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Influence of soil phosphorus and manure on phosphorus leaching in Swedish topsoils

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2 Abstract

3 In Sweden, subsurface transport of phosphorus (P) from agricultural soils represents 4 the primary pathway of concern for surface water quality. However, there are mixed 5 findings linking P in leachate with soil P and limited understanding of the interactive 6 effects of applied P sources and soil test P on P leaching potential. Identifying soils 7 that are susceptible to P leaching when manure is applied is critical to management 8 strategies that reduce P loadings to water bodies. Intact soil columns (20 cm deep) 9 from five long-term fertilization trials across Sweden were used in leaching 10 experiments with simulated rainfall to explore the interactive effects of dairy cow 11 (Bos taurus L.) manure application, soil test P and cropping system. Strong 12 relationships were observed between ammonium-lactate extractable P in soil and 13 dissolved reactive P (DRP) concentrations in leachate, although regression slopes varied across soils. For three soils, application of manure (equal to 21-30 kg P ha⁻¹) to 14 15 the soil columns significantly increased DRP leaching losses. The increase in DRP 16 concentration was correlated to soil test P, but with wide variations between the three 17 soils. For two soils leachate P concentrations after manure addition were independent 18 of soil P status. Despite variable trends in P leaching across the different soils, P 19 concentrations in leachate were always moderate from soils at fertilization rates 20 equivalent to P removal with harvest. Results clearly stress the importance of long-21 term P balance to limit P leaching losses from Swedish agricultural soils.

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23 Key words Phosphorus leaching · Ammonium lactate-extractable soil P · Rainfall

24 simulation · Long-term fertility experiments · Manure management

1 Introduction

2 Eutrophication of the Baltic Sea has increased algal blooms and anoxic conditions, 3 prompting an international accord to curb loadings of nutrients, particularly 4 phosphorus (P) (HELCOM 2007). Agriculture is the main source of Sweden's P 5 contribution to the Baltic Sea (SEPA 2008), accounting for roughly 50% of the total 6 anthropogenic load. Since agriculture in Sweden and Finland is mostly located on flat 7 landscapes where soils are drained (open ditches and subsurface tile drains), 8 subsurface transport of P represents the primary pathway of concern for downstream 9 surface water quality (Turtola and Jaakkola 1995; Ulén 1995). As leaching serves to 10 connect P at the soil surface with subsurface drains, understanding the factors 11 controlling P leaching through agricultural soils is key in assessing practices and 12 strategies aimed at mitigating diffuse P loads from Swedish agriculture (Ulén et al. 13 2007).

14 An extensive body of work has documented P leaching through soils, 15 emphasizing the soil-specific nature of P leaching potential and the varying influence 16 of management variables on P leaching processes (e.g. Djodjic et al. 2004; Kleinman 17 et al. 2009; Kang et al. 2011). Phosphorus leaching from soils varies widely, from 18 almost undetectable levels to several mg per litre of drainage water from arable and 19 grassland soils (Brookes et al. 1997; Sims et al. 1998). Bypass or preferential flow via 20 soil macropores represents one of the major transport mechanisms of P leaching 21 through well-structured soils (Jensen et al. 1998; Stamm et al. 1998; Simard et al. 22 2000). As a result, cropping systems or practices that preserve soil structure and 23 promote the maintenance of macropores (e.g. no-till and perennial forage systems) are 24 particularly susceptible to P leaching losses (Sims et al. 1998; Chardon and van 25 Faassen 1999). Phosphorus leaching was once seen as a phenomenon restricted to

coarse-textured soils but has now been widely documented in finer textured soils with extensive macropore networks (Djodjic et al. 1999; van Es et al. 2004).

2

3 Research to date has yielded mixed findings on the relationship between soil 4 test P and P leaching potential. A study by Heckrath et al. (1995), summarizing findings from shallow tile drains established in the Broadbalk (UK) cropping systems 5 6 trials, identified a clear threshold in Olsen-P of surface soils above which the potential 7 for leaching significantly increased. The 'change point' analysis performed by 8 Heckrath et al. (1995) sparked an array of studies investigating critical thresholds of 9 soil P above which P solubility and/or mobility increased significantly. For instance, 10 McDowell and Sharpley (2001) and Maguire and Sims (2002) reported significant 11 change points in the relationships between 0.01 M CaCl₂ extractable P and P sorption 12 saturation of surface soils, respectively, and P concentration in leachate from column 13 leaching experiments. In Sweden, Börling et al. (2004) reported strong relationships 14 between ammonium lactate (AL) extractable P, the dominant agronomic soil P test for 15 Scandinavia, and 0.01 M CaCl₂ extractable P, which is considered an indicator of 16 potentially leachable soil P. In another Swedish study, Ulén et al. (2011) found AL-17 extractable P to be a reliable P risk index for soil profiles with high clay content in a 18 catchment with overall balanced soil P level. However, in an intact soil column 19 leaching study in which a range of Swedish soils was assessed, no relationship 20 between AL-extractable P and leachate P was detected (Djodjic et al. 2004).

Application of manure to soils can temporarily elevate P concentrations in leachate from these soils, primarily as a result of transfer of manure P to infiltrating water. 'Rapid incidental transfers' (Preedy et al. 2001) of manure P to leachate are well documented (Geohring et al. 2001; Kleinman et al. 2005; 2009), with the greatest contributions of manure to leachate P typically occurring in the first leaching events after application (Chardon et al. 2007). In general, soluble P in the applied manure serves as the major source of P in leachate. Indeed, Kang et al. (2011) found that the water-extractable P concentration in manures and mineral fertilizers applied to soil columns was correlated with loads of dissolved reactive P (DRP) in leachate. However, little is known about the interactive effects of applied manures and fertilizers and antecedent soil properties and soil P status.

7 Given the lack of information on the relationship between soil P leaching and 8 soil test P measured as AL-extractable P in Swedish soils, and the limited insight into 9 the interactive effects of soil test P and applied P sources on P leaching potential, this 10 study sought to examine the role of soil test P and applied P sources on P leaching 11 from dominant types of agricultural soils in Sweden. Focus was on the topsoil which 12 usually has higher concentration of P than deeper soil-layers, and P leaching from the 13 topsoil was considered as potential P leaching which may reach deeper soil layers and 14 drainage tiles. Specific aims of the study were to investigate: i) relationships between 15 AL-extractable P and P leaching; ii) changes in P leaching from soils with varying 16 AL-extractable P following manure application; and, iii) effects of cropping system 17 properties on P leaching.

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19 Materials and Methods

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21 Site background and Soil description

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The Swedish long-term fertility experiments (LTFEs), initiated between 1957 and 1969, consist of 12 field trials located across Sweden, representing dominant soils and cropping systems in the country. More details on these experiments can be found in

1 Carlgren and Mattsson (2001) and Kirchmann et al. (1999; 2005). Soil columns were 2 collected from five of these 12 soils: Fjärdingslöv sandy loam (Oxyaquic. Hapludoll); 3 Ekebo loam (Oxyaquic Hapludoll); Bjertorp silty clay loam (not classified); 4 Klostergården silty clay loam (Oxyaquic Haplocryoll); and Högåsa loamy sand (Humic Dystrocryept). The experimental design of the LTFEs is similar for all soils, 5 6 with application rates of P varying in relation to P removed by harvested products on duplicate field plots (6.25 m x 20 m) (Table 1). The different P application rates have, 7 8 over time, resulted in substantially different soil P concentrations across treatments 9 within soils (Table 2), also demonstrated by Ehde (2012). Field plots with the 'low P' 10 treatment receive no mineral P applications, while P removed by harvested products is 11 replaced in the 'medium P' treatment. The 'high' and 'very high P' application rates 12 (Table 1) were intended to achieve slow and rapid increase, respectively, in soil P 13 status. Recent intensive soil monitoring of LTFEs has demonstrated that the AL-14 extractable P values are only elevated for about 2-4 months after mineral P application 15 and thereafter decline to approximately its original level. In the P replacement 16 treatment any permanent increase in AL-extractable P is minor (Djodjic and Mattsson 17 2013).

18 The crop rotation period is four or six years and includes cereals (barley 19 (Hordeum vulgare L.), winter wheat (Triticum aestivum L.) and oats (Avena sativa 20 L.)), oilseed rape (Brassica napus L.), and, at Ekebo and Fjärdingslöv, sugarbeet 21 (Beta vulgaris L.). Crop rotations are typical for the regions where the experiments 22 are situated. Two cropping systems are represented for each soil. In the manured 23 cropping system (MCS), a perennial forage crop is included, harvest residues are 24 removed and dairy manure (30 ton ha⁻¹) is applied every sixth year, except on soils in 25 southern Sweden (Ekebo loam and Fjärdingslöv sandy loam) where manure (20 ton

ha⁻¹) is applied every fourth year. On average, 9 kg P ha⁻¹ yr⁻¹ have been applied as 1 2 manure. The previous manure and P fertilizer application to this cropping system was 3 made at least six months prior to soil column collection. When manure is applied, the 4 rates of mineral fertilizer P are adjusted for the amounts of P applied in the manure. In the unmanured cropping system (UMCS) the harvest residues are incorporated into 5 6 the soil. The experimental fields have been conventionally ploughed on a regular basis. Normal tillage depth for conventional ploughing is 23 cm and the depth of the 7 8 tile drains is about 90 cm.

9 The carbon (C) content prior to soil column collection (2005-2007) was on 10 average 2.0% of air-dry topsoil in MCS and 1.8% in UMCS at the study sites 11 (Börjesson 2012). Other important soil properties are shown in Table 3. Except for 12 Fjärdingslöv, all soils are acidic in reaction.

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14 Soil and Soil Column Collection

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Topsoil samples (0-20 cm depth) were taken in autumn 2007 from all plots at the five sites, within the normal sampling routine for Swedish LTFEs (Börjesson 2012). At least 10 subsamples were randomly obtained from each plot and pooled into one composite sample.

Intact soil columns were collected from the four P level treatments in both MCS and UMCS at the five sites. In each field plot, four soil columns were taken at least 1.5 m from any edge of the field plot, from an area of approximately 2.25 m² (160 columns in total). The collection took place after harvest but before any tillage treatment in autumn. Soil columns from Fjärdingslöv sandy loam and Ekebo loam were collected in September 2007 and from Bjertorp silty clay loam in October 2008.

1 These soil columns were extracted by gently pressing polyvinyl chloride pipes (20 cm 2 in diameter and 20 cm long) into the soil using soil anchors and a hydraulic pump 3 system (Jarvis et al. 2008). A sharp steel cutting ring was placed at the lower end of 4 the pipes to facilitate insertion of the pipes into the soil. The Klostergården silty clay loam and Högåsa loamy sand soil columns were collected in September 2009. In 5 6 those cases, the plastic pipes were gently pressed into the soil by a tractor with a front 7 loader (Liu et al. 2012a). The columns were extracted and sealed using lids and plastic 8 bags.

9 Soil columns were then stored at 2 °C for approximately six months until they 10 were prepared for the leaching experiment. Column preparation involved removing 11 excess soil at the bottom along soil aggregate surfaces with a knife and vacuuming 12 loose particles. A nylon fabric with a mesh size of 50 µm was then placed at the 13 bottom of each column. Finally, the columns were placed on a base so that free 14 drainage could occur during the leaching experiment and leachate water was collected 15 in glass bottles.

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17 Leaching experiment

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19 Experiment 1

The leaching experiment was carried out in an indoor rainfall simulator (approximately 20 °C) before and after application of manure to the columns, in order to assess the contribution of soil P and applied manure P to leachate. The rainfall simulation was performed in rounds with 16 soil columns. In each round, all the soil columns came from the same soil and cropping system, but had different P levels. Simulated rainfall consisted of two parts de-ionized water and one part tap water, to

1 better resemble the chemical composition of natural rain water. The electrical conductivity of this simulated rainfall was 15 mS m⁻¹ and the chlorine (Cl) 2 concentration 13 mg L⁻¹. Irrigation was applied from air-atomizing spray nozzles 3 located 1.2 m above the centre of each soil column (Larsbo et al. 2009). The rainfall 4 intensity was 10 mm h⁻¹ (standard deviation 2.6 mm h⁻¹) and the simulated rainfall 5 6 was applied in three events lasting 2.5 h each, at 2-day intervals (76 mm in total) (Figure 1). The amount of applied water is more than would be expected under natural 7 8 field conditions in Sweden, but was necessary to produce a sufficient volume of 9 leachate for chemical analyses. After each 2.5 h rainfall simulation, the leachate was 10 collected and stored at 6 °C prior to analysis.

11

12 Experiment 2

13 Following an initial period of leaching with three simulated rainfall events for all soil 14 columns, fresh dairy cow (Bos taurus L.) manure collected on two occasions was 15 applied to soil columns from the manured cropping system (Figure 1) at a rate corresponding to 30 tonnes ha⁻¹. As a result of differences in manure composition 16 between collection occasions, the Fjärdingslöv sandy loam, Ekebo loam, and Bjertorp 17 silty clay loam columns received 30 kg P ha⁻¹ and the Klostergården silty clay loam 18 and Högåsa loamy sand columns received 21 kg P ha⁻¹. When the manure was 19 20 applied, 1 cm of soil was removed from the top of the soil column, the manure was 21 distributed over the surface (to within approx. 2 cm from the edge), and the soil was 22 then replaced on top of the soil column. After manure application, approximately 1.3 23 mm of water was added on two occasions with the rainfall simulator (Figure 1), to let 24 some of the P in the manure move downwards in the column and equilibrate with the soil. A second period of leaching with three rainfall events was then carried out withsoil columns from MCS (Figure 1).

3

4 Analysis of water and soil samples

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The concentration of total P (TP) was measured on unfiltered leachate samples after
acid digestion with potassium persulphate in sulphuric acid, and dissolved reactive P
(DRP) was measured in leachate after filtration (Schleicher & Schüll GmbH, Dassel,
Germany, membrane filter with pore diameter 0.2 μm). Both analyses were made
colorimetrically according to the method issued by the International Standards
Organization (ISO, 2003).

12 Soil samples were extracted with the AL method according to Egnér et al. 13 (1960). Phosphorus determination on the extracts was conducted by inductively 14 coupled plasma atomic emission spectroscopy (ICP-AES, OPTIMA 3000DV; Perkin 15 Elmer, Waltham, USA) according to Swedish standards (1993). The AL extraction 16 method is common in the Baltic region. Recent comparisons of the AL method with 17 the Olsen-P and Mechlich-3 methods using a set of 99 topsoil samples from seven 18 sites showed generally strong correlations with both Olsen-P and Mehlich-3 (Eriksson 19 et al. 2013). However, for acidic Swedish clay soils the AL method extracted 3.6 20 times more P than Olsen-P and 1.7 times more than Mehlich-3.

21

22 Calculations and statistical analysis

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24 Statistical analyses were performed using SAS software, version 9.2 (SAS Institute 25 2008). A mixed model approach was used for both experiments, following the

1 example of Littell et al. (2006). The mixed model included both cropping system and 2 soil as fixed categorical factors and AL-extractable P as covariate. Measurements 3 made in different soil columns within each plot were included as nested factors and 4 the different rainfall simulations were included as repeated factor with the correlation structure compound symmetry. The Kenward-Roger method was used to determine 5 6 degrees of freedom and fixed effects standard error. Assumptions of normality were checked by residual plots. Statistical results were considered significant at $\alpha = 0.05$. 7 8 Pairwise comparisons of means were adjusted using the Tukey-Kramer multiple 9 comparisons test. Using the mixed model, we also estimated and compared the slopes 10 of the regression of P leaching to the covariate AL-extractable P in the soil. Since the 11 rainfall simulations were performed in a consistent way, we did not account for the 12 fact that rainfall simulations were made in blocks with soil columns from the same 13 soil and cropping system.

14 For experiment 1, the dependent variables DRP and TP were transformed with 15 the natural logarithm (ln) because residuals were then closer to a normal distribution 16 and homoscedastic. Data on AL-extractable P were also In-transformed to linearize 17 the relationship between AL-extractable P and the dependent variables. The 18 dependent variables leachate volume and percentage of DRP in TP were not ln-19 transformed because residual plots were satisfactory without transformation. Cropping 20 system, soil and the interaction between cropping system and soil were set as fixed 21 effects. The ln-transformed values of AL-extractable P (ln-AL-P) were included as a 22 covariate, including the interaction between cropping system and ln-AL-P, the 23 interaction between soil and ln-AL-P, and the interaction between soil, cropping 24 system and ln-AL-P. The random effects in the model were as described above. The magnitude of the response in P leaching to elevated AL-extractable P level, also 25

referred to as the 'extraction coefficient' (Sharpley et al. 2002), was quantified by
 calculating the slope of the regression line for each soil.

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3 For experiment 2, with recent manure application, the increase in DRP and TP 4 leaching was calculated by subtracting the average concentration in leachate from 5 three rainfall simulations before manure application from the average concentration in 6 leachate from three rainfall simulations after manure application. We did not find any 7 time trend in concentrations between the three consecutive rain simulations and, 8 hence, the mean values of these were used in calculations. The increases in TP and 9 DRP were used as dependent variables and the residual plots were satisfactory 10 without ln transformation. The random effects in the model were due to the nested factors 'soil column' within the same soil and P level. Consequently, the interaction 11 12 between soil and P level was a random effect, while soil, AL-extractable P and the 13 interaction between soil and AL-extractable P were fixed effects. The effect of 14 application rate could not be separated from the effect of soil, and was not accounted 15 for in the analysis. The effect of application rate is therefore part of the effect of soil 16 in this model.

The percentage of DRP in TP in leachate water from experiment 2 was included in a mixed model as a dependent variable and the residual plots were satisfactory without ln transformation of the dependent variable. The fixed effects were: ln-AL-P, soil and the interaction between ln-AL-P and soil. The random effects in the model were due to the nested factors 'soil column' within the same soil and P level, and 'rainfall simulation' within the same 'soil column'.

2 **Results and Discussion**

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4 Experiment 1 – before recent manure application

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6	Leachate	Volumes
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No differences in leachate volume were found between the cropping systems in experiment 1 (Table 4), indicating that the variability in amount of applied water between simulation rounds was minor. Therefore, significant differences in leachate volumes between the different soils (p<0.04) probably reflect the varying hydraulic properties of the soils. Ekebo loam leached least water, while Högåsa loamy sand, which was the coarsest soil, was one of the two soils that leached the most (Table 5).

13 Another factor possibly explaining varying leachate volumes could be the 14 initial moisture conditions at the start of the experiment. All soil columns were 15 collected in September-October, which often is a time with moist soil conditions in 16 Sweden, and none of the soils were particularly dry. Soil columns were then stored 17 approximately six months at 2 °C prior to rain simulations. Initial soil moisture was 18 not measured, and was likely different between soil columns from different soils. 19 Slightly smaller leachate volumes in the first rainfall simulation compared with the 20 second and third were recorded for three of the soils (Ekebo loam, Högåsa loamy sand 21 and Klostergården silty clay loam), most likely due to lower initial soil water content. 22 However, total leachate volumes for individual soils were quite stable.

During three rainfall simulations, 76 mm of water was applied to the soil columns and based on values of porosity in Table 3, this represents 83-109% of the pore volume in the soil columns. However, the entire pore volume is usually not

1 equally active in water and solute transport (Flury et al. 1994; Gerke 2006) and the 2 effective pore volume is much smaller than the total pore volume in most soils. Some 3 reported values of effective pore volume for Swedish soils vary between 0.1-0.9 4 (Bergström et al. 2011; Ghafoor et al. 2013). The effect of this is that water applied on 5 top of a soil column is likely to reach the bottom before one entire pore volume has 6 passed. Only one soil in the present study was a weakly structured sand soil, while the 7 other soils contained 14-48% clay and in such soils preferential solute transport has 8 been shown to occur (Koestel et al. 2012). Both intensive rain and high water content 9 in the soil, which are conditions that were valid in the present study, are factors which 10 enhance macropore flow (Jarvis, 2007). Leachate water collected and measured in this 11 study was therefore a mix of water applied as simulated rainfall and water present in 12 the soil pores at the start of the experiment. However, bypass of binding sites by 13 preferential flow, breakthrough curves or other physical effects from the soil structure 14 were not evaluated in the present study.

Ponding was observed in a few soil columns directly after rainfall simulation, but generally the water infiltration was satisfactory. No systematic trends in ponding were observed by soil or cropping system and therefore the ponding was not likely to have had a large effect on the overall results in this experiment.

19

20 Soil P status and leachate P concentrations

Phosphorus concentration in leachate from the soil columns in the first three rainfall simulations increased with increasing AL-extractable P in the soil (Figure 2). Corresponding slopes for all soils were significantly different from zero for both DRP and TP as dependent variable (Table 5). The concentrations in the leachate from the rainfall simulations were relatively low for the LTFE plots with long-term fertilization

1 rates equivalent to P removal with harvested products. The mean TP concentration in the leachate was 0.12 mg L⁻¹, compared with nine-fold higher concentrations from 2 plots representing P application rates of replacement of P +15 or +20 kg P ha⁻¹ yr⁻¹. 3 The relationship between AL-extractable P and P leachate concentrations varied 4 across soils from different sites, with a significant interaction between soil and AL-5 6 extractable P (Table 4) and significantly different slopes between most soils (Table 5). 7 However, in the first rainfall simulation we cannot rule out that difference in P 8 leaching between soils was affected by different initial soil moisture. The results 9 confirm the controlling role of soil P, measured as AL-extractable P, on P leaching 10 from surface soils and are in agreement with Börling et al. (2004), who observed 11 significant relationships between AL-extractable P and CaCl2-extractable P (a 12 surrogate for leachate P) for surface soils (0-20 cm) from the same LTFEs.

13 Phosphorus-concentrations were relatively stable in rain simulations 2-3, 14 although less than the theoretical pore volume had leached through. In a study similar 15 to the present, Liu et al. (2012b) found constant P concentration in leachate from clay 16 loam topsoil columns even after eight rainfall simulations, representing approximately two pore volumes. The present experiments represented conditions with a high 17 18 groundwater level near water saturation in order to reproduce conditions with a high 19 risk of P leaching from the actual topsoil. Rubaek et al. (2010) were also able to 20 demonstrate relationships between P leaching and topsoil Olsen-P under conditions 21 favouring preferential flow. They used suction-controlled lysimeters in order to 22 simulate unsaturated conditions through topsoil (20 cm) columns. The results obtained 23 in the present study for topsoil contrast with those reported from deeper soil cores (1 24 m) from the Swedish LTFEs, for which no relationships were observed between AL-25 extractable P and P leaching (Djodjic et al. 2004). This suggests that AL-extractable P is an important explanatory variable for P leaching from surface soils, but that subsoil
 properties and water transport pathways can have a modifying effect on P leaching in
 deeper soil layers.

4

5 *Proportion of DRP in TP in leachate*

6 The percentage of DRP in TP in leachate was significantly different at different soil concentrations of AL-extractable P (Table 4) and the percentage increased with 7 8 increasing soil AL-extractable P (Figure 3). This is in agreement with Rubaek et al. 9 (2010), who observed that the greater the concentration of Olsen-P in the soil, the 10 greater the percentage of DRP in TP in leachate water. It is also in agreement with 11 Ahlgren et al. (2013), who used magnetic resonance spectroscopy to investigate P 12 forms in three of the Swedish LTFEs studied here. They found that the amount of 13 orthophosphate increased with increasing AL-extractable P, but that the amount of 14 phosphate monoesters was rather stable. As a result, the proportion of orthophosphate 15 in the soil increased with increasing soil P status. The interaction between AL-16 extractable P and cropping system was not considered significant (p=0.052).

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18 Cropping system, AL-extractable soil P and P concentration in leachate

Phosphorus removal with harvested products in the five LTFEs studied here has been less than the permitted maximum application rate of 22 kg P ha⁻¹ yr⁻¹ set by Swedish animal density regulations. On average, more P has been removed by harvested products in MCS (18 kg ha⁻¹ yr⁻¹) than in UMCS (12 kg ha⁻¹ yr⁻¹), and consequently application rates of P have been higher in MCS (Börjesson 2012). However, at the time of this study, AL-extractable P was generally lower in the MCS plots on Bjertorp silty clay loam and Klostergården silty clay loam (Table 2). In the three loamy soils with minor clay content (Ekebo, Fjärdingslöv, and Högåsa), historical P fertilization
at higher rates has generally led to a greater AL-extractable P in MCS than in UMCS.
The relative difference in applied P between MCS and UMCS was highest in plots
with low P level which received no mineral P (UMCS) or only manure (MCS).

5 Significant differences in both DRP and TP leaching from topsoil columns 6 were observed between the two cropping systems (Table 4), but these differences 7 were not consistent and were only significant for two of the soils. For Fjärdingslöv 8 sandy loam, the mean DRP and TP concentrations in the leachate were higher from 9 UMCS than from MCS (Table 5), despite UMCS having a generally lower soil P 10 status (Table 2). In contrast, for Klostergården silty clay loam, the mean TP 11 concentration in the leachate was higher from MCS than from UMCS (Table 5) 12 although this soil represented one of two with generally lower AL-extractable P in 13 MCS compared with UMCS (Table 2).

14 A high content of organic matter is suggested to be a factor which could 15 indirectly increase the P sorption capacity by inhibition of Al (aluminium) oxide 16 crystallization (Borggaard et al. 1990). Positive correlations between organic C and P 17 sorption were also reported in an earlier study which included the five soils used in 18 the present study (Börling et al. 2001). However, this could not be demonstrated in the 19 present study, where the relative differences in soil organic C between MCS and 20 UMCS were small, with an average range of only 6% for Klostergården and 15% for 21 the other soils. In addition, the general level of organic C content in the soil was rather 22 low (1.2 - 2.2%).

The three-way interaction between AL-extractable P, soil and cropping system was significant (p=0.03) when TP concentration in leachate was the dependent variable (Table 4). This means that there were differences in slopes between cropping

systems when looking at the soils separately, with a steeper slope in the UMCS for Fjärdingslöv sandy loam, Högåsa loamy sand, and Klostergården silty clay loam. Bjertorp silty clay loam had no differences in slopes between cropping systems, while Ekebo loam had a steeper slope in the MCS. However, differences in slopes were small. In conclusion, long-term moderate manure application in the LTFEs, which increased crop yield, did not seem to have had any overall effect on the P concentration in leachate, and hence on potential P leaching.

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9 Experiment 2 - P leachate concentration after recent manure application

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11 Soil P status and P concentration in leachate

12 Columns from Bjertorp silty clay loam, Fjärdingslöv sandy loam and Ekebo loam received 8 kg P ha⁻¹ more than the maximum application rate (22 kg P ha⁻¹ yr⁻¹) set by 13 14 Swedish animal density regulations. In Bjertorp silty clay loam and Fjärdingslöv 15 sandy loam, this application increased the mean concentration of DRP and TP in 16 leachate and the increase was correlated to AL-extractable P (Figure 4). In 17 Klostergården silty clay loam, which had received a P application very close to the maximum rate (21 kg P ha⁻¹ yr⁻¹), the DRP and TP concentration in leachate also 18 19 increased, but only the increase in DRP concentration was correlated to AL-20 extractable P (Table 7). However, significant differences in slopes and least squares 21 means were found across soils after recent manure application (Table 7). In Ekebo loam and Högåsa loamy sand there were no increase in DRP after manure application 22 23 (Table 7) and neither showed any significant correlation to AL-extractable P (Table 24 7). In experiment 1, for Högåsa loamy sand and Ekebo loam the DRP concentration in 25 leachate was higher before recent manure application at the high AL-extractable P

1 level than after manure application at the medium P level, which was also the case for 2 Klostergården silty clay loam. Consequently, the long-term build-up of soil P was 3 more important for leachate P concentration than recent manure application to these 4 soils. Similarly, Hahn et al. (2012) found that manure application did not override the 5 effect of soil P status on the P concentration in runoff water. Likewise, Liu et al. 6 (2012a) found that the build-up of soil P with long-term manure application was more important for potential P leachate losses than a single manure application to loamy 7 8 sand.

9 Temperature was approximately 20 °C while soil columns were placed in the 10 rainfall simulator and some mineralization of organic P may have taken place which 11 could have changed P concentration in soil solution over time. However, He et al. (2004) found that when dairy manure was mixed with soil at a rate of 28 mg dairy 12 manure P kg⁻¹ dry soil and incubated at 25 °C, the water extractable inorganic P 13 14 remained stable over time. In our experiment the application rate was <10 mg dairy manure P kg⁻¹ dry soil (calculated with dry bulk densities from Kirchmann et al. 1999; 15 16 2005), although, we did not mix the manure with the soil.

17

18 *Proportion of DRP in the leachate*

The percentage of DRP in TP (Fig. 3) was 49% on average across all soils after manure application, with Bjertorp silty clay loam having the highest percentage of DRP in TP and Ekebo loam the lowest (Table 7). Both soil and AL-extractable P had a significant effect on the percentage of DRP in TP after manure application (Table 6). The percentage of DRP in TP increased as AL-extractable P increased (Fig. 3) and slopes were not significantly different between soils (Table 6). Total P concentration in the manure was 0.69 and 1.0 g kg⁻¹ and reported literature values of the proportion 1 of inorganic P in Dairy cow manure are e.g. 54% (Pagliari and Laboski, 2012) and 2 63% (Barnett, 1994). However, of the P present in the leachate water, we cannot differentiate between the P that came from the manure and the P that came from the 3 4 soil and pore water solution present in the soil. Transport pathways differ between soils due to differences in shape and size of the pores, and this affects how fast 5 6 manure P is transported through the soil. Manure applications may also affect P adsorption properties in soils (Bolan et al., 1994), and consequently affect the source 7 8 of P that is found in leachate.

9

10 Other P leaching indices for leachate P concentrations

11 No totally consistent comparisons can be made with P saturation indices such as P 12 sorption maximum, P buffering capacity and degree of P saturation (Table 3), since 13 these parameters were only measured in UMCS and in experiment 2 we applied 14 manure to soil columns from MCS. However, it is interesting to note that Högåsa 15 loamy sand and Ekebo loam, i.e. the two soils with rather constant P concentration in 16 the leachate after manure addition, irrespective of soil P status, had the lowest degree 17 of P saturation. Fjärdingslöv sandy loam had the second highest degree of P saturation 18 as well as second highest DRP concentration in leachate. On the other hand, Bjertorp 19 silty clay loam had the highest concentration of DRP in leachate but not very high 20 degree of P saturation.

Manure application may increase soil P solubilization and decrease P adsorption due to increased amounts of organic acids (Bolan et al., 1994). This is a possible mechanism in the three soils in the present study where DRP leaching increased as AL-extractable P increased after recent manure application. In contrast, Guppy et al. (2005) suggested that increased leaching of DRP after manure application might be the result of P from the actual manure. This is in agreement with
the findings for Högåsa loamy sand and Ekebo loam in the present study, which had
more or less the same P leaching after recent application irrespective of soil P status.

4 Several other factors such as clay content or pH could not solely explain the 5 different results between the five soils in the present experiment. Overall, the results show the complexity of P leaching from the soil. Furthermore, the presence of 6 7 biopores and cracks in the soil columns was not evaluated in this study, but is likely to 8 have an effect on P leaching losses after P application (Djodjic et al. 1999; Glaesner et 9 al. 2011). Interactive effects between applied P sources of different types and soil test 10 P on P leaching potential need to be further investigated in order to identify soils that 11 are especially susceptible to P leaching and to avoid unnecessarily high P loadings to 12 water bodies.

13

14 Conclusions

15 This study showed that soils that receive moderate and infrequent applications of 16 manure in a cropping system which includes levs may not leach more P than an 17 unmanured cropping system with cereals. However, soils behave differently regarding 18 P leaching after recent manure application around current maximum permitted rates 19 set by Swedish animal density regulations. In some soils, there was an increase in 20 Dissolved Reactive P (DRP) concentration after dairy cow manure application and 21 this increase was significantly correlated to soil test P. In other soils, there was no 22 corresponding increase in P leaching after recent manure application. Identification of 23 soils that are especially susceptible to P leaching when manure is applied is important 24 in efforts to reduce P loadings to water bodies. Previous reports of an increased risk of 25 P leaching from soils with high soil test P were confirmed in this study, although a wide variation in the relationship was observed across soils. Phosphorus leaching was low at long-term fertilization rates equivalent to P removal with harvested products, which stresses the importance of long-term P balance to limit P leaching losses. Such a fine-tuned strategy also requires determination of P removal with harvested products, which in the Swedish long-term fertility experiments has been less than the national limit for P application, 22 kg P ha⁻¹ yr⁻¹, set through animal density regulations.

8

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- 1 Figure captions
- 2

Fig. 1 Timeline showing rainfall simulations and manure application. Manure was
only applied to soil columns from the manured cropping system (MCS). Simulated
rainfall was applied only three times (25 mm per event) to soil columns from the
unmanured cropping system (UMCS).

7

Fig. 2 Relationships between ammonium lactate (AL) extractable P and concentration
of dissolved reactive phosphorus (DRP) and total phosphorus (TP) in leachate from
the three rainfall simulations in experiment 1. Each dot represents the mean of the
three rainfall simulations and four soil columns from the same P-level, cropping
system and soil. Error bars with standard deviation. Data transformed with the natural
logarithm.
Fig. 3 Relationship between ammonium lactate (AL)-extractable P in the soil (data

transformed with the natural logarithm) and the percentage of dissolved reactive P
(DRP) in total P (TP) in leachate. Error bars with standard deviation. See Tables 4-7
for more information about the relationships.

18

Fig. 4 Increase in concentration of dissolved reactive phosphorus (DRP) and total
phosphorus (TP) in leachate after manure application related to ammonium lactate
(AL)-extractable P. Each dot represents the mean of four soil columns from the same
P level, cropping system and soil. Error bars with standard deviation.

1 Tables

2 Table 1 Application rates of P to field plots, in relation to P removed with harvest

P level	P application rate (kg ha yr ⁻¹)				
	Fjärdingslöv,	Bjertorp, Klostergården,			
	Ekebo	Högåsa			
Low 0		0			
Medium	Replacement	Replacement			
High	Replacement + 15	Replacement + 20			
Very high Replacement + 30		Replacement + 30			

3 4

Table 2 Texture and concentration of ammonium lactate-extractable P (mg kg⁻¹) at different P levels infield plots. Results from composite topsoil samples (0-20 cm depth) taken in the autumn 2007

		Manured cropping system (MCS) Unmanured cropping system					em (UMCS)		
Texture		Low	Me- dium	High	Very high	Low	Me- dium	High	Very high
Bjertorp	Silty clay loam	19	24	70	101	18	28	87	123
Ekebo	Loam	36	70	164	236	24	46	109	173
Fjärdingslöv	Sandy loam	26	58	132	208	15	23	111	183
Högåsa	Loamy sand	29	42	80	133	18	26	84	116
Klostergården	Silty clay loam	23	29	100	121	34	47	117	152

Low, medium, high and very high refer to P levels with varied P application in relation to P removed

with harvested products, see Table 1. Börling et al. (2001) reported P-AL for the low P level with standard deviations $<1 \text{ mg kg}^{-1}$.

Table 3 Selected chemical and physical properties of soils used in the experiment. Degree of P saturation using ammonium lactate-extractable Fe, Al and P (DPS-AL) calculated according to Ulén (2006) on data from topsoil samples taken in 2010 in the high P level treatment in the unmanured cropping system (Anders E Lindsjö, personal communication)

	Texture ^a	PSC _{max} ^b	Sand	Silt	Clay	Porosity	pH^{a}	Organic C ^e	Са	PBC ^a	Fe _{ox} +Al _{ox} ^b	DPS-AL
		(mmol kg ⁻¹)	(%)	(%)	(%)	(%)		(%)	(cmol kg ⁻¹)	(I kg ⁻¹)	(mmol kg ⁻¹)	(%)
Bjertorp	Silty clay loam	8.8	nd	nd	30 ^a	nd	6.6	1.8	nd	4.7	111	22
Ekebo	Loam	10.2	47 ^d	36 ^d	18 ^d	44 ^d	6.5	2.2	7.9 ^d	5.3	108	13
Fjärdingslöv	Sandy loam	6.0	62 ^d	24 ^d	14 ^d	35 ^d	7.5	1.2	11.3 ^d	3.2	70	29
Högåsa	Loamy sand	10.0	77 ^c	15 ^c	7 ^c	46 ^c	5.8	1.8	3.8 ^c	4.7	116	18
Klostergården	Silty clay loam	6.9	9 ^c	44 ^c	48 ^c	45 ^c	6.9	1.8	21.3 ^c	3.9	89	33

Topsoil samples were taken from the unmanured cropping system (low P level).

 PSC_{max} : maximum P sorption capacity; Fe_{ox} + Al_{ox} : ammonium oxalate-extractable Al and Fe ; PBC: P buffering capacity ; nd: not determined. ^a Börling et al. (2001), ^b Börling et al. (2004), ^c Kirchmann et al. (2005), ^d Kirchmann et al. (1999), ^e Börjesson (2012), ^f Djodjic et al. (2004)

Table 4 Main effects and interactions (p-values) from the statistical analyses of P leaching, before recent manure application, at different concentrations of AL-extractable P. Dissolved reactive P, TP, percentage DRP in TP and leachate volume were used as dependent variables. Data on DRP, TP and AL-extractable P were transformed with the natural logarithm in the analysis

	Dependent variable (p-value)								
	DRP	ТР	% DRP in TP	Leachate volume					
CS	0.3	0.007	0.3	0.8					
AL-P	<0.0001	<0.0001	< 0.0001	0.6					
Soil	0.006	<0.0001	0.1	0.04					
AL-P * CS	0.9	0.2	0.05	0.9					
AL-P * Soil	<0.0001	< 0.0001	0.09	0.5					
Soil * CS	0.008	0.002	0.2	0.8					
AL-P * Soil * CS	0.07	0.03	0.1	0.8					

DRP: Dissolved Reactive Phosphorus; TP: Total Phosphorus; CS: Cropping System; AL-P:

Ammonium Lactate extractable P

Table 5 Estimates of least squares means and slopes with p-values, for P leaching before recent manure application. Leachate volume, DRP and TP were used as dependent variables in the statistical analysis. The LS means are mean values of leachate concentration or leachate volume, from three 25 mm rainfall simulations and from four soil columns. Values of DRP, TP and AL-extractable P were transformed with the natural logarithm, and the unit before transformation was mg L^{-1} for DRP and TP. In a, slopes with p-values for DRP and TP and LS means of leachate volumes for the different soils are shown. In b, LS means for DRP and TP of different soils and cropping systems are shown separately. Backtransformed geometric means are given within brackets. In c, comparisons between cropping systems for each soil are shown.

Soil	DRP -		TP -		Leachate volume (mm)			
a)								
	<u>Slope</u>	<u>p-value</u>	<u>Slope</u>	<u>p-value</u>	<u>LS Mean</u>	<u>SEM</u>		
Bjertorp	2.2 с	<0.0001	1.3 b	<0.0001	28 c	1.6		
Ekebo	1.2 a	<0.0001	0.58 a	<0.0001	17 a	1.6		
Fjärdingslöv	1.6 b	<0.0001	1.2 b	< 0.0001	19 a, b	1.5		
Högåsa	2.1 b, c	<0.0001	1.6 c	< 0.0001	27 с	1.5		
Klostergården	2.9 d	<0.0001	2.1 d	<0.0001	24 b, c	1.5		
b)								
	LS Mean	<u>SEM</u>	LS Mean	<u>SEM</u>				
Manured cropp	ing system (I)							
Bjertorp	-1.4 (0.25) c	0.20	-0.83 (0.44) d	0.11				
Ekebo	-4.0 (0.019) a	0.22	-3.0 (0.05) a	0.12				
Fjärdingslöv	-2.6 (0.072) b	0.19	-2.0 (0.14) b	0.10				
Högåsa	-2.5 (0.083) b	0.18	-1.5 (0.23) c	0.094				
Klostergården	-1.5 (0.22) c	0.19	-0.63 (0.53) d	0.097				
Unmanured cro	pping system (II)							
Bjertorp	-1.2 (0.30) c	0.19	-0.52 (0.59) c	0.098				
Ekebo	-3.4 (0.033) a	0.18	-2.6 (0.08) a	0.095				
Fjärdingslöv	-1.5 (0.22) c, b	0.18	-1.0 (0.35) b	0.095				
Högåsa	-1.7 (0.18) c, b	0.19	-1.1 (0.33) b	0.10				
Klostergården	-2.4 (0.093) b	0.19	-1.3 (0.28) b	0.099				
c)								
Comparisons of	LS Means betwee	en cropping	systems (p-valu	es shown)				
Bj I vs. Bj II	0.9		0.5					
Ek I vs. Ek II	0.6		0.3					
Fj I vs. Fj II	0.01		< 0.0001					
Hö I vs. Hö II	0.2		0.2					
Kl I vs. Kl II	0.09		0.004					
Values within c	olumns and section	ons with dif	ferent letters are	significant	ly different.			

Values within columns and sections with different letters are significantly different.

DRP: Dissolved Reactive Phosphorus; TP: Total Phosphorus; SEM: Standard Error of the Mean

Table 6 Main effects and interactions (p-values) from the statistical analyses of P leaching, after recent

 manure application, at different concentrations of AL-extractable P in five soils

	Dependent variable (p-value)									
	Increase DRP	Increase TP	% DRP in TP							
Soil	0.05	0.03	<0.0001							
AL-P	<0.0001	0.003	<0.0001							
AL-P*Soil	0.0003	0.009	0.2							

DRP: Dissolved Reactive Phosphorus; TP: Total Phosphorus; AL-P: Ammonium Lactate extractable P

Table 7 Estimates of least squares means and slopes for each soil from the statistical analysis of increase in P leaching after recent manure application. Increase in dissolved reactive P (DRP), increase in total P (TP), and percentage DRP in TP were used as dependent variables

	Increase DRP (mg L ⁻¹)					Increase TP (mg L ⁻¹)				% DRP in TP				
Soil	LS Mean	p-value	SEM	Slope	p-value	LS Mean	p-value	SEM	Slope	p-value	LS Mean	SEM	Slope	p-value
Bjertorp	1.7 d	<0.0001	0.1	0.17 c	<0.0001	2.0 c	<0.0001	0.2	0.17 b	0.0006	0.82 d	0.02	0.11	0.0004
Ekebo	0.0098 a	0.9	0.08	0.00054 a	0.9	0.2 a, b	0.2	0.1	0.0047 a	0.8	0.15 a	0.02	0.048	0.1
Fjärdingslöv	0.95 c	<0.0001	0.07	0.042 b	0.002	1.6 c	<0.0001	0.1	0.047 a	0.02	0.58 c	0.02	0.13	<0.0001
Högåsa	-0.042 a	0.6	0.07	-0.0076 a	0.68	-0.13 a	0.3	0.1	-0.026 a	0.4	0.40 b	0.02	0.15	0.0001
Klostergården	0.44 b	0.0002	0.08	0.040 a, b	0.04	0.54 b	0.002	0.1	0.030 a	0.3	0.58 c	0.02	0.12	0.0003

Values within columns with different letters are significantly different.

SEM: Standard Error of the Mean

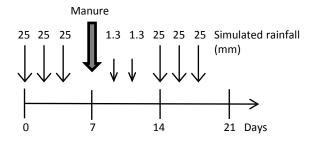


Figure 1.

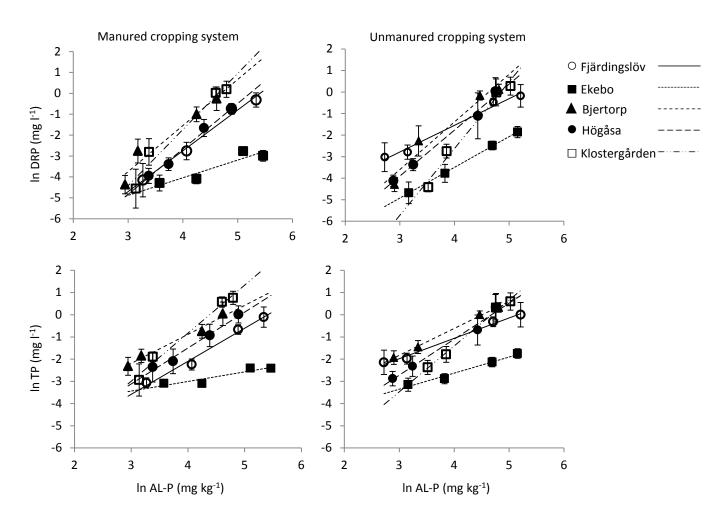


Figure 2.

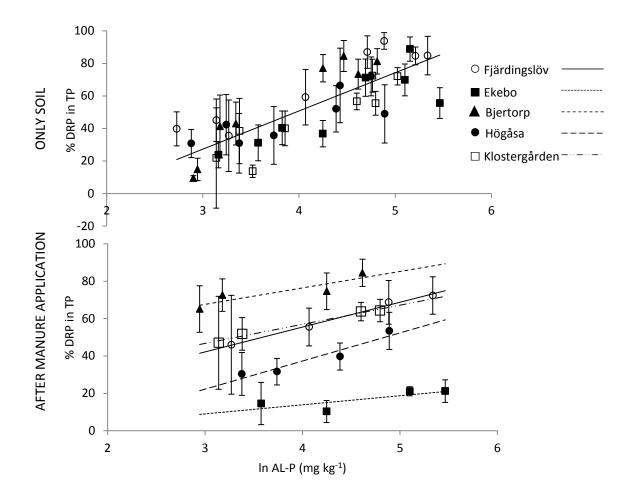


Figure 3.

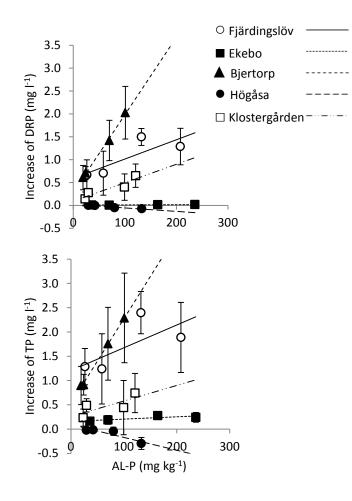


Figure 4.