% in Rogaland was exported as plant products.

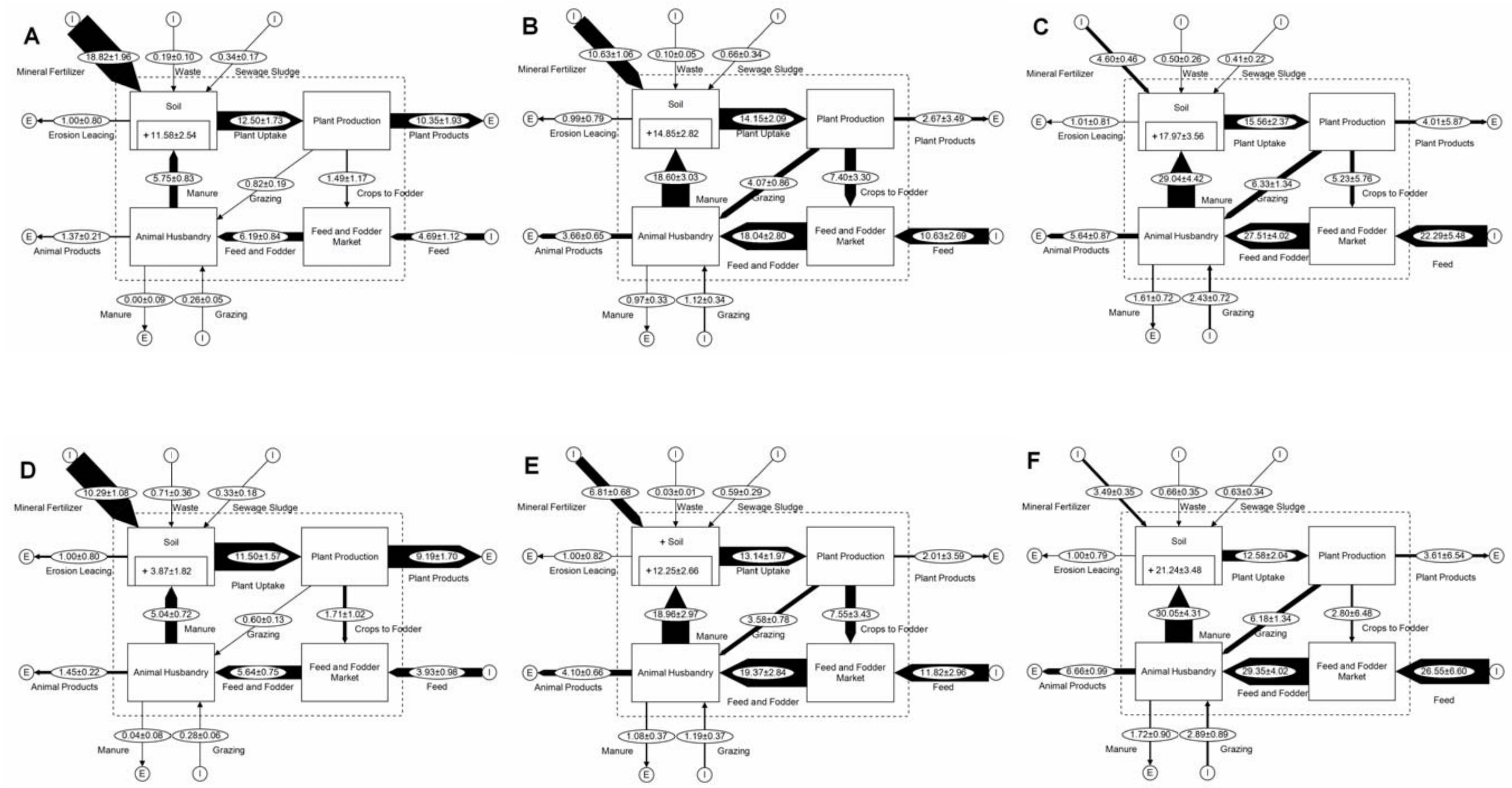

Figure 7. Phosphorus fluxes and soil stock in three different Norwegian regions (A, D – Akershus; B and E – Sør-Trøndelag; C and F – Rogaland). All values in kg phosphorus per ha per year, averaged for 3 years (A, B, C – 1997-1999; D, E, F – 2009-2011).

Table 1. Comparison of the main phosphorus fluxes and soil phosphorus stock change in Akershus, Sør-Trøndelag and Rogaland between two time points.

	Akershus			Sør-Trøndelag			Rogaland			
Flux/stock	1997-1999	2009-2011	Change	1997-1999	2009-2011	Change	1997-1999	2009-2011	Change	Unit
Mineral Fertilizer	18.8	10.3	-45%	10.6	6.8	-36%	4.6	3.5	-24%	kg P/ha-yr
Crop Yield	12.5	11.5	-8%	14.1	13.1	-7%	15.6	12.6	-19%	kg P/ha-yr
Crop Products	10.3	9.2	-11%	2.7	2.0	-25%	4.0	3.6	-10%	kg P/ha-yr
Crops to local Fodder Market	1.5	1.7	15%	7.4	7.5	+2%	5.2	2.8	-46%	kg P/ha-yr
Imported Fodder to local Fodder Market	4.7	3.9	-16%	10.6	11.8	+11%	22.3	26.5	+19%	kg P/ha-yr
Grazing (uncultivated land)	0.3	0.3	+8%	1.1	1.2	+6%	2.4	2.9	19%	kg P/ha-yr
Grazing (cultivated land)	0.7	0.6	-9%	4.1	3.6	-12%	6.3	6.2	-2%	kg P/ha-yr
Fodder to Animal Production	6.2	5.6	-9%	18.0	19.4	+7%	27.5	29.3	+7%	kg P/ha-yr
Animal Manure to Soil (cultivated land)	5.7	5.0	-12%	18.6	19.0	+2%	29.0	30.0	+3%	kg P/ha-yr
Animal Products	1.4	1.4	+6%	3.7	4.1	+12%	5.6	6.7	+18%	kg P/ha-yr
Soil Stock Change	11.6	3.9	-67%	14.8	12.2	-18%	18.0	21.2	+18%	kg P/ha-yr

Soil phosphorus budget was positive in all three counties for both time points (1997-1999 and 2009-2011) and varied depending on agricultural production system. It was 11.6 kg/ha-yr in Akershus, 14.8 kg/ha-yr in Sør-Trøndelag and 18.0 kg/ha-yr in Rogaland. Due to long time application of mineral fertilizer in excess to plant uptake (from 1950 to 2007, data not shown), the highest net accumulation in the soil was in Akershus (ca. 930 kg/ha by 1997-1999). Rogaland had much lower net accumulation (ca. 480 kg/ha).

The comparison between phosphorus fluxes average for 1997-1999 and 2009-2011 (Figure 7, Table 1) reveals the development in agriculture in these three counties over last years. The most notable change is a decrease in mineral fertilizer use by 45% in Akershus, 36% in Sør-Trøndelag and 24% in Rogaland. Feed concentrate input is still high in Sør-Trøndelag and Rogaland, where it increased by 11% and 19% respectively. In Akershus phosphorus soil budget is now 3 times lower then 12 years ago, mostly due to reduced mineral fertilizer use and 12% less phosphorus in manure, though output from soil through uptake by plants decreased by 8%. Import of feed concentrates and export of crop products decreased by 16% and 11% respectively in Akershus. In Sør-Trøndelag soil budget decreased as well, though not so much – by 18%, due to decrease in fertilizer use. This effect was partially compensated by 7% decrease in plant uptake. Crop products output became 25% lower, but animal products output increased by 12%. And feed concentrates input to Sør-Trøndelag increased by 11%. Rogaland is the region where the soil budget is increasing. Average for 2009-2011 soil phosphorus budget is 18% higher then 12 years earlier. The amount of phosphorus taken up by plants decreased by 19%; most of it is used for production of animal fodder or consumed through grazing. Phosphorus import with feed concentrates increased by 19% and the total input to animal production with feed, fodder and grazing increased by 7%. Output in animal products increased by 18%. Phosphorus intake, deposition and excretion by different animal categories for both time points are shown in the Table 11 (Appendix 9).

3.2. Long-term phosphorus budgets of Norwegian regions

Long-term changes in phosphorus soil budget (input less output) over years in Akershus, Sør-Trøndelag and Rogaland are shown in Figure 8. In all three counties, the budget was increasing until mid 1970s, then leveled, and decreased from mid 1980s to mid 1990s. After mid 1990s, the budget is still decreasing in Akershus, leveled in Sør-Trøndelag, and increases in Rogaland.

The output did not change much over time, and the budget pattern more or less follows the input pattern, i.e. the increase of budget coincides with the increase of input, and decrease of budget coincides with decrease of input (Figure 9). The decrease in input since mid- 1980s coincided with the decline in mineral fertilizer use (Figure 10). The input is still decreasing in Akershus – the region with dominant crop production – as mineral fertilizer use is still declining. In the same time, the input of phosphorus with manure is increasing in Rogaland – area with high livestock density, and Sør-Trøndelag – county with mixed agriculture system (Figure 10). And the growing input with manure compensates the decrease in mineral fertilizer use in Sør-Trøndelag and overcomes it in Rogaland.

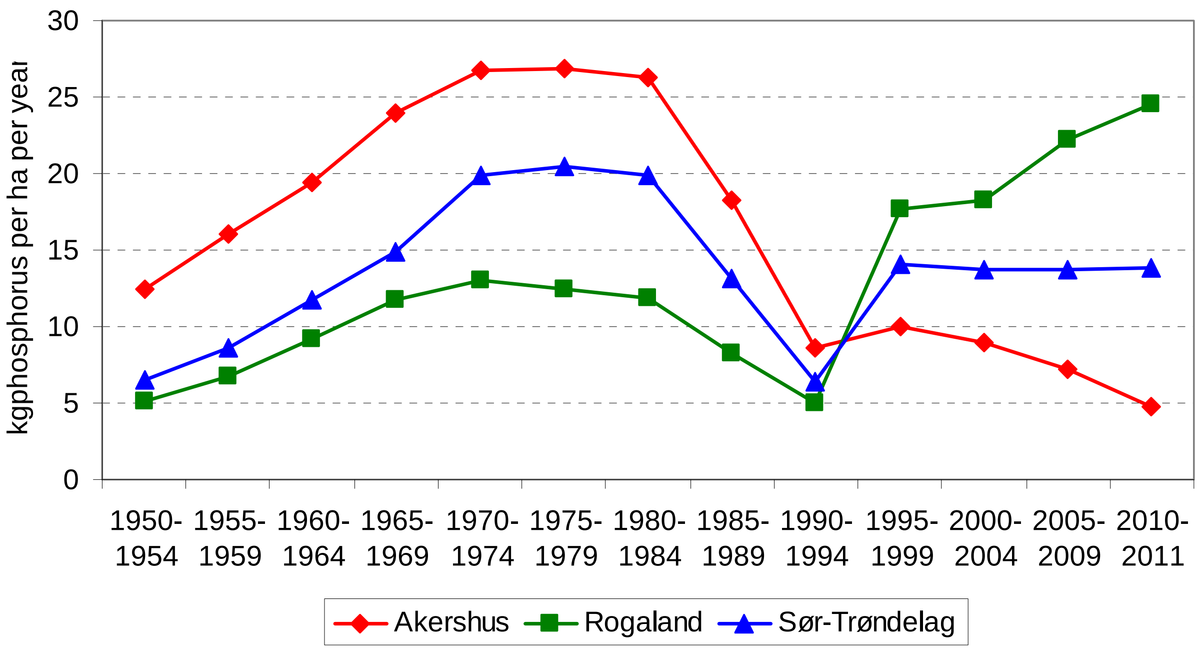

Figure 8. Phosphorus soil budget (input less output) in agricultural land in three Norwegian counties in 1950-2011. Each data point is 5-years average. The minimum during 1990-1994 may be the result of the shift toward more intensive animal production or accounting for more animal groups in statistics since this period.

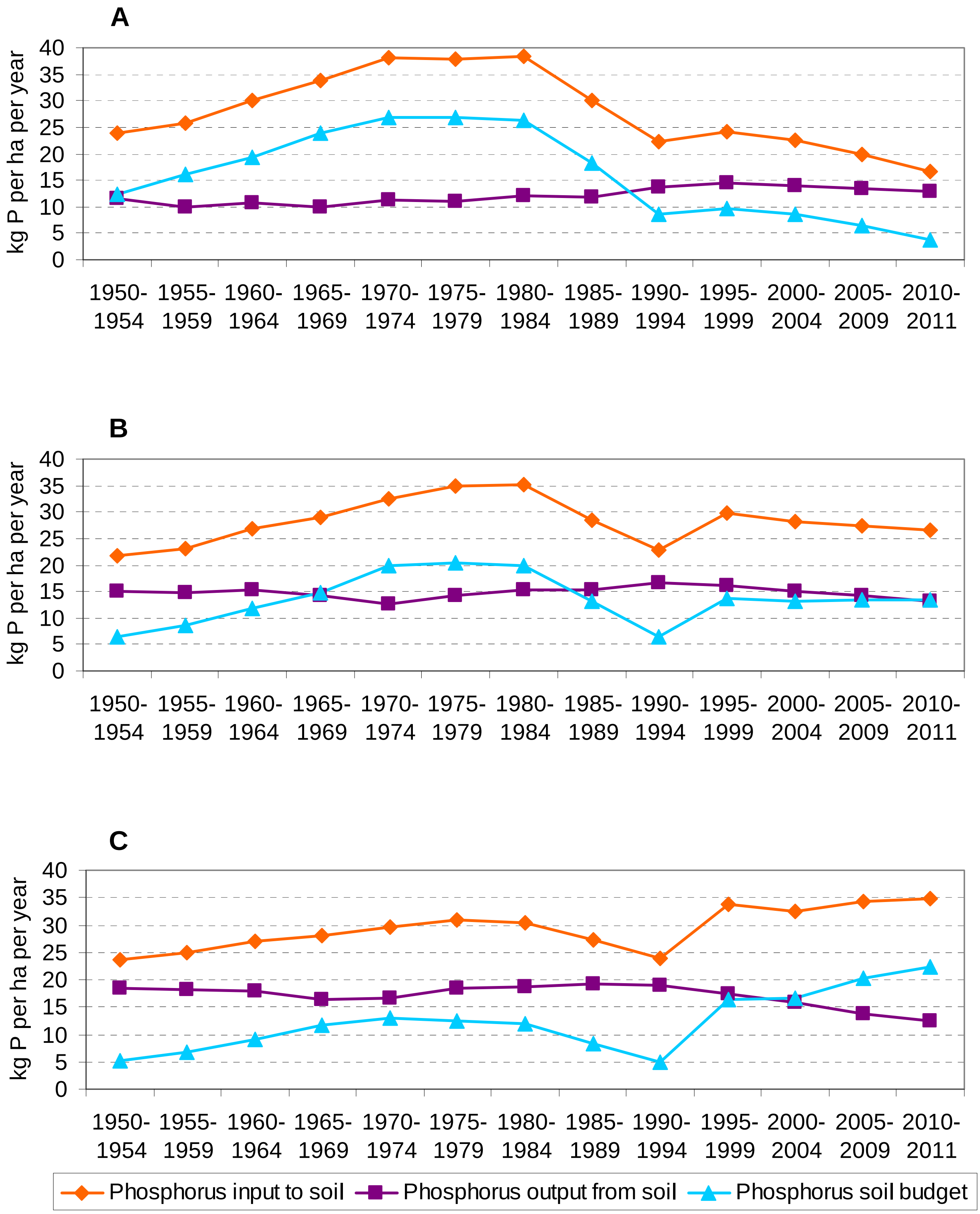

Figure 9. Long-term phosphorus fluxes and budgets in agricultural soil of Akershus (A), Sør-Trøndelag (B) and Rogaland (C). Input is a sum of phosphorus amount in mineral fertilizer, animal manure, composted waste and sewage sludge; output is a sum of phosphorus removal through plant uptake and losses due to erosion and leaching; budget is difference between input and output. Each data point is 5-years average.

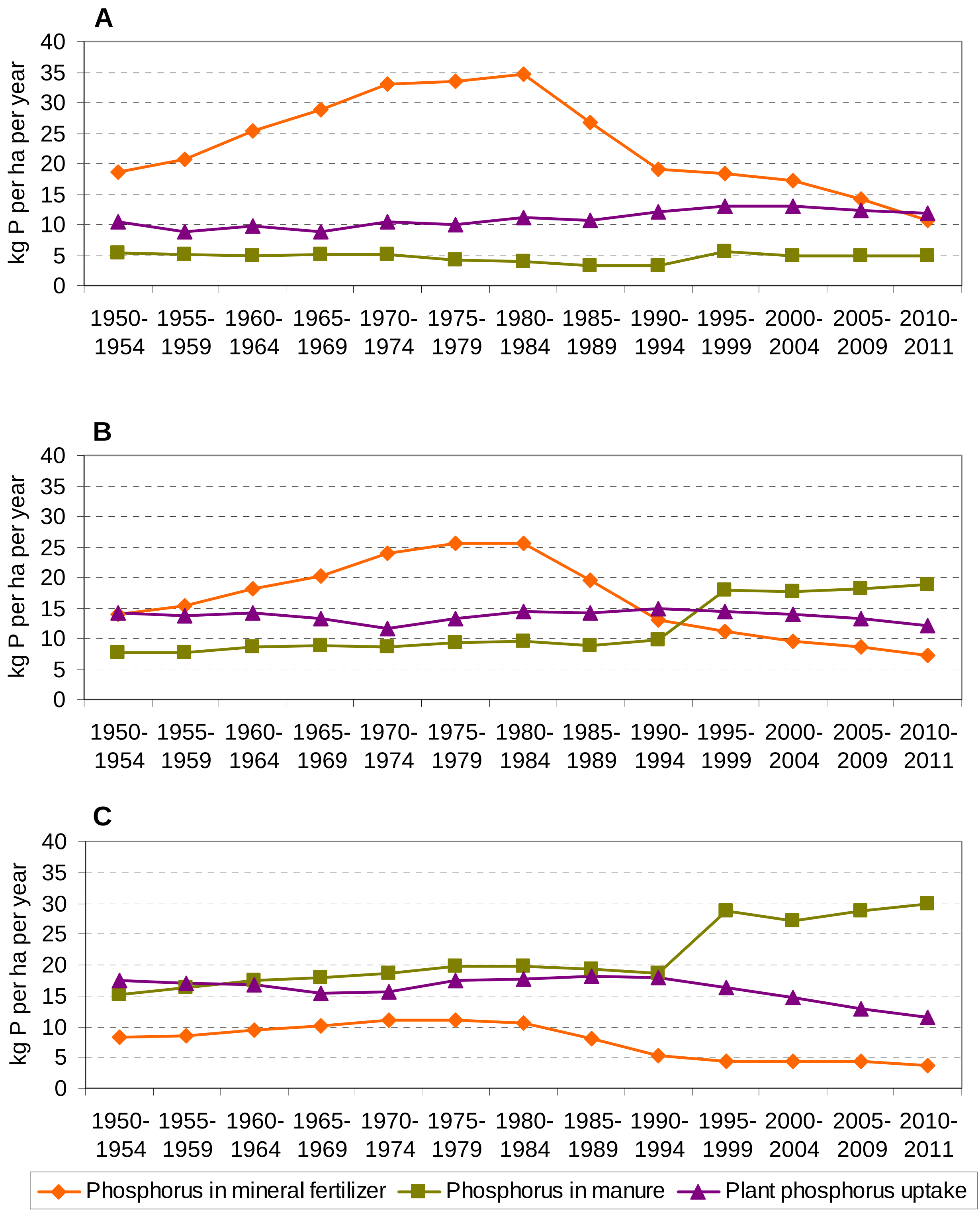

Figure 10. Main fluxes in agricultural soil of Akershus (A), Sør-Trøndelag (B) and Rogaland (C) from 1950 to 2011. Each data point is 5-years average.

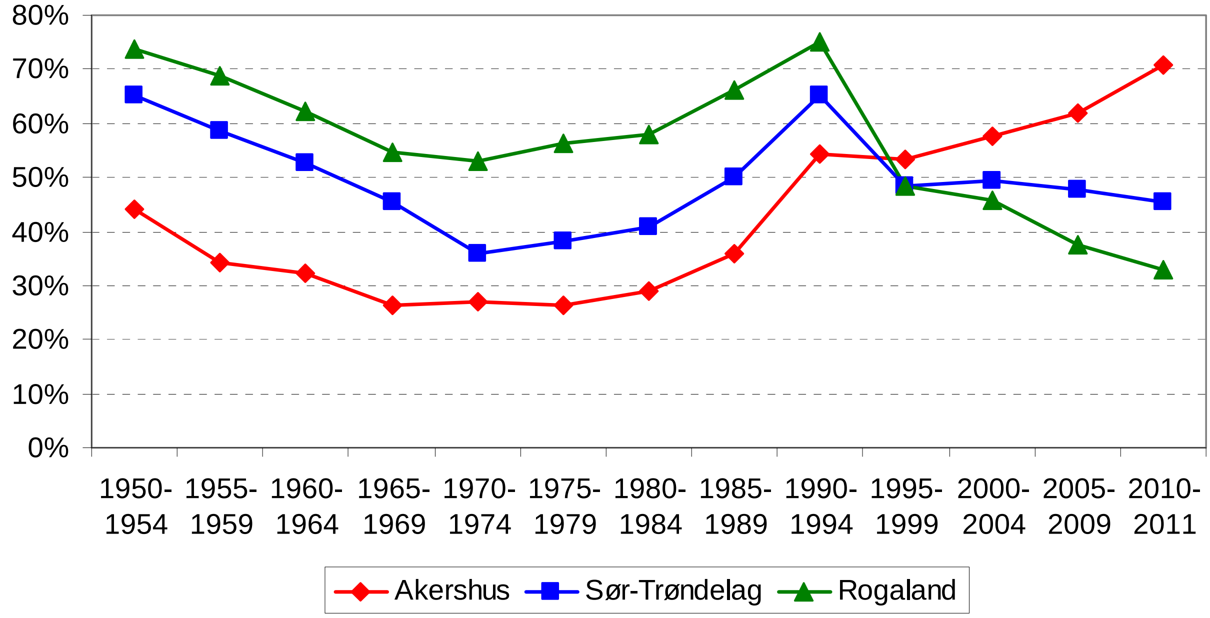

Figure 11. Changes in phosphorus soil efficiency over time. Each data point is 5-years average. The peak efficiency in 1990-1994 coincides with the minimum budget (Figure 8).

Long-term changes in soil phosphorus efficiency (plant uptake divided by sum of all inflows to soil) in Akershus, Sør-Trøndelag and Rogaland (Figure 11) inversely reflected the changes in phosphorus soil budget over the same period (Figure 8): the higher the input and, consequently, soil budget, the lower the phosphorus soil efficiency. The peak efficiency (Figure 11) in 1990-1994 coincides with the minimum budget (Figure 8). Since mid-1990s the highest efficiency is observed in Akershus, the lowest in Rogaland.

3.3. Phosphorus accumulation in soil and soil phosphorus test

Long-term phosphorus net accumulation in agricultural soil is presented on the Figure 12. The highest accumulation since 1950, over 1000 kg phosphorus per ha by 2011, is found in Akershus, mostly due to long-term mineral fertilizer application in high quantities with excess to plant uptake. In Sør-Trøndelag phosphorus net accumulation in agricultural soil is over 800 kg/ha, in Rogaland around 700 kg/ha by 2011. There is no data for total soil phosphorus, so the calculated net-accumulation is considered as the lower border of total phosphorus content and is compared with the results of P-AL soil phosphorus test (Figure 12).

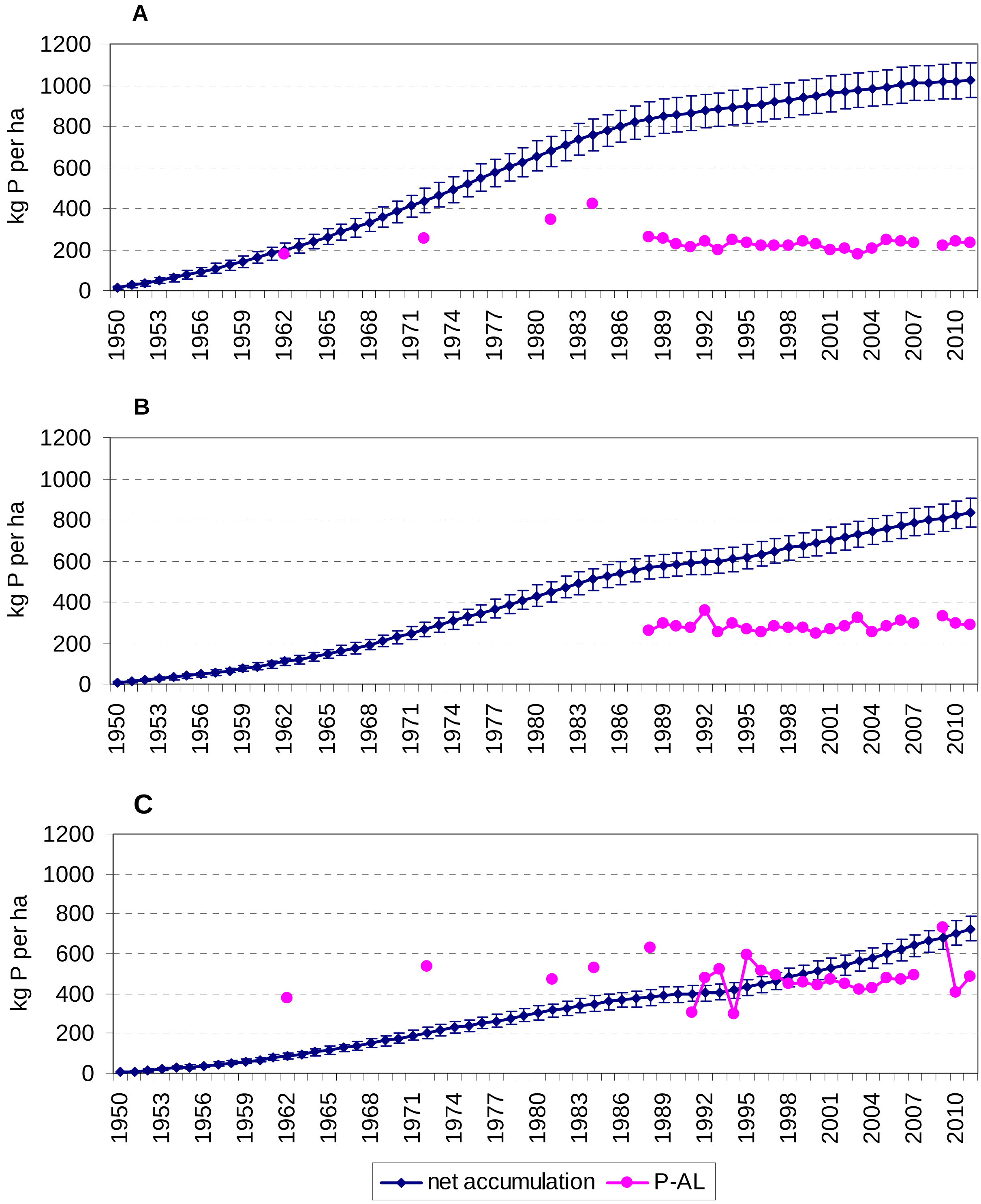

Figure 12. Phosphorus soil stock accumulation and results of P-AL soil phosphorus test in three Norwegian counties for years 1950-2011. A – Akershus, B – Sør-Trøndelag, C – Rogaland. Error bars are shown for 95% confidence interval. P-AL data for 1960-1985 are from Krogstad et al., 1987; for 1988-2011 from Jordsdatabanken (Grønlund, 2013).

Graphs demonstrate that available phosphorus content, according to P-AL measurement, was more or less stable over the whole monitoring period and did not reflect the increase in soil accumulation. During last five years (2007-2011) P-AL test showed around 22±1% of net accumulation in Akershus by 2007-2011, 37±3% in Sør-Trøndelag and 77±21% in Rogaland.

4. Discussion and outlook

4.1. Uncertainties

Sources of uncertainties include variations in material phosphorus content, errors in material flow estimation, and under- or overestimation of losses through erosion and leaching, where one can only be certain of order of magnitude. In addition, consistency of statistic databases cannot be checked. To minimize the influence of these uncertainties the flows were cross-checked where possible. In order to account for variations in data quality, incertainty propagation was carried out. The largest uncertainties were observed for either minor flows or flows that do not influence the main conclusions of the present study.

4.2. Changes in phosphorus fluxes and budget over time

Mineral fertilizer use was declining since mid-1980s mostly due to environmental concern, and decreases dramatically during last several years due to changes in fertilization recommendations, as balanced phosphorus fertilization strategy was introduced in 2007-2008 in many areas in Norway (Krogstad et al., 2008). There may be some other reasons like increasing inorganic fertilizer prices, production cost reduction measures or improvement in fertilization strategy as, for example, in France (Senthilkumar et al., 2012b).

The comparison of phosphorus fluxes on the county level based on dominating agricultural production system showed notable differences between regional soil phosphorus budgets within one country. In crop production region (Akershus) the main phosphorus fluxes are plant uptake, mineral fertilizer and crop products (Figure 7D). Inorganic fertilizer use decreased dramatically over past years, but it is still over 10 kg/ha, that is higher than national average that is around 8 kg phosphorus per ha total utilized agricultural area, though for arable land it is 13 kg/ha (Ulen et al., 2007). Soil phosphorus budget dropped by factor of 3 over last decade, so balanced budget may be expected in the nearest future. However, this region is heavily dependent on mineral fertilizer input.

In Rogaland, with its dominant animal farming, the most important fluxes are feed and fodder (especially imported feed), animal manure and plant uptake (Figure 7F). Soil budget is growing since mid-1990s, when input in plant manure exceeded plant uptake (Figure 10C).

But even though input in manure is more than sufficient to compensate plant uptake, mineral fertilizer use is still 3.5 kg/ha. Plant uptake is continuously decreasing during last decades (Figure 10C), while input of feed concentrates is increasing (Figure 7C and 7F). Rogaland is a region with highest accumulation of phosphorus excreted with manure – 30 kg/ha-yr, while national average is 12 kg/ha (Ulen et al., 2007), and is highly dependent on input of feed concentrates.

Sør-Trøndelag, an area with mixed farming, combines features of crop production and animal production areas. The dominant fluxes are feed and fodder, animal manure, plant uptake and, to a lesser extent, mineral fertilizer (Figure 7E). Inorganic fertilizer use decreased during past years, but it is still 6.8 kg/ha. In this region phosphorus input to soil with animal manure is more than sufficient to compensate phosphorus removed from soil by plant uptake. The need of mineral fertilizer application may be questionable, as it could be replaced with manure produced within region. However, technical difficulties, first of all geographical segregation of crop and animal producing farms even within the county, prevent substitution of imported inorganic fertilizer with manure. Practically all crops produced in Sør-Trøndelag are used locally for animal fodder (Figure 7E). However, locally produced fodder is not enough – the region is dependent on feed concentrates input, as well as Rogaland.

The same pattern was earlier observed for four French regions: approaching balanced budget and high dependence on mineral fertilizer input in crop producing region (Centre), high accumulation due to animal manure and still not zero fertilizer use in animal production region (Brittany), and not complete substitution of mineral fertilizer with animal manure in mixed-farming regions (Lorraine and Aquitaine) (Senthilkumar et al., 2012b).

4.3. The relevance of soil phosphorus test for the assessment of phosphorus accumulation in soil

The results of monitoring of available phosphorus content in soil with P-AL test (Krogstad et al., 2008, Grønlund, 2013) did not reflect the increase in soil accumulation over the long period (Figure 12). It may indicate that a large proportion of accumulated phosphorus is transferred to unavailable form (”fixed” phosphorus pool). However, the phosphorus budget might have been overestimated in this study. First, output with plant uptake may be underestimated. Calculation included plant uptake of cereals (wheat, barley, oats, rye and triticale) green forage, silage and hay. Vegetables, fruit and berries were not accounted for. In addition, statistical data for green fodder, silage and hay may be lower than actual harvest due to large losses during harvest and conservation (Bleken and Bakken, 1997). Losses due to erosion and leaching may be higher than assumed as they vary considerably from site to site (Ulen et al., 2007; Rød et al., 2009; Ulen et al., 2012). A part of phosphorus surplus may be transferred to subsoil due to downward movement of phosphorus (Oehl et al., 2002; Rubæk and Hekrath, 2008). However, when no significant phosphorus accumulation in the lower soil horizon is observed, the most likely explanation is phosphorus "fixation" – conversion to non-extractable form (McCollum, 1991).

At present (by 2007-2011) P-AL soil test shows around 22% of net accumulation in Akershus, 37% in Sør-Trøndelag and 77% in Rogaland. Soil phosphorus availability depends on the properties of the source of phosphorus applied to soil rather then total amount of added phosphorus (Ebeling et al., 2003b). The highest results of P-AL test in Rogaland, the region with the lowest accumulated soil stock, may be explained by higher availability of phosphorus originated from manure. Total phosphorus content in manure may vary a lot, but almost 70% of phosphorus in manure is available (Dou et al., 2000; Shen et al., 2011). That is because, though 50-90% of phosphorus in manure is inorganic (Dou et al., 2000; Shen et al., 2011), high content of organic matter in soil, especially due to manure application, increases phosphorus availability (Ebeling et al., 2003a).

4.4. Possible implications for phosphorus management

Present study showed the relevance of main problems concerning phosphorus use for the situation in Norwegian agriculture. Area of crop production (Akershus) is heavily dependent on mineral fertilizer input. Area of high livestock density (Rogaland) is heavily dependent of feed import and, in the same time, accumulates phosphorus in soil stock at a growing rate. Even area of relatively mixed production system (Sør-Trøndelag) depends on mineral fertilizer and feed import and accumulates phosphorus in soil. So, Norwegian agriculture contributes to the global problem of phosphorus resource depletion and local problems of decreasing water quality due to excessive phosphorus run-off. Such production system is unsustainable with respect to phosphorus (Schröder et al., 2010) and needs adjustment of inputs to outputs. Proper management of agricultural phosphorus cycle is required.

When mineral fertilizer is concerned, the main strategies to mitigate resource scarcity are higher prices, more efficient resource use, introduction of alternatives and recovery of the resource after use (Cordell et al., 2009). The measures taken in order to reduce mineral fertilizer use have met a number of challenges. Liberal use of phosphate fertilizer has always been motivated by concerns about crop yield decline and resulting economical losses (Schröder et al., 2010). Moreover, farmers would not easily believe that their fertilizer use could affect water bodies far away (Schröder et al., 2010), they considered fertilizer as the product that was useful in getting high yields and in building up the soil for the future. Norwegian farmers reacted negatively on such measures to decrease fertilizer use as ambient tax on fertilizer while being quite positive to ambient tax on pesticides (Vedeld, 2002). However, in areas with high soil phosphorus content there is no need for excess fertilizer use (Krogstad et al., 2008). The application of fertilizer near plant roots in the period of greatest crop demand or more frequently but with smaller quantities may reduce losses and increase efficiency (Tilman et al., 2002). The ability of plants to take up phosphorus or to utilize it varies with genotype, and the plant uptake capacity can be enhanced by the simbiosis with miccorizal fungi (Schröder et al., 2011). Therefore, to increase the efficiency of mineral fertilizer, different fertilizer placement methods may be used, as well as improved crops and Mycorrizas.

The more efficient use includes the reduction of losses, which is also highly desirable from the environmental perspective. Soil management practices that are efficient in phosphorus losses reduction are a decrease in autumn ploughing, growing of catch crops, and implementation of constructed wetlands (Bechmann et al., 2008). Ploughless tillage may reduce total phosphorus run-off losses by 10-80% compared to conventional ploughing (Ulen et al., 2010). However, eutrophication management based on phosphorus control did not result in expected improvement of water quality after over two decades of reduced phosphorus input (Jarvie et al., 2013). Reduced tillage and no-till practices that are used to control phosphorus erosion losses have their trade-offs. They may lead to higher run-off losses of dissolved phosphorus. Control of diffuse phosphorus pollution in the areas with high content of dissolved phosphorus should be based first of all on lowering of the phosphorus soil accumulation (Kleinman et al., 2011).

The use of mineral fertilizer was reduced in many crop producing areas, and currently the main concern is phosphorus input in manure. At present, high positive phosphorus budget typically originates from animal waste in regions with high livestock density and cropland insufficient to spread the produced manure (MacDonald et al., 2011). In Rogaland the soil phosphorus budget is growing from mid-1990s. Accumulation of manure may increase phosphorus inputs to surface water. For example, in Feixi county in Central China the phosphorus loss to water was found higher than the entire phosphorus input to soil with livestock manure (Wu et al., 2012). The possible measures to avoid the consequences of manure accumulation include exporting manure, adjusting livestock diet and lowering livestock density.

The recycling of manure phosphorus from areas with high livestock density to crop production areas was suggested both at a national and a global scale (MacDonald et al., 2011; Bateman et al., 2011). Export of manure from animal production areas can lighten accumulating phosphorus burden. In the areas of crop production (like Akershus) imported manure can substitute mineral fertilizer use. However, manure export may meet a number of challenges. At present it does not seem to be profitable from the economical point of view, as mineral fertilizer is relatively cheap and easy to use, while manure is bulky and otherwise difficult to transport. Up to the year 2008 the prices of mineral fertilizer were to low to stimulate production and use of renewable fertilizer (Cordell et al., 2012). In addition, the quality of waste-based products may be believed to be low (Schröder et al., 2010). People may be reluctant to buy food products that have been grown using manure due to, for example, belief in pathogen hazard. However, if animal manure is treated by composting or used for biogas production, the resulting compost or soil conditioner will no longer pose pathogen hazard. The use of treated manure as a fertilizer can reduce dependence on mineral fertilizer supply and will allow to close the phosphorus cycle where animal and crop production are spacially combined (Tilman et al., 2002). The areas of mixed agriculture, like Sør-Trøndelag, are most likely to achieve substitution of mineral fertilizer with manure, although at present waste treatment and waste-based products application are not optimized for high phosphorus recycling efficiency (Bøen and Grønlund, 2008). One of the possible solutions might be separation of phosphorus-rich solid fraction from nitrogen-rich liquid fraction (Christensen et al. 2009; Cordell et al., 2012). This will facilitate transport and make distribution and dosage of nutrients easier. Where manure export or manure processing are not feasible, the on-site production of feed concentrates should be considered (Hermans and Vereijken, 1995), but it will require sufficient area for plant production.

Another option is adjustment of animal diet. The use of phosphorus in animal diet may often excess nutritional requirements. In US dairy farms diet of dairy cows often contains 20-25% excess phosphorus (Toor et al., 2005). Lower phosphorus in animal diet would result in reduction of phosphorus excretion in manure, and, as a consequence, lower phosphorus accumulation in soil and potential phosphorus losses to water (Toor et al., 2005). Increased inorganic phosphorus in the diet of dairy cattle can increase phosphorus availability in the manure (Ebeling et al., 2003b), so use of different diet components may result in manure with desired properties. Phytase supplements to pig and poultry diet enhance phosphorus availability in feed. Lower phosphorus diet may be introduced then, and phosphorus excretion in manure will decrease. Moreover, it was shown that genetically modified pigs with introduced gene of bacterial phytase did not need phosphate additives to their feed and their manure contained up to 75% less phosphorus then that from non-genetically modified pigs (Elser and Bennett, 2011). However, it is unlikely that use of genetically modified organisms for food purposes will be ever approved in Norway, or large-scale breeding of genetically-modified animals will take place in other countries. But addition of phytase produced by genetically-modified bacteria is practiced.

Lowering the livestock density will decrease phosphorus accumulation in the areas of intensive animal production. But decreasing production, in its turn, will have economical and social consequences. So, while from environmental perspective extensification of agricultural food production is desirable, it should better be intensified from the perspective of land, water, labor and energy use efficiency (Schröder et al., 2010).

A possible solution can be the expansion of organic agriculture. The nutrient surplus is lower and nutrient efficiency is higher in organically managed farms compared to conventional farms (Steinshamn et al., 2004; Tully and Lawrence, 2011).

For implementation of policy measures aimed at sustainable nutrient management, economical incentives are needed. Such incentives may include subsidies or tax relief on manure export, manure processing, low phosphorus feed and phytase addition to feed, as well as taxes on phosphate rock-based inputs where alternatives are available, or on phosphorus intensive bio-energy consumption (Schröder et al., 2010; Cordell et al., 2012).

5. Conclusions

The regional phosphorus flow analysis has shown the influence of agricultural production system on the main phosphorus flows and phosphorus soil stock. The crop producing region (Akershus) is highly dependent on mineral fertilizer import, although this input is decreasing over time. The mainly animal producing region (Rogaland) is dependent on feed concentrates input, and accumulates large amount of phosphorus in manure. In the mixed production region (Sør-Trøndelag) manure does not completely replace mineral fertilizer, and agricultural production is dependent on input of feed concentrates; this indicates fragmentation between fodder production and animal husbandry.

The phosphorus soil budget was positive in an all three regions examined over the entire analyzed period, but shows differences between the three Norwegian regions depending on the agricultural production system. In Akershus (crop production) the budget was decreasing since 1970s and is currently the lowest, while in Rogaland (animal production) it is growing since 1990s, and is now the highest. Nevertheless, the accumulated phosphorus stock in soil is the highest in Akershus, the crop-producing region, due to long-term excessive mineral fertilizer application. The stock is lower in the region with mixed agriculture (Sør-Trøndelag) and the lowest in the region with dominating animal production (Rogaland). The results of P-AL test show the different proportion of calculated lower border of total phosphorus content, depending on main phosphorus source, and do not reflect phosphorus soil accumulation. Apparently, a large part of accumulated phosphorus can not be revealed by this test because it is transferred into unavailable form (”fixed” phosphorus pool).

The situation in the Norwegian agricultural system with respect to phosphorus is characterized by high dependence on inputs with simultaneous soil accumulation and can therefore be called unbalanced. From the system perspective, the measures to mitigate this unbalance should aim at the recycling of phosphorus from the areas of accumulation where it causes pollution to the areas of need where it can become a resource. Practical implementation of this principle can however meet a number of challenges: physical, economical and even social.

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Appendix 1. Phosphorus reactions in soil

Absorption and precipitation

Phosphate anions can be bound to soil constituents and become difficult for plant uptake (Tan, 2011). Two types of mechanisms can be defined:

(1) Phosphate retention refers to adsorbed phosphate that can be extracted with dilute acid, relatively available to plants. Acid soil contains significant amount of Al^{3+}, Fe^{3+}, Mn^{2+} ions. Basic soil contains Ca^{2+} ions. Phosphate may be absorbed on the colloid surface with Al^{3+}, Fe^{3+}, Mn^{2+} ions as bridges (metal bridging or co-adsorption). Such phosphates are still readily available to plants. Adsorption like this can also occur with Ca-saturated clays.

- In Al-rich/acid soil

$[Al^{3+}]^{1/3}[H_2PO_4] = (Al^{3+})^{1/3}(H_2PO_4)$; Clay – Al – H_2PO_4

- In Ca-rich/basic soil

$[Ca^{2+}]^{1/2}[H_2PO_4] = (Ca^{2+})^{1/2}(H_2PO_4)$; Clay – Ca – H_2PO_4

The retention can also take place with protonated OH groups (in clays containing oxilhydroxy, aluminol, ferrol and silanol surfaces) in two steps:

- adsorption (rapid):

$–Al–OHH^+ + H_2PO_4^- \leftrightarrow –Al–OHH^+–H_2PO_4^-$

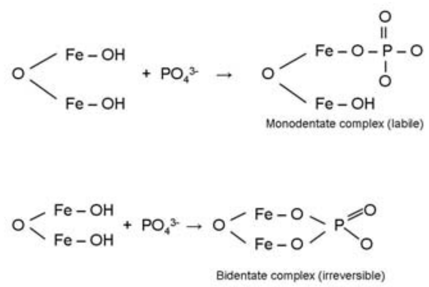
- penetration into crystal structure (slow, several weeks or longer):

$–Al–OHH^+–H_2PO_4^- \rightarrow –Al–H_2PO_4 + H_2O$

(2) Phosphate fixation refers to phosphate not extractable with dilute acids that are not readily available to plants ("fixed").
Fixation reaction takes place between phosphate and Fe or Al ions and Fe or Al hydrous oxides, or between phosphate and silicate minerals, and results in phosphates insoluble in water, relatively non-available to plants.

$Al^{3+} + H_2PO_4^- \rightarrow Al(H_2PO_4)_3 \downarrow$

Fixation can also take place by formation of complexes or chelates (Tan, 2011).



Reaction with silicate clays:

- $Al_2Si_2O_5(OH)_4$ (kaolinite) → $Al(OH)_2H_2PO_4$ (variscite)
- $Al_2Si_2O_5(OH)_4$ (kaolinite) → $2Al(OH)_2^{+}$ + $Si_2O_5^{2-}$
- $2Al(OH)_2 + 2H_2PO_4^{-} \rightarrow Al(OH)_2H_2PO_4$

(Bohn et al., 1985).

Phosphate fixation in alkaline soil (pH > 7):

- $3Ca^{2+} + 2PO_4^{3-} \rightarrow Ca_3(H_2PO_4)_2 \downarrow$
- $3Ca\ CO_3 + 2PO_4^{3-} \rightarrow Ca_3(H_2PO_4)_2 \downarrow + 2CO_2 \uparrow$
- dibasic calcium phosphate dihidrate $CaHPO_4{\cdot}2H_2O$
- octocalcium phosphate $Ca_4H(PO_4)_3$
- hydroxiapatite $[Ca_5(H_2PO_4)_3(OH)]$

In acidic soil (pH < 5.5)

- amorphous Al and Fe phosphates
- variscite ($AlPO_4{\cdot}2H_2O$
- stregnite ($FePO_4{\cdot}2H_2O$)

(Tan, 2011).

Immobilization

Biological fixation

Etherification with glucose → Glucose-6-phosphate ester
Pyrophosphate bonding in ATP
Phosphates + humic acid → phosphohumate complexes or chelates
(Tan, 2011).

Appendix 2. Soil types

Table 2. Soil types, their properties and land use (Smith and Smith, 2011, UIDAHO, 2013; NIFL, 2013).

Soil order	Properties	Land use
Alfisol	Moderately leached, Accumulated clays, Shallow penetration of humus, Naturally fertile	Cropland Grazing Forest Savanna Grassland
Andisol	Formed in volcanic ash, High water-holding capacity, High P-fixation	Forest Cropland Pasture
Aridisol	Very dry Low organic matter content, High base content Accumulated calcium carbonate, silica, salts, gypsum Prone to salinization	Rangeland Wildlife habitat Irrigated cropland
Entisol (all soil types that do not fit into one of the other 11 orders)	Newly formed Great diversity, Unconsolidated parent material	Cropland Pasture Rangeland Forest Wildlife habitat Urban
Gelisol	Permafrost within 2 m from surface, Large quantity of organic carbon due to slow rate of organic decomposition	Wildlife habitat
Histosol	Wet, Poor drainage, Contain at least 20-30% organic matter by weight, bulk density less than 0.3 g/cm^3 (½ or more of upper 80 cm is organic) Bog and muck soil	Forest Cropland Urban Recreation Wildlife habitat

Soil order	Properties	Land use
Inseptisol	Young (slightly developed) Shallow	Cropland Pasture Forest Rangeland Wildlife habitat
Mollisol	Deep, Fertile, Large amount of organic material Rich in bases Prone to calcification Most productive, agricultural soil	Cropland Pasture Forest Rangeland
Oxisol	Highly weathered Infertile Very low nutrient reserves Rich in Kaolinite, Fe and Al oxide minerals, humus High P retention Can be productive with input of lime and fertilizers	Rain-fed crop
Spodosol	Sandy, Acidic, Accumulated amorphous mixtures of organic matter and Al and Fe, Naturally infertile Responsive to good management Requires addition of lime	Forest Wildlife habitat Cropland Pasture
Ultisol	Weathered Acid Low native fertility Low base content Ca-, Mg-, K-deficient Accumulated clay Contain Fe oxides	Forest Cropland Pasture
Vertisol	Clayey Have cracks Shrink when dry, swell when moist Slow permeability, irrigation can result in waterlogging and elevated salinity	Rangeland Cropland Pasture

Appendix 3. Soil phosphorus tests

Table 3. Soil phosphorus tests.

	Ammonium lactate, P-AL test	Mehlich-3 Test	Bray-Kurtz P1 test	Olsen test
Extracting solution	ammonium lactate	acetic acid, ammonium nitrate, ammonium fluoride, nitric acid and the chelate, EDTA	dilute hydrochloric acid and ammonium fluoride solution	weak sodium bicarbonate
Correlation		0.83-0.99	0.74-0.94	0.73-0.96 for alkaline soil conditions
Detection limit		1 ppm (dry soil basis)	1 ppm (dry soil basis)	
Recommended for	acid soil	acid/neutral soil	acid/neutral soil	alkaline soil
May show % of total	9-18%	6-9%*	5-7%	2-4%
Reference	Neyroud and Lischer, 2003; Eriksson et al., 2013	Watson and Mullen, 2007; Neyroud and Lischer, 2003	Watson and Mullen, 2007; Neyroud and Lischer, 2003	Watson and Mullen, 2007; Neyroud and Lischer, 2003

* Mehlich-3 extractable P concentrations of 42 soils from US and Canada represented from 0.5% to 72% of soil total P (Hesterberg, 2011).

Appendix 4. System definition

Table 4. Parameters.

	Parameter	Unit	Reference
AA	Agricultural area	ha	SSB, 1974; SSB, 1974-1996; SSB, 1982; SSB, 1995; StatBank, 2013
FRS	Fertilizer sold	tons	Mattilsynet, 2010; 2011; 2012, Bøen, 2013; Nyhus, 2013
P_{FR}	P concentration in fertilizer	%	Calculated from Mattilsynet, 2010; 2011; 2012, Bøen, 2013; Nyhus, 2013
WS	Waste composted	1000 tons	StatBank, 2013
CR	Crop harvest: Wheat Barley Oats Rye and Triticale Potato Green fodder and silage Hay	1000 tons	SSB, 1974; SSB, 1974-1996; SSB, 1982; SSB, 1995; StatBank Norway, 2013
FDS	Feed concentrates sold	tons	Breen, 2013
NA	Number of animals	heads	SSB, 1974; SSB, 1974-1996; SSB, 1982; SSB, 1995; StatBank, 2013
APR	Animal products: Horse, Cattle, Pigs Sheep, Poultry Milk Eggs	tons 1000 l tons	StatBank, 2013
PD	P discharge with wastewater	tons	StatBank, 2013

	Parameter		Unit	Reference
P_{WS}	P concentration in waste	0.15	%	Briseid et al., 2010
PR	P removal by WWTP	40-90	%	Berge and Mellem, 2012
SU	Sewage sludge used in agriculture	50	%	Berge and Mellem, 2012
P_{EL}	P losses due to erosion and leaching	1	kg/ha-yr	Rød et al., 2009
P_{FD}	P concentration in feed	0.5	%	Calculated from Breen, 2013
P_{CR}	P concentration in crops:			
	Wheat	0.40	% of dry matter	Antikainen et al., 2005
	Barley	0.38		
	Oats	0.39		
	Rye and Triticale	0.36		
	Potato	0.21		
	Green fodder and silage	0.32		
	Hay	0.24		
DM_{CR}	Dry matter concentration in crops:	86	%	Antikainen et al., 2005
	Wheat	86		
	Barley	86		
	Oats	86		
	Rye and Triticale	22		
	Potato	23		
	Green fodder and silage	83		
	Hay			
P_{APR}	P concentration in animal products:			
	Horse	0.71	%	Antikainen et al., 2005; Matvaretabellen, 2013
	Cattle	0.71		
	Pigs	0.55		
	Sheep	0.55		
	Poultry	0.67		
	Milk	0.09		
	Eggs	0.24		

	Parameter		Unit	Reference
P_{EM}	P excreted in manure:			
	Horse	7	kg/animal-yr	Karlengen, 2012
	Dairy cows	14.4		
	Beef cows	8		
	Other cattle	9		
	Sheep	2		
	Milking goats	1.2		
	Pigs for breeding	5.8		
	Slaughter pigs	0.45		
	Laying hens	0.13		
	Broilers	0.04		
SAL	Slaughtering age lambs	160	days	Ringdal et al., 2012
GOL	Grazing on uncultivated land, lambs	90	days	Karlengen, 2012
GR	Grazing share cattle	20-40	%	SSB, 1974; SSB, 1974-1996; Tine, 2013
	Grazing share sheep	50	%	Assumed
P_{NTR}	Nutrient requirements:			
	Horse	7.3	kg/animal-yr	Merck, 2013
	Dairy cows	20		
	Beef cows	9		
	Other cattle	6		
	Sheep	2		
	Milking goats	1.6		
	Pigs for breeding	6		
	Slaughter pig	1		
	Laying hens	0.16		
	Broilers	0.05		
	Lambs	0.005	kg/animal-day	Estimate from MSU, 2013

Table 5. System variables.

	Variables
$X_{0\text{-}1MF}$	Mineral fertilizer
$X_{0\text{-}1CC}$	Compost
$X_{0\text{-}1SS}$	Sewage sludge
$X_{4\text{-}1}$	Manure to soil (cultivated land)
$X_{1\text{-}2}$	Plant uptake (excluding plant residue)
$X_{1\text{-}0}$	Erosion and leaching
$\Delta S1$	Soil stock change
$X_{2\text{-}0}$	Crop products
$X_{2\text{-}3}$	Fodder
$X_{2\text{-}4}$	Grazing (cultivated land)
$X_{0\text{-}4}$	Grazing (uncultivated land)
$X_{0\text{-}3}$	Imported feed
$X_{3\text{-}4}$	Feed and fodder
$X_{4\text{-}0\text{-}AP}$	Animal products: meat, milk and eggs
$X_{4\text{-}0\text{-}MN}$	Manure to soil (uncultivated land)

Table 6. Equations.

	Mass Balance Equations
(1)	$X_{0\text{-}1MF} + X_{0\text{-}1CC} + X_{0\text{-}1SS} + X_{4\text{-}1} = X_{1\text{-}2} + X_{1\text{-}0} + \Delta S1$
(2)	$X_{1\text{-}2} = X_{2\text{-}3} + X_{2\text{-}4} + X_{2\text{-}0}$
(3)	$X_{2\text{-}3} + X_{0\text{-}3} = X_{3\text{-}4}$
(4)	$X_{3\text{-}4} + X_{2\text{-}4} + X_{0\text{-}4} = X_{4\text{-}0\text{-}AP} + X_{4\text{-}0\text{-}MN} + X_{4\text{-}1}$ for every group of animals
	Model approach equations
(4a)	$X_{3\text{-}4} + X_{2\text{-}4} = \Sigma\ NA \times P_{NTR}/AA$ For 1950-1995 $X_{4\text{-}0}$ was not calculated due to the absence of data for animal production by county, $X_{3\text{-}4}$ is therefore quantified according to nutritional requirements.
(5)	$X_{0\text{-}1MF} = FRS \times P_{FR}/AA$
(6)	$X_{0\text{-}1CC} = WS \times P_{WS}/AA$
(7)	$X_{0\text{-}1SS} = SU \times PD \times PR/(1 - PR)/AA$
(8)	$X_{2\text{-}4} = (X_{3\text{-}4} + X_{2\text{-}4}) \times GR$ for every group of animals except lambs
(8a)	$X_{3\text{-}4} = 0$ for lambs
(9)	$X_{2\text{-}3} + X_{2\text{-}0} = \Sigma\ CR \times P_{CR}/AA$
(10)	$X_{1\text{-}0} = P_{EL}$
(11)	$X_{0\text{-}3} = FDS \times P_{FD}/AA$
(12)	$X_{4\text{-}0\text{-}AP} = \Sigma\ APR \times P_{APR}/AA$ = (Σ carcasses weight × P concentration in animals + amount of milk × P concentration in milk + amount of eggs × P concentration in eggs) /AA
(13)	$X_{4\text{-}1} = \Sigma\ NA \times P_{EM}/AA$ for all animals except lambs
(13a)	$X_{4\text{-}0\text{-}MN} = GOL/SAL \times (X_{3\text{-}4} + X_{2\text{-}4} + X_{0\text{-}4} - X_{4\text{-}0\text{-}AP})$ for lambs
(14)	$X_{3\text{-}4} + X_{2\text{-}4} + X_{0\text{-}4} = NA \times SAL \times P_{NTR}/AA$ for lambs
(15)	$X_{0\text{-}4} = GOL/SAL \times (X_{3\text{-}4} + X_{2\text{-}4} + X_{0\text{-}4})$ for lambs

Appendix 5. Analytical solution

$X_{0\text{-}1MF}$ = FRS × P_{FR} /AA

$X_{0\text{-}1CC}$ = WS × P_{WS} /AA

$X_{0\text{-}1SS}$ = SU × PD × PR /(1 – PR) /AA

$X_{1\text{-}0}$ = P_{EL}

$X_{0\text{-}3}$ = FDS × P_{FD} /AA

$X_{1\text{-}2}$ = ((Σ CR × P_{CR}) + (Σ APR × P_{APR} + Σ NA × P_{EM}) × GR + (SAL-GOL) ×NAlambs × P_{NTR})/AA

$X_{2\text{-}0}$ = ((Σ CR × P_{CR}) + (Σ APR × P_{APR} + Σ NA × P_{EM})× GR + (SAL-GOL) ×NAlambs × P_{NTR} – (1-GR)/GR × (Σ APR × P_{APR} + Σ NA × P_{EM})× GR – FDS × P_{FD} – (Σ APR × P_{APR} + Σ NA × P_{EM})× GR – (SAL-GOL) ×NAlambs × P_{NTR})/AA

$X_{2\text{-}3}$ = $X_{3\text{-}4}$ – $X_{0\text{-}3}$ = ((1-GR)/GR × (Σ APR × P_{APR} + Σ NA × P_{EM})× GR – FDS × P_{FD}) /AA

$X_{0\text{-}4}$ = GOL × NAlambs × P_{NTR} /AA

$X_{2\text{-}4}$ = (GR × (Σ APR × P_{APR} + Σ NA × P_{EM}) + (SAL-GOL) ×NAlambs × P_{NTR})/AA for 1996-2011

$X_{2\text{-}4}$ = ($X_{3\text{-}4}$ + $X_{2\text{-}4}$) × GR = (($X_{4\text{-}0\text{-}AP}$ + $X_{4\text{-}1}$) × GR = Σ NA × P_{NTR}× GR + (SAL-GOL) × NAlambs × P_{NTR})/AA for 1950-1995

$X_{3\text{-}4}$ = (1-GR) × (Σ APR × P_{APR} + Σ NA × P_{EM})/AA

$X_{4\text{-}1}$ = (Σ NA × P_{EM} + (SAL-GOL)/SAL × (NAlambs × P_{NTR} × SAL – APR × P_{APR}))/AA

$X_{4\text{-}0\text{-}MN}$ = GOL/SAL × (NA × P_{NTR} × SAL – APR × P_{APR})/AA

$X_{4\text{-}0\text{-}AP}$ = Σ APR × P_{APR}/AA

ΔS1 = (FRS × P_{FR} + WS × P_{WS} + SU × PD × PR /(1 – PR) + Σ NA × P_{EM} + (SAL-GOL)/SAL × (NAlambs × P_{NTR} × SAL – APR × P_{APR} – (Σ CR × P_{CR}) – (Σ APR × P_{APR} + Σ NA × P_{EM}) × GR – (SAL–GOL) × NAlambs × P_{NTR})/AA – P_{EL}

Appendix 6. Mineral fertilizer data

Table 7. Calculation of phosphorus concentration in mineral fertilizer productss in Norway for the year 2011.

Product type	Consumption, tons (Mattilsynet, 2011)	Phosphorus, tons (Mattilsynet, 2011)	Resulting phosphorus concentration
NP-fertilizer 12-23	857	197	22.99%
NPK-fertilizer 6-5-20	1829	95	5.19%
NPK-fertilizer 11-5-18	897	41	4.57%
NPK-fertilizer 12-4-18	22333	893	4.00%
NPK-fertilizer 15-4-12	930	34	3.66%
NPK-fertilizer 17-5-13	17	1	5.88%
NPK-fertilizer 18-3-15	34668	901	2.60%
NPK-fertilizer 19-4-12	3958	150	3.79%
NPK-fertilizer 21-3-8	1987	52	2.62%
NPK-fertilizer 21-4-10	11081	399	3.60%
NPK-fertilizer 22-2-12	40942	696	1.70%
NPK-fertilizer 22-3-10	106988	2996	2.80%
NPK-fertilizer 25-2-6	116838	1869	1.60%
NPK-fertilizer 27-3-5	21080	548	2.60%
P-fertilizer 0-8-0	148	12	8.11%
P-fertilizer 0-20-0	5	1	20.00%
PK-fertilizer 0-5-17	331	16	4.83%

Table 8. Phosphorus content in sold mineral fertilizer products in Akershus, Sør-Trøndelag and Rogaland for the year 2011 (tons). Data on sales from (Mattilsynet, 2011), P concentration from Table 7.

	Akershus			Sør-Trøndelag			Rogaland		
Product	Sold, tons	P conc.	P content, tons	Sold, tons	P conc.	P content, tons	Sold, tons	P conc.	P content, tons
NP-fertilizer 12-23	187	22.99%	43.0	46	22.99%	10.6	33	22.99%	7.6
NPK-fertilizer 6-5-20	269	5.19%	14.0	10	5.19%	0.5	155	5.19%	8.0
NPK-fertilizer 11-5-18	0	4.57%	0.0	4	4.57%	0.2	380	4.57%	17.4
NPK-fertilizer 12-4-18	1070	4.00%	42.8	499	4.00%	20.0	1619	4.00%	64.8
NPK-fertilizer 15-4-12	930	3.66%	34.0	0	3.66%	0.0	0	3.66%	0.0
NPK-fertilizer 17-5-13	2	5.88%	0.1	0	5.88%	0.0	0	5.88%	0.0
NPK-fertilizer 18-3-15	333	2.60%	8.7	2506	2.60%	65.2	5352	2.60%	139.2
NPK-fertilizer 19-4-12	394	3.79%	14.9	764	3.79%	29.0	0	3.79%	0.0
NPK-fertilizer 21-3-8	8	2.62%	0.2	208	2.62%	5.4	161	2.62%	4.2
NPK-fertilizer 21-4-10	955	3.60%	34.4	576	3.60%	20.7	0	3.60%	0.0
NPK-fertilizer 22-2-12	828	1.70%	14.1	2136	1.70%	36.3	5762	1.70%	98.0
NPK-fertilizer 22-3-10	16227	2.80%	454.4	6604	2.80%	184.9	398	2.80%	11.1
NPK-fertilizer 25-2-6	9384	1.60%	150.1	7440	1.60%	119.0	10466	1.60%	167.5
NPK-fertilizer 27-3-5	3884	2.60%	101.0	2289	2.60%	59.5	759	2.60%	19.7
P-fertilizer 0-8-0	0	8.11%	0.0	5	8.11%	0.4	5	8.11%	0.4
P-fertilizer 0-20-0	0	20.00%	0.0	0	20.00%	0.0	1	20.00%	0.2
PK-fertilizer 0-5-17	17	4.83%	0.8	3	4.83%	0.1	1	4.83%	0.0
Total			912			552			538

Appendix 7. Phosphorus excretion by livestock

Table 9. Phosphorus excretion by domestic animals. Comparison of data from different data sources.

Animal group	m^3 manure/ animal-year (5,7) [1]	kg P/ ton manure (1,3,8,9) [1]	kg P/ animal-year Calculated based on (1,3,5,7,8,9)	kg P/ animal-year (2) [1]	kg P/ animal-year (7) [1]	kg P/ animal-year (6) [1]	kg P/ animal-year (4) [1]	kg P/ animal-year (10) [2]
Horses	9-9.6	1	9.0-9.6		8	7		7
Dairy cows	18	0.48-0.67	8.6-12.1	14.82	12.6	14	14.4	14.4
Other cows	15.6	0.45-0.6	7.0-9.4			9.3	7.8	8
Other cattle								9
Heifers 0-24 months							10.6	
Oxen 0-18 months							7.5	
Young cattle 0-12 months	5-7.2	0.51-0.6	2.6-4.3		2.8	4.7		
Young cattle 12-24 months	7.2-10	0.51-0.6	3.7-6.0		3.8	4.7		
Heifers 0-6 months	3	0.51-0.6	1.5-1.8	2.24				
Heifers 6-12 months	6.6	0.51-0.6	3.4-4.0	2.24				
Heifers 12-24 months	10.2	0.51-0.6	5.2-6.1	4.6				
Oxen 0-6 months	3	0.51-0.6	1.5-1.8	3.3				
Oxen 6-12 months	6.6	0.51-0.6	3.4-4.0	3.3				
Oxen 12-18 months	10.2	0.51-0.6	5.2-6.1	4.21				
Sheep winter feeding	1-1.8	1.13-1.7	1.1-3.1	1.39	1.2	2		2
Dairy goats winter feeding	1-1.8	1.2	1.2-2.2		1.2	2		1.2
Pigs for breeding	4.5-4.8	0.89-1.5	4.0-7.2	6.88	5.9	5.6	5.8	5.8
Pigs for slaughtering	2-4.8	0.89-1.5	1.8-7.2	1.15	2	0.778	0.45	0.45
Laying hens	0.028-0.060	4-8.1	0.1-0.5	0.185	0.2	0.175	0.128	0.13
Broilers	0.010-0.070	6-7.2	0.1-0.5	0.010	0.070	0.010	0.006	0.04

[1] References: (1) Bioforsk, 2013; (2) Bolstad, 1994; (3) Daugstad et al., 2012; (4) Karlengen et al., 2012; (5) Knutsen and Magnussen, 2011; (6) Lovdata, 2013; (7) Nesheim et al., 2011; (8) Tveitnes, 1993; (9) Øgaard, 2008.
[2] (10) Used in the present study for the period from 1996 to 2011.

Appendix 8. Feed production data

Table 10. Raw products sold for feed production in 2010, tons

Product	Raw products sold in Norway, tons	Phosphorus concentration in raw products	Resulting P content, tons	Phosphorus concentration in feed
Corn (Mais)	54113[1]	0.10%[2]	54	
Sorghum	23168[1]	0.29%[2]	66	
Corn (Mais) groats	31469[1]	0.10%[2]	31	
Wheat	232569[1]	0.37%[2]	863	
Rye/Triticale	12525[1]	0.36%[2]	44	
Barley	507881[1]	0.29%[2]	1447	
Oats	255013[1]	0.52%[2]	1316	
Bran	74597[1]	1.10%[2]	821	
Molasses	61719[1]	0.03%[3]	19	
Herring flour	12374[1]	0.02%[4]	2.4	
Corn (Mais) gluten	24349[1]	0.57%[5]	139	
Soya flour	215527[1]	0.60%[2]	1293	
Rapeseed pellets	91091[1]	0.30%	273	
Oilseeds, peas	19594[1]	0.31%[2]	61	
Fish silage	9319[1]	1.90%[6]	177	
Vitamin/mineral	78874[1]	5%	3944	
Total	1811198		10550	0.58%

References:
[1] Data from the Norwegian Agricultural Authority (Breen, 2013)
[2] Matvaretabellen, 2013
[3] Nutritiondata, 2013
[4] Tangkanakul et al.,, 2005
[5] Stein, 2013
[6] Hertrampf and Piedad-Pascual, 2000

Appendix 9. Phosphorus balance in the process "Animal Husbandry"

Table 11. Phosphorus balance of main animal categories in Akershus, Sør-Trøndelag and Rogaland. Intake is the sum of phosphorus consumed with feed and fodder and through grazing both on cultivated and uncultivated land (quantified using mass balance). Deposition is assumed to be equal to amount of phosphorus in animal products (carcasses of animals slaughtered, milk and eggs). Excretion is phosphorus amount in manure excreted both on cultivated and uncultivated land.

County	Animal groups	1997-1999 Intake	1997-1999 Deposition	1997-1999 Excretion	2009-2011 Intake	2009-2011 Deposition	2009-2011 Excretion	Unit
Akershus/ Oslo	Horse	0.2	0.0	0.2	0.3	0.0	0.3	kg P/ha-yr
	Cattle	4.1	0.6	3.4	3.2	0.5	2.6	kg P/ha-yr
	Sheep/Lambs	0.5	0.5	0.0	0.5	0.4	0.1	kg P/ha-yr
	Goats	0.0	0.0	0.0	0.0	0.0	0.0	kg P/ha-yr
	Pigs	0.8	0.0	0.8	0.7	0.0	0.7	kg P/ha-yr
	Poultry	1.4	0.3	1.1	1.5	0.3	1.2	kg P/ha-yr
	Total	6.9	1.4	5.6	6.2	1.3	4.9	kg P/ha-yr
Sør-Trøndelag	Horse	0.2	0.0	0.2	0.3	0.0	0.2	kg P/ha-yr
	Cattle	17.5	3.0	14.6	14.0	2.5	11.5	kg P/ha-yr
	Sheep/Lambs	3.3	0.3	3.0	3.4	0.2	3.2	kg P/ha-yr
	Goats	0.0	0.0	0.0	0.0	0.0	0.0	kg P/ha-yr
	Pigs	0.7	0.1	0.6	0.5	0.1	0.4	kg P/ha-yr
	Poultry	1.3	0.3	1.1	5.8	1.2	4.5	kg P/ha-yr
	Total	23.0	3.6	19.4	24.0	4.1	19.9	kg P/ha-yr
Rogaland	Horse	0.2	0.0	0.2	0.2	0.0	0.2	kg P/ha-yr
	Cattle	22.5	3.2	19.3	19.2	3.8	15.4	kg P/ha-yr
	Sheep/Lambs	7.0	1.4	5.6	8.3	2.0	6.3	kg P/ha-yr
	Goats	0.02	0.00	0.02	0.02	0.00	0.02	kg P/ha-yr
	Pigs	2.9	0.2	2.6	3.7	0.3	3.4	kg P/ha-yr
	Poultry	3.3	0.7	2.6	7.5	1.4	6.1	kg P/ha-yr
	Total	35.9	5.6	30.3	39.0	7.5	31.6	kg P/ha-yr