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SUSTAINABLE PHOSPHORUS REMOVAL IN ONSITE WASTEWATER TREATMENT

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SUMMARY

Övergödning är ett utbrett miljöproblem (nationellt och internationellt) som i förlängningen kan orsaka skador i form av bland annat minskad biologisk mångfald, minskade rekreativvärden och försämrade livsmedels- och dricksvattensproduktionsmöjligheter. I Sverige är Östersjön en speciell angelägenhet och fosfor har pekats ut som det viktigaste bidraget till övergödning i stora delar av Östersjön. Av de olika bidragen av fosfor till Östersjön anses enskilda avlopp stå för drygt 10 %. Det relativt stora bidraget från enskilda avlopp har gjort att tuffare regelverk har införts som kräver mellan 70-90 % fosforavskiljning. De ökade kraven har inneburit att lämpligheten att använda konventionella markbaserade reningstekniker (infiltrationer och markbäddar) har ifrågasatts. Förbättrad fosforreningsteknik har utvecklats och krav på sådan teknik har blivit vanligare. Det har dock varit svårt att uppskatta den miljömässiga nyttan med att införa förbättrade fosforreningstekniker eftersom kunskapen om fosforfastläggning i konventionella markbaserade reningstekniker (både i absoluta termer och ur mekanistisk synvinkel) är begränsad. Dessutom har eventuella risker för negativa miljöeffekter (på grund av exempelvis ökad energi- och resursförbrukning) i samband med införande av förbättrad fosforreningsteknik aldrig utvärderats ordentligt.

I denna avhandling har huvudsyftet varit att förbättra beslutsunderlaget gällande miljömässigt hållbar hantering av enskilda avlopp genom att; (i) förbättra kunskapen kring fosforfastläggningsmekanismer i jord och reaktiva filter material, (ii) fastställa fosforfastläggningskapaciteten i markbaserade reningssystem, (iii) från ett miljömässigt helhetsperspektiv utvärdera prestandan hos markbaserade reningssystem i jämförelse med reningstekniker med förbättrad fosforavskiljning.

I reaktiva filtermaterial studerades fastläggningsmekanismer genom att undersöka i vilka kemiska former som den fastlagda fosfor förelåg med hjälp av X-ray Absorption Near Edge Structure (XANES) teknik (en typ av röntgenabsorptionsspektroskopi). I de flesta filtermaterialen som studerades (Filtra P, Polonite®, Wollastonit och Absol) dominerades fosforfraktionerna av kalciumfosfater. I två materialtyper (Filtralite® P och masugnsslagg (BFS)) uppträdde dock en betydande andel av fosfor som aluminiumfosfater eller som fosfor bunden till järn- eller aluminium(hydr)oxider. I markbaserade reningssystem studerades fastläggningsmekanismer, fastläggningskapacitet och rörlighet genom olika typer av skak- och kolonnförsök utförda på jordprover från äldre (10-30 år) anläggningar samt massbalansberäkningar. På grund av ett tydligt samband mellan oxalatlöslig fosfor och oxalatlöslig aluminium samt med stöd från kemisk jämviktsmodellering drogs slutsatsen att aluminium spelar en viktig roll för fosforfastläggningen i dessa system. Antingen genom att fosfor fälls ut som aluminiumfosfater eller att fosfor binds till aluminium(hydr)oxidator. Massbalanserna visade att mellan 320-820 g P m⁻³ hade ackumulerats i de markbaserade systemen vilket tyder på att fastläggningskapaciteten i den omättade zonen mellan infiltrationsyta och grundvattenyta/dräneringskikt i många fall överskrids. Den fastlagda fosfor visade sig också vara lätttröglig i vissa jordar.

Som metod för att utvärdera det miljömässiga hållbarhetsperspektivet användes livscykelanalys (LCA). En LCA modell utvecklades som kunde hantera skillnader i lokala förutsättningar och som kunde användas för att göra

utvärderingar ur såväl ett regionalt perspektiv (Östersjöregionen) som ett lokalt perspektiv. Från modelleringen erhöles resultat som tydde på att: (i) ur ett per capita perspektiv är emissionerna av övergödande ämnen samt förlusterna av fosforresurser som uppstår genom enskilda avlopp i regel påtagliga i jämförelse med emissionerna av växthusgaser och försurande gaser, (ii) reningssystem med förbättrad fosforavskiljning bidrar i högre grad till utsläpp av växthusgaser och försurande gaser än markbaserade system, (iii) ytvattenbelastande markbaserade system (markbäddar) bidrar i sådan grad till övergödning att det i många fall (även ur ett östersjöperspektiv) bör vara befogat att införa förbättrad fosforavskiljning, (iv) om man bortser från att fosfor inte kan återvinnas så visar sig markbaserade system med grundvattenutsläpp (infiltrationer) vara miljömässigt fördelaktiga i de fall där tillräckliga avstånd till ytvatten föreligger.

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LIST OF APPENDED PAPERS

This thesis is based on the following papers which are referred to in their corresponding Roman numerals and can be found in Appendix 1-5.

Papers included in the thesis:

- I. Weiss P, Eveborn D, Kärrman E, Gustafsson J.P. (2008) Environmental systems analysis of four on-site wastewater treatment options. *Resources Conservation and Recycling*, 52(10), 1153–1161.
- II. Eveborn D, Gustafsson J.P, Hesterberg D, Hillier S. (2009) XANES Speciation of P in Environmental Samples: An Assessment of Filter Media for on-Site Wastewater Treatment. *Environmental Science & Technology*; 43(17), 6515–6521.
- III. Eveborn D, Gustafsson J.P, Kong D. (2012) Wastewater treatment by soil infiltration: long-term phosphorus removal. *Journal of Contaminant Hydrology*, 140, 24-33.
- IV. Eveborn D, Gustafsson J.P, Elmefors E, Yu L, Lung E, Renman G. (2013) Phosphorus in soil treatment systems: accumulation and mobility. Manuscript.
- V. Eveborn D, Kiessling H.S, Kärrman E (2013). Environmental systems analysis on enhanced phosphorus removal in onsite wastewater treatment. Manuscript.

Relevant reports not included in the thesis:

Eveborn D, Holm C, Gustafsson J.P. (2009) *Fosfor I infiltrationsbäddar fastläggning, rörlighet och bedömningsgrunder* (Report 2009-07)(In Swedish). Swedish Water & Wastewater Association, Stockholm, Sweden.

Eveborn D, Gustafsson J.P, Elmefors E, Ljung E, Yu L, Renman G. (2012) *Kvantifiering av fosforläckage från markbaserade avlopps-system* (JTI skriftserie 2012:3) (In Swedish). JTI - Swedish Institute of Agricultural and Environmental Engineering, Uppsala, Sweden.

NOMENCLATURE AND ABBREVIATIONS

AP	Acidification potential
EP	Eutrophication potential
GHG	Greenhouse gas(es)
GWP	Global warming potential
GW discharge STS	Soil treatment system with groundwater discharge
(hydr)oxide	Sorbent occurring as oxide, hydroxide or oxyhydroxide
OWT	Onsite wastewater treatment
TPL	Technospherical phosphorus losses
STS	Soil treatment system(s)
SW discharge STS	Soil treatment system with surface water discharge

ABSTRACT

Aquatic eutrophication is a serious environmental problem that occurs all over the world. To protect surface waters (in particular the Baltic Sea), the regulatory pressure on onsite wastewater treatment (OWT) systems have increased in Sweden. Stringent requirements have led to uncertainties regarding the capability of conventional treatment techniques (soil treatment systems (STS)) to remove phosphorus (P), but they have also stimulated the development and introduction of enhanced P treatment techniques. In this thesis the accumulation and mobility of P as well as the chemical P removal mechanisms were studied in soils and reactive filter media. This knowledge was then used in environmental systems analysis. A model based on life cycle assessment (LCA) methodology was developed to evaluate the overall environmental performance of conventional and enhanced P treatment systems under various local conditions. The P accumulation in the studied STS varied ($320\text{-}870\text{ g m}^{-3}$) and the accumulated P was rather mobile in some soils. Phosphorus compounds were identified in alkaline reactive filter media (calcium phosphates predominated) by means of X-ray Absorption Near Edge Structure (XANES). In sandy soils from STS aluminium was found to be a key element for P removal, as evidenced by a strong relationship between oxalate-extractable P and Al. The LCA studies indicated that enhanced P treatment systems may be beneficial from an eutrophication and P recycling perspective but causes increased impacts in terms of global warming and acidification. Despite the drawbacks, enhanced P treatment techniques should be considered suitable substitutes to surface water discharge STS under most conditions. This is because the latter systems have such a strong eutrophication impact. On the other hand, under appropriate conditions, STS with groundwater discharge may be advantageous. These systems generally caused low environmental impacts except for the dispersion of P resources.

Keywords: Onsite wastewater treatment, Soil treatment system, Phosphorus, Removal mechanisms, Environmental impacts, Life cycle assessment

1. INTRODUCTION

Eutrophication of aquatic environments is a serious environmental problem for human kind that appears all over the world (Smith, 2003). Nutrient enrichment in water environments can in the end lead to decreased biodiversity, reduced recreational values and damages on essential resources such as food and drinking water supply. In Sweden the Baltic Sea is a particular concern because of the many ecosystem services that this water environment provides to the Swedish population. Many parts of the Baltic Sea environment may be growth-limited by phosphorus (P) (Boesch et al., 2006) and consequently the limitation of P loads to the Baltic Sea environment should be a priority

measure. The emissions of P to the Baltic Sea are due to several human activities including agriculture (45 %), municipal wastewater treatment plants (20 %), industries (17 %) and onsite wastewater treatment (OWT) systems (12 %) (Brandt et al., 2009).

To protect water environments in general and the Baltic Sea in particular, the regulatory pressure on OWT systems has increased considerably in Sweden. Current regulations require up to 90 % P removal (Swedish EPA, 2006). These stringent requirements have led to uncertainties regarding whether the capability of conventional treatment techniques (soil treatment systems (STS)) is sufficient. The

fact that several recent Swedish reports have been published on the subject reflects the uncertainties that exist regarding these systems (Ridderstolpe, 2009; Palm et al., 2012). Furthermore, as a consequence several enhanced P treatment techniques have been developed, and such techniques have become increasingly common.

The development and implementation of new P removal techniques may in general be beneficial from an eutrophication perspective. However, the risk for adverse environmental side effects from large-scale implementation of enhanced P treatment has not been seriously reviewed. Moreover, the actual benefits from an eutrophication perspective have been difficult to assess because of limited knowledge about the mechanisms and potential of P removal in conventional STS. Sustainable development needs holistic perspectives and such perspectives have to a large degree been missing in the current management of OWT systems in Sweden.

The overarching aim of this study was to improve the knowledge base for an environmentally sustainable management of OWT systems in Sweden. To fulfill this aim, the following research goals were established:

- To investigate and improve the knowledge about the mechanisms behind P removal in soils and reactive filter media (Paper II, III, IV)
- To clarify the P removal potential of conventional STS (Paper III, IV)
- To evaluate the overall environmental performance of STS in comparison to enhanced P treatment systems (Paper I, V)

Soils studied in the thesis are limited to sand fractions that meet Swedish design criteria for STS. The distinct P focus in the geochemical field means that other nutrients and pollutants are covered more briefly in sustainability evaluations. Another significant limitation is that the enhanced P treatment systems that are evaluated belong to a category which can be easily integrated within current sewer infrastructure. This means that source separating treatment

systems are not covered in the thesis. Sustainability evaluations have been directed only towards environmental issues and have taken starting point in Swedish scenarios.

2. BACKGROUND

To obtain improved knowledge within this field of research a multidisciplinary approach is required. In particular, two diverse scientific disciplines are covered: environmental geochemistry and environmental systems analysis. In this chapter the theoretical basis for specific areas within these disciplines that are relevant for the thesis are discussed and a background to the research question is given.

2.1. Onsite wastewater treatment – Swedish perspectives

Onsite wastewater treatment systems refer to treatment facilities that serve individual homes or small groups of households. These systems are common in rural areas in Sweden (Ridderstolpe, 2009) as well as in many other countries.

2.1.1. Historical outlook and current status

The issues that arise from the disposal of urine and faeces from human settlements are from many perspectives related to the introduction of running water and waterborne toilets. The ways in which the awareness and the attention about OWT issues have changed throughout history were discussed by Evehorn (2010). Briefly, the main focus has shifted from disposal issues towards health issues and then on to eutrophication issues. In recent years there has been a growing interest for a more comprehensive sustainability perspective.

According to Ek et al., (2011) there are nearly 700 000 OWT systems in Sweden (here defined as systems designed for less than 200 persons). Nearly 450 000 of these systems are used on a daily basis (for permanent living) and about 130 000 systems consist only of a septic tank (and therefore they do clearly not fulfil legal requirements). Among the systems with further treatment, STS counts for 70 % and

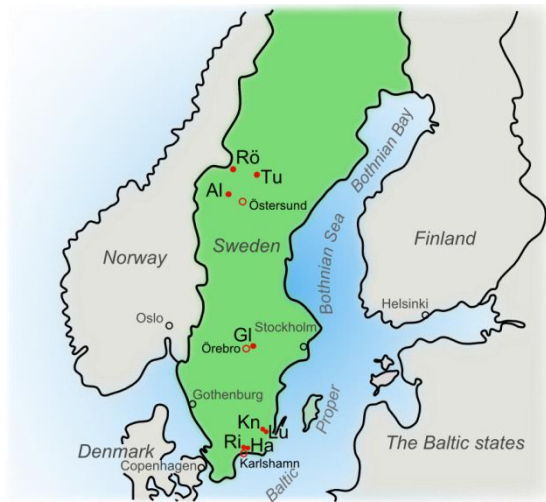


Figure 1. Map of the Baltic sea region with sample sites (soil treatment systems) indicated

most of the remaining systems are grey water systems with separate WC streams connected to collecting tanks.

2.1.2. Legal aspects

In Sweden the control and authorization of OWT systems are managed through the environmental and health authorities at the local municipalities and a legal permission is needed prior to construction. Since 2011 the Swedish Agency for Marine and Water Management (SwAM) has the general responsibility for OWT on a national level and there are legal guidelines available (Swedish EPA, 2006) that municipalities are expected to follow. The legal guidelines may be considered as a narrowed interpretation of the current legislation (mainly the environmental code; (Swedish Ministry of the Environment, 1998). In 2006 the guidelines replaced an older practical handbook ((1987) with the same legal meaning) and meant a conceptual change by moving from technical instructions towards more target-oriented rules. Among the targets, the stipulated treatment efficiencies are the most prominent ones. They imply that between 70 and 90 % P removal, 90 % BOD removal and up to 50 % N removal should be achieved. The removal requirements for N and P should be adjusted with respect to the sensitivity of the local environment. Even though one of the general goals of the Swedish environmental

legislation (Swedish Ministry of the Environment, 1998) is to further a sustainable development, the OWT guidelines (Swedish EPA, 2006) treat aspects as recovery and resource management very briefly.

2.1.3. The role of eutrophication

At the moment, eutrophication aspects are probably considered the most important issue associated with OWT in Sweden. This focus has been kept up for about two decades and ultimately it reflects the environmental concerns regarding the Baltic Sea (Fig. 1). The awareness of the serious environmental situation in the Baltic Sea was growing from the late 1960s and onwards. While some of the most acute toxicological issues in the Baltic Sea have been successfully mitigated, the eutrophication issues have been very difficult to tackle (Elmgren, 2001). The updated regulations regarding OWT systems (Swedish EPA, 2006) can in many respects be seen as a consequence of the objective to protect the Baltic Sea from eutrophication.

In a Baltic Sea eutrophication perspective P should in general be considered the most important nutrient to treat in OWT systems. This is not only because P is the limiting nutrient in many parts of the Baltic Sea environment (Boesch et al., 2006) but also a natural consequence of the relevance of P from OWT systems from a load apportionment perspective. The N leakage from OWT in relation to the total diffuse N leaching is minimal (Olshammar et al., 2009) while the P leakage from OWT systems is estimated to about 10 % of the total anthropogenic P load to the Baltic Sea from Swedish coasts (Brandt et al., 2009). If we compare data for the mean annual nutrient leakage from Swedish farmland (Johnsson et al., 2008) with data on the estimated nutrient stream from 2.5 inhabitants (in accordance with the method used in the Baltic Pollution Load Compilations; Ek et al. (2011)), it can be estimated that an average OWT stream is equal to the diffuse leaching from about 2 Ha of farmland (in the case of P). On the other hand, the same reasoning based on N will yield around 0.5 Ha of farmland per

OWT system. Thus, in contrast to the agricultural nutrient leaching, OWTs are concentrated point sources which should be considered easier to treat and to manage.

Even though the focus in Sweden many times is on the Baltic Sea, the relevance of P is at least as important in the local inland perspective. Recent studies in UK have observed that OWT systems can be critical for the eutrophication status of stream waters especially during low water flow (Withers et al., 2011). Because the ecological risk associated with algal growth in rivers is largely linked to soluble reactive P during low flow periods (Jarvie et al., 2006), OWT systems must be taken seriously even though their role in the annual nutrient budget may be limited.

2.1.4. Soil treatment technologies

As stated above, the most common onsite treatment technique used in Sweden is septic tanks with subsequent soil treatment. This is also a frequently applied technique in many other parts of the world. Australia, North America, Canada and parts of Europe use STS extensively in rural areas (Butler & Payne, 1995; USEPA, 2002; Beal et al., 2005; Ridderstolpe, 2009; Gill, 2011).

Soil treatment systems are a diverse group of treatment systems. However, they all share a common element: soil, which is fundamental for the removal processes responsible for the treatment of nutrients, organic substances, pathogens and other polluting elements present in the wastewater. The terminology concerning STS is far from consistent, and the praxis of OWT construction varies between regions and countries. Other widely used terms for soil treatment systems are e.g. “soil infiltration systems” (e.g. Zanini et al., 1998; Cheung & Venkitachalam, 2006), “soil absorption systems” (Postma et al., 1992; Beal et al., 2005), or simply “septic systems”. However, the term “septic system” is somewhat ambiguous since it may also refer to a system without any other treatment than a septic tank. Concerning soil treatment, two distinct different system designs should be noted: those with discharge to groundwater

(GW) and those with discharge to surface waters (SW). The latter ones (sometimes denoted as sand filters (Gill et al., 2009b; Wilson et al., 2011) are applied particularly in areas with soils that have a low infiltration capacity. Such systems count for about 30 % of the STS in Sweden (Ek et al., 2011). These systems use imported sand as a soil infiltration medium and they have a drainage system at the bottom that pipes the discharge to a surface water recipient. However, in real world applications, there are mixes of the above designs in which varying proportions of effluent discharges to groundwater. In Sweden, a negligible part of the drained systems is sealed, and sealing is not encouraged in the national guidelines (Swedish EPA, 2003; Ridderstolpe, 2009). In addition to the core STS designs described above, there is a growing number of prefabricated infiltration units that are built in as an integral part of many modern STS. Interview studies show that these systems have a significant market share in Sweden and constitute a significant part of the total number of STS (Kiessling, 2013).

2.1.5. Technologies for enhanced phosphorus removal

The interest and demand for more efficient P removal in OWT systems have resulted in a growing number of suppliers offering treatment solutions with high P removal capacity. Three different concepts for enhanced P removal can be identified:

- Source separation
- Chemical precipitation
- Reactive filters

In source-separated systems urine and/or faeces (which contains the bulk of the nutrients present in wastewater) is separated from other wastewater streams at the source. However, this thesis focuses on P removal technologies that are easily integrated within the current infrastructure. Both chemical precipitation and reactive filters satisfy this criterion. Chemical precipitation is a well-known technology that has been adopted from the municipal wastewater treatment engineering sector. In these systems precipitation agents such as aluminium or

iron chlorides are used to enforce the precipitation of aluminium or iron phosphates. These compounds accumulate in the organic sludge in the septic tank. As the sludge volume increases when chemical precipitation is used, there will be need for a larger septic tank or more frequent septic tank emptying. In most other respects the OWT system can be implemented according to conventional standards.

Reactive filter technology is a younger and less commercially developed technology. The main concept behind reactive filters is to make use of a material with strong P-sorbing properties that is porous enough to admit a long-term infiltration of wastewater. During this process P is trapped (chemically through precipitation or adsorption) in the filter material. This treatment solution can be managed very easily without the need for any advanced control and regulation technology. The robustness that is expected to follow with less need for process control in combination with a potential for affordable costs have been arguments for the development of reactive filter media for application in OWT systems (Baker et al., 1998). Another argument has been to close the P loop by searching for materials that will be possible to apply in agriculture and thus recover the P bound to them (Cucarella et al., 2008). From a literature review it appears that the first research publications on reactive filter media (natural sand and gravel excluded) were published in the late 1990s (Johansson, 1997; Mann, 1997; Zhu et al., 1997; Baker et al., 1998; Sakadevan & Bavor, 1998). In many cases these studies were linked to the development of constructed wetlands for wastewater treatment, which is still a common focus for research on reactive filter media. The latest reviews on the topic (Cucarella & Renman, 2009; Vohla et al., 2011) reveal the large number of natural and engineered products that have been studied for their potential as reactive P removal media.

The commercial products that are (or have been) available and promoted as reactive media on the Swedish market are Filtra P (Nordkalk Oyj Abp), Filtralite® P (Saint-

Gobain Weber) and Polonite® (Biotech AB). According to Rivera et al. (2012), Biotech (the leading provider) estimates there may be as many as 1 000 treatment units with these materials in Sweden. Hence, the real-world implementations are still few and it is unusual with more than 20 units within a single municipality (Rivera et al., 2012). On the Swedish market, the application manufacturers generally have chosen to implement the reactive filter technology in compact units (containing up to 1 m³ material), which requires frequent exchanges (the life span is a few years, at best). All the current commercial available reactive media are chemically driven by a combination of a high pH and a large release of Ca (Paper II), which in turn results in a negligible biological activity within the filter material. Consequently, biological pre-treatment is needed and the filter modules are sensitive to high organic loads (Nilsson, 2012). Therefore, in contrast to the chemical precipitation technique, reactive filter modules constitute the last treatment component in the OWT system and depending on the type of pre-treatment that is used, part of the P will be trapped earlier in the treatment process.

2.2. Assessing sustainability

An overarching aim of this project was to improve the knowledge base for an environmental sustainable management of OWT systems in Sweden. Three interrelated base components are usually used at conceptualization of sustainability: economy, environment and society. Here the environment (which is in focus in this thesis) may be seen as the most fundamental component because both society and economy depends on the environment while the opposite dependence is not equally strong (Giddings et al., 2002).

In a scientific context we need a framework to explore sustainability in a systematic and objective way. One framework that is focused on the environmental component is the Life Cycle Assessment (LCA) methodology (Bjorklund, 2002), which is a form of environmental systems analysis.

2.2.1. Environmental system analysis

In LCA, the environmental impacts throughout the whole life cycle (cradle to grave) of a product or a service are investigated (Rebitzer et al., 2004). To explore the whole lifecycle of a product, a large number of input data is needed. These data quantify flows of relevant substances, energy and resources. As for all kinds of modelling the quality of the input parameters will be critical for the quality and accuracy of the results (Huijbregts et al., 2001; von Bahr & Steen, 2004). This paradigm is important for this thesis and it ties together the work in two different scientific disciplines. In other words, refinements of input data through studies within the geochemistry field will improve the output of environmental sustainability modelling in the environmental systems analysis field (Fig. 2). The interaction between LCA and geochemistry studies have been strong, as uncertainties revealed in the input data (Paper I) led me to conduct studies in the geochemistry field (Paper II, III and IV). In the end all generated knowledge has been integrated (as far as possible) in a final environmental impact modelling approach (Paper V).

2.2.2. Aspects considered

It is not a straightforward task to define the key aspects, i.e. *impact categories* in LCA terminology, when to investigate the environmental sustainability of different wastewater treatment solutions. The starting point for this project has been the legislation, guidelines and objectives put forward in the Swedish society, and the emphasis has been on environmental quality rather than on human health. All environmental aspects analysed in the project are well established, and they are reflected or expressed in both legislation (Swedish Ministry of the Environment, 1998) and in the national environmental objectives of Sweden (Environmental Objectives Council, 2009). However, the choices made in this project have also been influenced by the specific relevance of the aspects within the OWT field as well as by the availability of data and tools. For

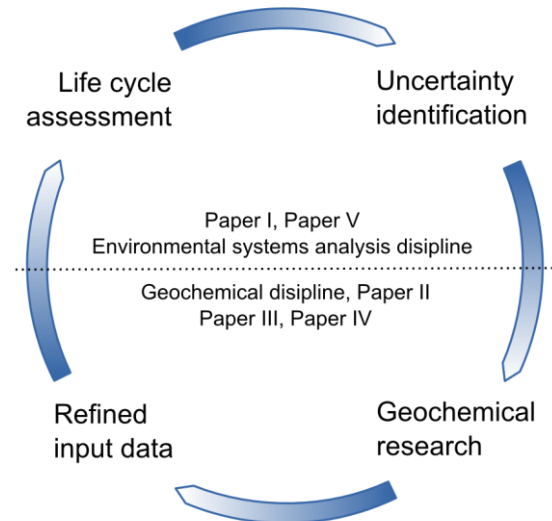


Figure 2. Illustration of the interaction between the geochemical and the environmental systems analysis research field.

example, human health aspects associated with insufficient treatment in OWTs would be relevant to consider but have been neglected much because of lack of both data and suitable methods.

2.2.3. Previous work

The number of environmental systems analyses targeting OWT systems is limited. One study that concerns small-scale treatment plants (<2000 persons) was recently conducted (Yildirim & Topkaya, 2012). For somewhat larger but still small applications there are more references (e.g. Tidåker et al., 2006; Gallego et al., 2008). Dixon et al., (2003) performed an LCA to compare reed beds with aerated biological filters. Moreover, a few studies with some similarities to the approach in Paper V have been conducted to develop a risk-oriented model framework for OWT systems in Australia (Carroll et al., 2006b) and to implement support for OWT systems in some hydrologic transport models in USA (Jeong et al., 2011). However, the most relevant analogue to the work conducted within this project and that used a holistic environmental approach for OWT systems is probably the work of Tidåker et al. (2007b), who studied the environmental impact of some different source separation

techniques compared to chemical precipitation.

2.2.4. Methodological issues

The LCA technique has frequently been criticised because of several methodological issues that are difficult to manage in a consistent and correct way and which may lead to discrepancy between the outcomes of studies despite similar goals and study objects (Reap et al., 2008). From a scientific point of view this is a serious problem. Since there is no means of validating the outcome from a LCA model, the reliability of the results may be difficult to judge. The most obvious source of unreliability in LCA is perhaps the variation in input data quality (Bjorklund, 2002). However, what should probably be considered more important and difficult to handle is e.g. allocation when dealing with processes with multiple outputs, selection of system boundaries and awareness of all relevant processes (Reap et al., 2008). To simplify, the latter issues could be boiled down to “ask a silly question, and you get a silly answer”.

Within this thesis, two methodological difficulties deserve special attention: eutrophication impact modelling and the difficulties with expressing the value of nutrient recovery. Eutrophication impact modelling deserves attention since eutrophication probably is the most discussed issue related to OWT systems today (at least in Sweden). In this regard, a fundamental problem with LCA is that the traditional impact models are not capable to consider spatial differences in the sensitivity of the aquatic environment (such as limiting nutrients, eutrophication status and natural nutrient retention). Traditionally a simplistic site-independent approach is taken to assess the environmental interference in terms of a *potential impact* (Gallego et al., 2010). More fine-tuned approaches can be cumbersome to manage since the activities in an LCA are often heterogeneously distributed over a range of large and diverse regions.

Within eutrophication impact modelling, most impact models take their starting point in the elemental composition of an average

algae ($C_{106}H_{263}O_{110}N_{16}P$); this is referred to as the Redfield ratio (Huijbregts et al., 2001). The potential eutrophication impact is expressed as the potential contribution to biomass production or the potential contribution to oxygen depletion, usually expressed in terms of PO_4 equivalents (e.g. Lindfors, 1995; Guinée, 2002). In both cases the Redfield ratio is used to estimate the characterization factors for substances that contribute to eutrophication. In general (as a consequence of the concept of *potential impact*) both N and P are assumed to be limiting nutrients for algae growth, while other elements are not.

In wastewater treatment eutrophication is an important impact category and most of the nutrient emissions have a distinct receiver. Thus, it is reasonable to distinguish between N and P emissions so that the impacts on the recipient can be judged from the actual nutrient status perspective. Both Paper I and Paper V handle N and P separately in the impact assessment. In Paper V we also normalized the impacts of N and P separately to both local (Swedish) and regional (Baltic Sea) data sets.

The other methodological issue of specific interest for this study is the difficulties to express the value of nutrient recovery. The problem is primarily related to the shortcomings in the current LCA praxis for expressing the impacts of the depletion of non-renewable resources (e.g. metals, phosphorus or valuable fossil deposits). However, nitrogen recycling is not affected by this as nitrogen fertilizers are primarily manufactured by means of N fixation from the air. The nitrogen production is therefore limited by the access to energy rather than by the access to a limited N resource. By contrast, P fertilizer production is completely dependent on mining activities (depletion of irreplaceable infinite resources). The depletion of finite resources is supposed to be represented by the indicators available for abiotic resource consumption. However, from our own experience (Weiss, 2007; Paper I) the logic behind the present indicators fails to recognize the P consumption.

According to Steen (2006) there are, in principle, four concepts behind the indicators of abiotic resource depletion: “those based on energy or mass, those based on relation of use to deposits, those based on future consequences and those based on exergy consumption or entropy production”. Schneider et al. (2011) argue that the differences between most of the used indicators will have a very small effect on the outcome. However, a major drawback of most indicators is that the depletion potential is summed over a large set of substances that fulfil very different needs in the society and can't be replaced by each other. This fact has been recognized by others (Brentrup et al., 2002). Thus, in Paper I, in which we tried using the ADP (abiotic depletion potential) indicator (Guinée, 2002), the depletion of P resources became unimportant in relation to the use of fossil fuels. This behaviour should be considered a shortage. It may be possible to gain energy and plastics from other resources than from fossil fuels, but it is theoretically and practically impossible to grow plants without phosphorus.

The easiest way around the problem discussed above is to separately report the P recycling potential. Several LCA reports have done so (Tidåker et al., 2007a; Tidåker et al., 2007b) including the first one within this thesis (Paper I). A drawback with such an approach is that the P resource issue is difficult to illustrate in the same context as other impacts (the recycling potential is something positive). Therefore, in Paper V a new impact category was defined, *Technospherical Phosphorus Losses* (TPL). This impact category is simply a measure of the amount of P in a technical system that is lost/dispersed without posing a critical benefit in food production.

2.3. Relevant phosphorus chemistry

Generally, in soil as well as in reactive filter media, P mobility is controlled by two main mechanisms:

- Precipitation/dissolution reactions driven by the equilibrium state in the pore water

- Sorption/desorption to functional groups on the surfaces of soil or reactive media

In practice it can be difficult to distinguish between these two types of reactions but they are quite different from a theoretical viewpoint. Precipitation reactions are generally slower than sorption reactions but are not directly limited by the amount of available surface area of the sorbent. A critical factor for both these mechanisms is the pH value (Arai & Sparks, 2007; Devau et al., 2009). At low pH values (< 7) the adsorption of PO₄-P to surfaces of iron (Fe(III)) and aluminium (hydr)oxides is generally considered important (Goldberg & Sposito, 1984; Gustafsson et al., 2012). Precipitation of iron or aluminium phosphates may also occur under such circumstances especially at high P concentrations (Gustafsson et al., 2012). At medium high pH values, P adsorption onto carbonates is possible (So et al., 2011). Precipitation of calcium phosphates may also occur, particularly if the pH value is > 9 (Johansson & Gustafsson, 2000; Gustafsson et al., 2012).

2.3.1. Properties of wastewater

Phosphorus is present in wastewater mainly as inorganic phosphate. The concentration is variable as a consequence of our daily habits (e.g. dietary, use of chemicals such as detergents as well as the daily water consumption). Even extraneous water can be important for the overall concentration of the wastewater, especially in treatment systems with larger pipe networks. Based on Swedish data, Ek et al. (2011) estimated the typical P concentration in wastewater from OWTs in Sweden to around 10 mg/L (based on a water consumption of 170 L/d and the current ban of P-containing detergents). This is roughly a factor of 1000 higher than the typical P concentration in natural waters (Correll, 1999). Wastewater also has a high salt concentration and it contains substances that can interfere chemically with the phosphate ion. As an example, humic substances can both block soil functional groups (Guppy et al., 2005; von Wandruszka, 2006) and inhibit calcium

phosphate precipitation (Alvarez et al., 2004; Song et al., 2006). As a consequence, the P chemistry involved when a soil or filter media is exposed to wastewater may become different from the predominant P chemistry in natural environments.

2.3.2. Phosphorus removal in soil treatment systems

Soil treatment systems have a long tradition and have been extensively studied. Early scientific work was concerned primarily with health, hydraulically and technical aspects (see e.g. SOU, 1955). However, P contamination from STS was subject to a number of early studies in the 70s and 80s (Reneau & Pettry, 1976; Sawhney & Starr, 1977; Jones & Lee, 1979; Gilliom & Patmont, 1983; Whelan, 1986; Chen, 1988). Thus, P attenuation in STS was discussed already in the 1970s (at least in North America). Research that covers Swedish soils and conditions is rare but some scientific studies were conducted in the 1980s (e.g. Nilsson & Stuanes, 1987; Pell & Nyberg, 1989). Since the mid-1990s the scientific work on ground water contamination from STS have progressed, particularly thanks to Robertson and coauthors who monitored a number of Canadian septic ground water plumes in detail for many years (Robertson, 1995; Robertson et al., 1998; Zanini et al.,

1998; Robertson & Harman, 1999; Robertson, 2003; Zurawsky et al., 2004; Robertson, 2008; Robertson, 2012).

Phosphorus removal will be expected first and foremost in the unsaturated subsoil beneath the drainage field (Fig. 3), but further retention is expected also in the ground water system if the treatment system is not drained. Chemical rather than biological processes are thought to be important for the long term P removal in STS. However, the biology is known to impact several chemical properties such as pH and redox potential. Robertson (2003) suggested that sewage oxidation processes, such as nitrification of ammonium and degradation of organic matter, are responsible for the often observed pH decrease in the soil below septic drain fields. Such pH decreases has also been observed in our own studies (Paper III, Paper IV) and may have effects on the P chemistry by influencing sorption processes involving iron and aluminum and by impairing the conditions for formation of calcium phosphates and adsorption to carbonates. A limited number of studies have been devoted to monitoring P removal in the unsaturated subsoil. However, both high (Pell & Nyberg, 1989; Lowe & Siegrist, 2008; Gill et al., 2009a), variable (Nilsson &

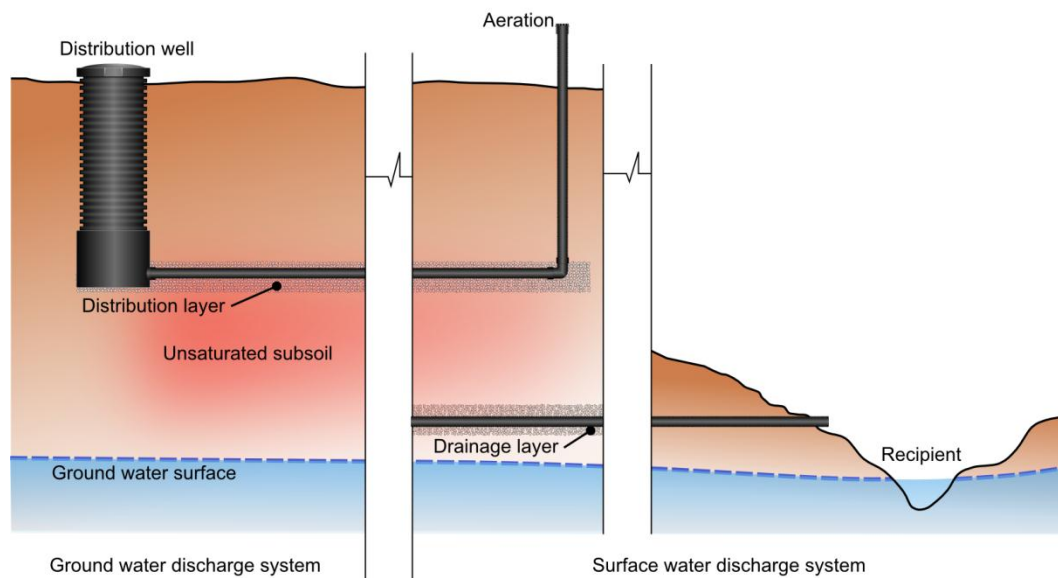


Figure 3. Description of typically designed soil treatment systems with surface and groundwater discharge

Stuanes, 1987; Carroll et al., 2006a) and low (Aaltonen & Andersson, 1996, Paper III) P removal capacities have been reported. An important limitation in most of these studies is that investigations have been conducted on young treatment systems and for a short period of time compared to the life span of a STS.

Whether the groundwater system should be seen as an integral part of the actual treatment system or not is a subjective matter. From a legal point of view it may be doubted whether this is the case (Swedish EPA, 2006), but the groundwater system can be a huge sink for P during favorable geochemical and hydrological circumstances. The reactive and adsorptive properties of P make it tempting to conclude that P would be rapidly immobilized in the groundwater system. Consequently, many early studies (e.g. Jones & Lee, 1979) as well as some recent reviews (Beal et al., 2005; CEEP, 2006) ruled out the risk for P contamination through groundwater transport. In fact, however, the mobility of P in groundwater has been shown to be variable. Whereas certain ground water systems effectively immobilize phosphate (Robertson, 2003) others (primary those on calcareous soils) do not (Robertson et al., 1998; Robertson, 2003; Robertson, 2008). Even in decommissioned systems continued migration of the phosphate plume has been observed on calcareous soils (Robertson & Harman, 1999).

There are few examples on direct identification of P minerals in STS. Whelan (1986) identified P precipitates rich in Fe and Ca but only in systems treating black water (toilet separated wastewater). In systems with more diluted wastewater no precipitates were found at all. However, from a closer inspection of the paper of Whelan (1986), it seems that the characterization was for the biomat rather than for the underlying soil. Zanini et al. (1998) and Robertson (2012) identified both Fe-rich and Al-rich P precipitates in soils beneath septic drainage pipes but no calcium phosphates (even though some samples were taken from calcareous soils). The role

of Al and Fe for the P removal is in agreement with the geochemical conditions in the subsoil (pH generally below 7). Indirect support for such processes have been presented in several studies (Robertson, 1995; Zurawsky et al., 2004) including our own work (Paper III, Paper IV). Through the set-up of a 1D reactive transport model that considered a wide range of chemical and biological processes Spiteri et al. (2007) was able to simulate the advance of phosphorus in two of the sites earlier monitored by Robertson. According to Spiteri and co-workers the model inferred that fast sorption reactions to oxides and carbonates were crucial for the P immobilization rather than precipitation reactions. However, their model did not include possible precipitation of aluminum phosphates (such as variscite). On the other hand evidence from certain sites indicate that precipitation may be the predominating process (Robertson, 2012).

2.3.3. Phosphorus removal in reactive filter media

It is difficult to be specific about chemical P removal mechanisms in reactive media since the materials can have completely different properties. However, if we narrow it down to the materials that are or have been commercially available in Sweden, these have many similarities in terms of chemistry including high pH and high calcium content. Filtralite® P differs somewhat from Polonite® and Filtra P, as it also contains considerable amounts of aluminium. There are several studies on the removal efficiency of these materials, both on the laboratory scale (Zhu et al., 1997; Hylander et al., 2006; Adam et al., 2007; Renman et al., 2009; Herrmann et al., 2012) and on the field scale (Heistad et al., 2006; Jenssen et al., 2010; Renman & Renman, 2010). As a consequence of the high pH and high calcium content in the materials, calcium phosphate precipitation is thought to be the predominant P removal mechanism. Some evidence supporting this statement has been presented earlier (Johansson & Gustafsson, 2000; Gustafsson et al., 2008), and additional evidence is provided in this thesis (Paper II).

3. MATERIAL AND METHODS

3.1. Sites, soils and reactive media (Paper II, III, IV)

A number of soils from different STS in Sweden as well as a selection of reactive media with different P loading history (Table 1) have been investigated by means of different analytical and experimental methods. At selection of soil samples we prioritized STS that complied with Swedish design guidelines and had an age of at least 10 years. Samples were taken at eight sites (Fig. 1): Alsen (Al), Halahult (Ha), Rötiviken (Rö), Tullingsås (Tu), Knivingaryd (Kn), Luvhult (Lu), Ringamåla (Ri) and Glanshammar (Gl). The soil samples were collected in the unsaturated subsoil beneath the distribution pipes at the depths 0-5, 5-15, 15-30, 30-60 and 60-100 cm. In addition a reference sample was collected from each site, which represented soil that had not been exposed to P-containing wastewater.

Six different types of reactive media were included in the study: Filtralite® P (FTE), Filtra P (FAP), Polonite® (PTE), Absol (AOL), blast furnace slag (BFS) and natural wollastonite (WTE). Each reactive filter medium was represented by 1-3 different samples which originate from various field applications and laboratory experiments (acquired from other research activities).

The unused soils and the reactive media differed particularly as regards the pH values and calcium contents (Table 1).

3.2. Phosphorus mineral characterization (Paper II)

Characterization of P in reactive filter media was performed through a combination of spectroscopic analysis methods (Paper II). Knowledge of the chemical speciation of P in reactive filter media is useful for optimizing and predicting the behaviour of practical applications during utilisation as well as during any subsequent recycling phase and can provide support for enhanced assumptions when the environmental sustainability should be evaluated.

Analyses by means of X-ray absorption near edge structure (XANES) spectroscopy,

X-ray powder diffraction (XRPD) and attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy were conducted. The two latter techniques were used more as complementary methods as they usually failed to identify any P species due to low P concentrations in the samples. However, they played an important role in verifying the purity of standards used for XANES data interpretation. The wavelengths of the beam used in XANES spectroscopy interact with the sample materials on an atomic level and generates element-specific absorption spectra. The spectra are derived from changes in the X-ray absorption coefficient around the element of interest (the absorber) and spectral features near the edge depend on the average local coordination environment of all absorber atoms in the sample (Kelly et al., 2008).

The XANES data for all reactive media listed in Table 1 were collected at beamline X-15B at the National Synchrotron Light Source, Brookhaven National Laboratory, NY. The beamline was operated in fluorescence mode and the fluorescence signal was measured using a solid state Ge fluorescence detector. The sample and X-ray flight path inside the sample compartment was purged with He gas. Data interpretation was carried out by a linear combination fitting (LCF) approach (Tannazi & Bunker, 2005), which was conducted by means of the Athena software (v0.8.056) (Ravel & Newville, 2005). Spectra for the following eight inorganic references compounds were used as standards: amorphous calcium phosphate (ACP), octacalcium phosphate (OCP), hydroxyapatite (HA), brushite (BTE), monetite (MTE), hydrated aluminium phosphate (AIP), and P adsorbed to aluminium oxide (boehmite) (Alox-P) or ferrihydrite (Feox-P).

3.3. Mass balance calculations (Paper III, IV)

To estimate the long-term treatment performance achieved in the unsaturated subsoil of the studied STS mass balance calculations has been performed.

Table 1. Description of soil and filter media investigated in this study

Name	Provider	Description	Major elements a (g kg ⁻¹ dw ⁻¹)				Ref pH _b	pH _c	Grain size (mm)	References
			Si	Al	Ca	Fe				
Filtralite® P (FTE)	Maxit Group, Norway	Expanded clay aggregates doped with limestone	-	20.3	30.5	5.8	10.7	7.5-9.6		d
Filtra P (FAP)	Nordkalk Oyj Abp, Finland	Granules of heated limestone, gypsum and iron	14.6	11.1	312	41.3	12.9	7.8-12.5	2-16	e, f
Polonite® (PTE)	Biotech Sweden AB,	Thermally treated and crushed calcium-silica bedrock	241	27	245	16.5	12.4	8.2-8.5	2-6	e, f
Absol (AOL)	Yxhult/Svesten Sweden AB,	Sorbent for oils, paint spills etc. (contains sand, crushed concrete and heated limestone)	232	10	194	8.2	9.5	9.3		-
Water cooled blast furnace slag (BFS)	SSAB Mercox Sweden AB,	By-product from the steel industry	155	69.7	216	3.11	9.4	8	0.5-4	e, f
Wollastonite (WTE)	Aros Mineral Sweden AB,	A calcium-silicate mineral (mining residues)	276	54.6	151	21.5	9.4	9.3	1-3	e, f
Alsen (Al)	Municipality of Krokomb, Sweden	Unsaturated subsoil from STS near Östersund	-	-	-	-	7.26	7.04-7.28	-	Paper III
Rötviken (Rö)	Municipality of Krokomb, Sweden	Unsaturated subsoil from STS near Östersund	-	-	-	-	7.52	7.28-7.38	-	Paper III
Tullingsås (Tu)	Municipality of Strömsund, Sweden	Unsaturated subsoil from STS near Östersund	371	73	10	44	6.45	4.74	d50=1.02	Paper III, IV
Halshult (Ha)	Municipality of Karlshamn, Sweden	Unsaturated subsoil from STS near Karlshamn	383	61	30	24	8.94	7.33-7.44	d50=3.93	Paper III, IV
Knivingaryd (Kn)	Municipality of Nybro, Sweden	Unsaturated subsoil from STS near Nybro	367	81	10	29	6.84	5.19-5.46	d50=1.45	Paper IV
Ringamåla (Ri)	Municipality of Karlshamn, Sweden	Unsaturated subsoil from STS near Karlshamn	395	55	17	23	8.89	5.87-6.06	d50=1.5	Paper IV
Glanshammar (Gl)	Private homeowner, Sweden	Unsaturated subsoil from STS near Örebro	421	58	10	34	5.89	3.87-4.95	d50=0.21	Paper IV
Luvehult (Lu)	Private homeowner, Sweden	Unsaturated subsoil from STS near Nybro	371	78	10	35	6.25	4.96-5.11	d50=1.64	Paper IV

^aTotal concentrations from element analysis, ^bpH values determined in reference samples (initial pH), ^cpH values in wastewater exposed samples or column effluents (for soils are pH values in samples down to 15 cm below the infiltration surface reported), ^d(Adam et al., 2007), ^e(Gustafsson et al., 2008), ^f(Gustafsson et al., 2011)

Accumulated P was determined in all soil samples from different soil treatment sites by calculating the difference between total P concentration in the subsoil sample and the total P concentration in corresponding reference sample. The calculated accumulations, dry bulk densities for each soil and volumetric data were then used to calculate the total amount of P removed in the subsoil (typically down to 1 m below the distribution level). The total amount of P was finally compared to an estimated P load on the system. Estimation of P loads was based on the number of persons connected to the system or if data were available, measurements of P mass flow. The total P accumulation at each site was typically based on the depth distribution of the P accumulation found at a single sample location (5 samples). However, at Tullingsås site a more detailed study were undertaken by conducting depth distribution sampling at five different locations (Paper III). In Paper III total P concentrations were analysed by means of nitric acid digestion and P determination with inductively coupled plasma emission spectroscopy (ICP-OES). In Paper IV borate fusion in combination with P determination using an inductively coupled plasma atomic emission spectroscopy (ICP-AES) and a quadrupole mass spectrometer (ICP-QMS) was used.

3.4. Soil analyses and batch experiments (Paper III, IV)

To investigate the chemical mechanisms behind the observed P accumulation in the different subsoils, samples and reference samples was chemically and physically characterized and a number of batch experiments were conducted. Oxalate-extractable Al, Fe and P were determined by means of the method described by Van Reeuwijk (1995).

For soil samples from the sites Lu, Kn, Ri, Ha, Rö and Al two types of batch experiments were conducted: desorption of P in response to acid and base additions and sorption of P in response to different P additions. In the desorption experiment equilibrations were carried out using 0.01 M

NaNO₃ as background electrolyte (to have a fixed and realistic ionic strength), different acid additions ranging from 0 to 2.6 mM HNO₃ or base additions ranging from 0 to 1.7 mM NaOH. Field moist samples (4 g) were suspended in 35 cm³ (Paper III) or 30 cm³ (Paper IV) solution and equilibrated for 5 days in an end-over-end shaker. After equilibration the samples were centrifuged and the pH value was determined on the unfiltered supernatant using a Radiometer combination electrode immediately after centrifugation.

The sorption experiments were set up in a similar manner. Phosphate additions (NaH₂PO₄) corresponding to concentrations between 0 and 0.5 mM (0-15 mg P L⁻¹) were used instead of acid or base additions. The liquid to solid ratio was 30:3 (Paper III) or 30:4 (Paper IV). Inorganic PO₄-P concentrations in equilibrated solutions were measured colorimetrically on filtered supernatants (0.2 µm Acrodisc® PF filter) by means of the acid molybdate method by use of flow injection analysis. All batch experiments were conducted at room temperature (21°C) and all samples were in duplicate.

3.5. Chemical speciation modelling (Paper III)

For some STS sites (Al, Ha, Rö and Tu) the conditions for precipitation of solid phases were evaluated to further explore which P removal mechanisms that might be important in the unsaturated subsoil. Activities of Ca²⁺, Al³⁺ and PO₄³⁻ were calculated from the batch experiments by means of the chemical equilibrium modelling software Visual MINTEQ (Gustafsson, 2009). The P phases included in the investigation are listed in Table 2. As input for the modelling the following parameters/concentrations were used: temperature, alkalinity, pH, Ca²⁺, Mg²⁺, Al³⁺, Fe²⁺, K⁺, Na⁺, PO₄³⁻, SO₄²⁻, Cl⁻, F⁻, and NO₃⁻. Concentration of anions and cations were determined by ion chromatography and inductively coupled plasma optical emission spectrometry (ICP-OES), respectively. However, the NO₃⁻

concentration was calculated based on the additions of NO_3 from NaNO_3 and HNO_3 and the inorganic $\text{PO}_4\text{-P}$ concentrations and pH were determined as described for the batch experiments. At $\text{pH} > 5$ (i.e., in all extracts except for those from the Tu 0–5 cm sample), the calculated Al^{3+} activity was higher than the one calculated from the solubility of “soil” $\text{Al}(\text{OH})_3(\text{s})$ (Gustafsson et al., 2001). The Al^{3+} activity was then modelled as fixed by the solubility of $\text{Al}(\text{OH})_3(\text{s})$, in agreement with procedures used for lake water (Sjöstedt et al., 2010). For the Tullingsås 0–5 cm sample, for which undersaturation with respect to $\text{Al}(\text{OH})_3(\text{s})$ was indicated, the Al^{3+} activity was instead calculated using the Stockholm Humic Model (Gustafsson et al., 2001), as described by Sjöstedt et al. (2010).

3.6. Column studies (Paper IV)

Large columns ($\Phi = 300$ mm) with wastewater-loaded soils from the sites Tu, Lu, Kn and Ri were set up to monitor the actual P discharge from the unsaturated subsoil of old STS. Sampled and homogenized layers from the four sites were added to the columns in the natural layer sequence with a distribution layer and drainage layer of macadam at the top and at the bottom (Fig. 4a). The depth of the soil columns were adjusted to 1 m. For the Ri column for which the depth of the real STS was less than 1 m, additional soil sampled from the bottom layer was added to reach a depth of 1 m. The column experiments were reproduced in duplicate and included two reference columns loaded with inert silica sand (SiO_2 content 99.8 %). Leachate was collected in polyethylene containers (40 L).

The column experiment was performed in two steps: first, 12 weeks loading with wastewater and subsequently 9 weeks loading with deionized water. The hydraulic loading rate (Fig. 4b) was adapted to the hydraulic loading rates given in the Swedish guidelines (Swedish EPA, 2003) and the columns were fed intermittently with 3 hours interval. The wastewater was taken from a 1 m^3 buffer tank (Fig. 4 b), which was completely refilled once a week with

fresh wastewater pre-treated mechanically by means of a drum screen (see Table 3 for results from characterizations).

After the end of the wastewater loading period, the buffer tank was cleaned and filled up with deionized water. The columns were then fed with deionized water for a period of 9 weeks. Loading rates and other conditions were identical to the preceding period. However, as a consequence of hydraulic failure (clogging), the reference columns had to be run using a lower loading rate.

In both experimental periods, effluent samples were taken once a week. The pH value was determined instantly after collection and an additional sample volume were frozen (-18 °C) and later analysed for total P. Total P concentration was determined through acid digestion followed by colorimetric analysis (the acid molybdate method).

3.7. Environmental system analysis (Paper I, V)

LCA methodology (Pennington et al., 2004; Rebitzer et al., 2004) was conducted to set up the environmental systems analysis models used in this thesis. The approaches and objectives in the two studies (Paper I and Paper V) were slightly different. System boundaries (Fig. 5) and impact categories varied somewhat as well as the exact types of studied treatment technologies. The results from other parts of the thesis (Paper II, III, IV) as well as a more complete literature review allowed for considerable improvements in the input data quality in the later model (Paper V). In particular a new approach to estimate the P removal performance of STS was used. In Paper I we simply assumed that GW discharge STS achieved 70 % P removal (as could be inferred from current legislation). In contrast we assumed in Paper V that the unsaturated subsoil had a limited P removal capacity equivalent to 350 g P m^{-3} (based on Paper III and IV). For groundwater discharge STS a simple first-order decay model was adopted to estimate

Table 2. Phosphate species included in speciation modeling

Reaction	$\log K_s$ (25°C) ^a	ΔH_r (kJ/mol) ^a
HAp: $\text{Ca}_5(\text{PO}_4)_3\text{OH}(\text{s}) + \text{H}^+ \rightleftharpoons 5\text{Ca}^{2+} + 3\text{PO}_4^{3-} + \text{H}_2\text{O}$	-44.3b	0
OCP: $\text{Ca}_4\text{H}(\text{PO}_4)_3(\text{s}) \rightleftharpoons 4\text{Ca}^{2+} + 3\text{PO}_4^{3-} + \text{H}^+$	-47.95c	-105 ^c
DCP: $\text{CaHPO}_4(\text{s}) \rightleftharpoons \text{Ca}^{2+} + \text{PO}_4^{3-} + \text{H}^+$	-19.28	31
DCPD: $\text{CaHPO}_4 \times 2\text{H}_2\text{O}(\text{s}) \rightleftharpoons \text{Ca}^{2+} + \text{PO}_4^{3-} + \text{H}^+ + 2\text{H}_2\text{O}$	-19.00	23
ACP2: $\text{Ca}_3(\text{PO}_4)_2(\text{s}) \rightleftharpoons 3\text{Ca}^{2+} + 2\text{PO}_4^{3-}$	-28.25c	-87 ^c
ACP1: $\text{Ca}_3(\text{PO}_4)_2(\text{s}) \rightleftharpoons 3\text{Ca}^{2+} + 2\text{PO}_4^{3-}$	-25.5c	-94 ^c
Variscite: $\text{AlPO}_4(\text{s}) \times 2\text{H}_2\text{O}(\text{s}) \rightleftharpoons \text{Al}^{3+} + \text{PO}_4^{3-} + 2\text{H}_2\text{O}$	-22.07d	-9.4 ^e

^a Unless otherwise stated, the values are from Smith et al. (2003).

^b Solubility of HAp at 21°C (McDowell et al., 1977).

^c Calculated from raw data given by Christoffersen et al. (1990).

^d Lindsay (1979).

^e Woods & Garrels (1987).

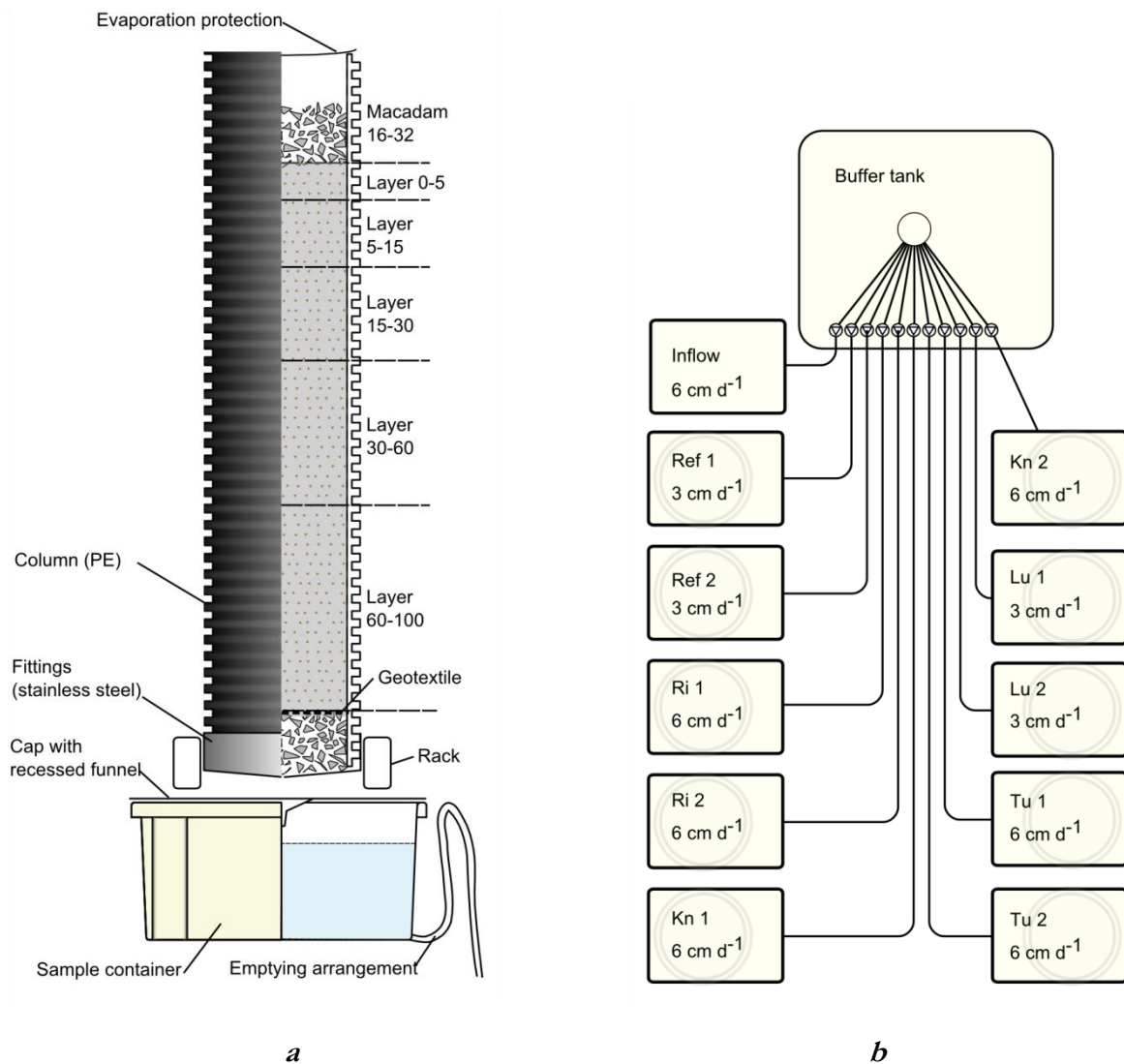


Figure 4. Layout of columns and sample containers (a) and schematic view over the column experiment including soils and loading rates (b).

Table 3. Characteristics of wastewater used in the column study and analytical methods utilized. All values in mg L^{-1} (except pH).

	Week 1	Week 5	Week 9	Analytical method
BOD7	120	140	160	ISO 5815:1989
CODCr	280	280	370	Kuvette test (Dr Lange LCK814)
SS	110	90	140	EN 872:2005
Tot-N	56.1	59.5	75.3	ex- SS 028131-1 oxidation with peroxodisulphate
NH ₄ -N	41.8	40.5	49.8	ISO 11732, flow analysis and spectrometric detection
NO ₂ -N	0.25	0.22	0.47	ISO 6777:1984, molecular absorption spectrometric method.
NO ₃ -N + NO ₂ -N	0.70	0.62	0.35	SS 028133-2 appendix A, reduction of nitrate with copperized cadmium followed by spectrophotometric detection
Fe	0.14	0.28	0.19	ISO 17294-2, ICP-MS
Al	0.62	0.60	0.72	ISO 17294-2, ICP-MS
Ca	81	89.4	132	ISO 17294-2, ICP-MS
Alk	537	517	569	ISO 9963-1:1994, acidimetric titration
TOC	85.4	61.3	84.6	SS-EN 1484 ed. 1. High temperature catalytic oxidation
pH	8.3	8.1	8.1	SS 028122-2

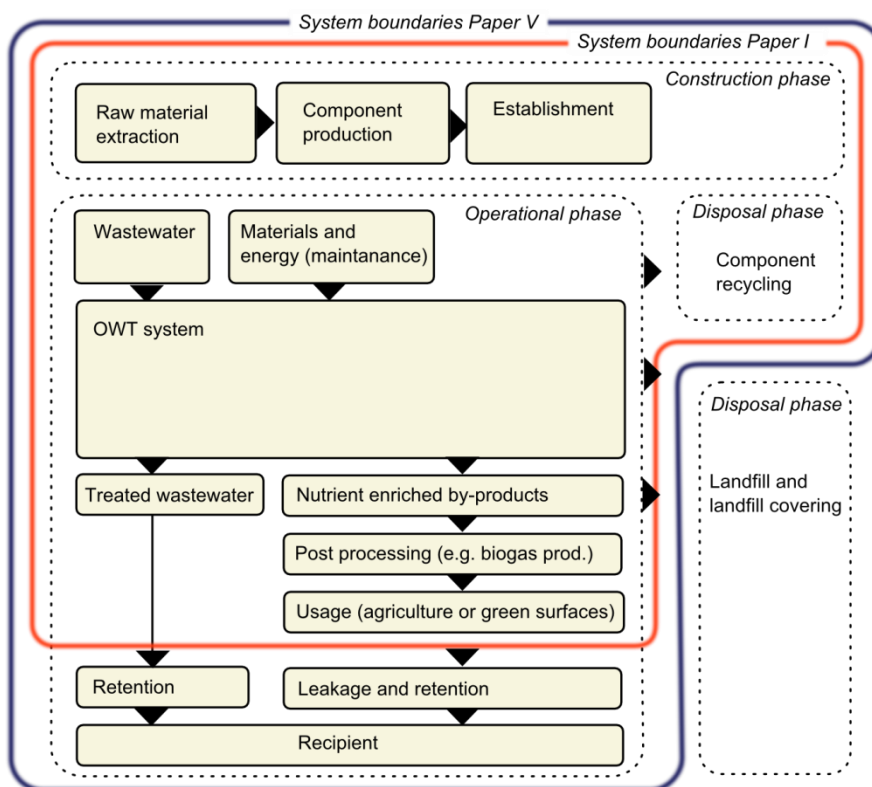


Figure 5. Comparison of system boundaries used in the LCA models in Paper I and Paper V respectively.

P retention in groundwater. The velocity of the P in the groundwater was assumed to be 1 m yr^{-1} and the half-life for P equal to 30 years. These conditions are similar to those described by Robertson (2008).

In Paper I the Stockholm County was chosen as a case study area, which means that most input data used were representative for this region. A consequential LCA was conducted to evaluate the environmental performance of groundwater discharge STS, two reactive filter systems with the filter media Filtralite® P and Filtra P and one chemical treatment system with polyaluminium chloride as the precipitation agent. In addition, a low-grade system was included, with only septic tank treatment. Impact categories studied included energy use, depletion of abiotic resources, global warming potential (GWP) and eutrophication potential (EP).

In Paper V the model was designed to allow for an evaluation of a more universal application (compared to the Stockholm County case study). Data applicable for Swedish conditions were used as input and the baseline scenario were compared to diverse type cases which considered e.g. hydrological and demographical differences in Sweden. The objective was to highlight

the dependence of local conditions and the importance of the eutrophication perspective (local or Baltic Sea-oriented). The systems evaluated were SW and GW discharge STS (conventional techniques), a reactive filter with Polonite® as the filter medium, and a chemical treatment system with polyaluminium chloride as the precipitation agent. The two enhanced P treatment systems were both combined with a compact biologic filter module. The impact categories studied were GWP, acidification potential (AP), technospherical phosphorus losses (TPL) and EP. The eutrophication potential were divided into subcategories for a P limited and N limited environment respectively (EP(P) and EP(N)). The TPL category is a self-defined measure of the amount of P that is lost/dispersed without posing a critical benefit in food production.

4. RESULTS

4.1. Wastewater impacts on soil/mineral characteristics

In most samples a significantly lowered pH was observed in the used materials compared to the references (Table 1). In Paper III and Paper IV we also observed larger amounts of oxalate-extractable metals in P loaded soils (Fig. 6).

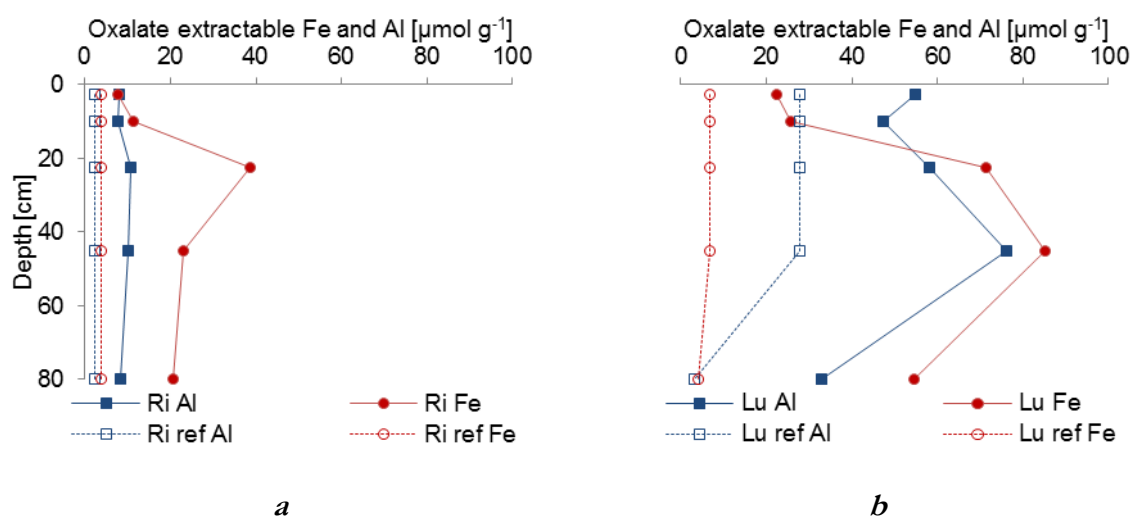


Figure 6. Depth distribution of oxalate-extractable Al and Fe in samples and reference samples at site Ri (a) and site Lu (b).

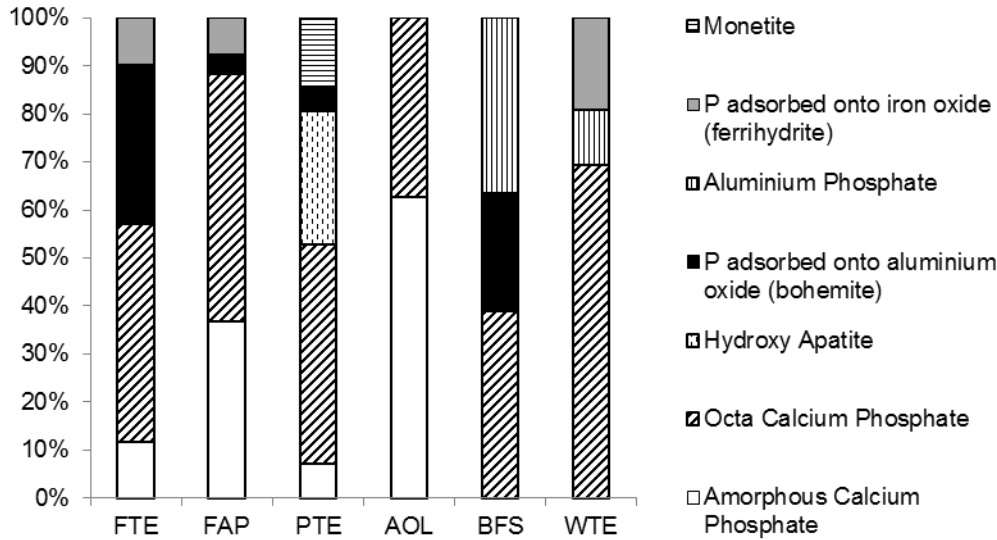


Figure 7. Distribution of phosphorus mineral phases in alkaline filter materials according to XANES analysis results

4.2. Phosphorus removal mechanisms

According to the XANES analysis (Fig. 7), all of the investigated reactive filter media had been enriched with calcium phosphates. The distribution of calcium phosphate mineral phases differed substantially among the studied materials (Fig. 7). In soil samples a strong relationship was observed between Al and P in the oxalate extracts (Fig. 8). However, the high ratio of oxalate-extractable P to Al in the filter beds (~ 0.7)

was larger than would be expected if sorption to Al compounds was the only important process. Evidence for a role of aluminium phosphates in the removal processes were obtained through chemical speciation modelling with data from the batch experiments. The calculated ion activity products for Variscite ($\text{AlPO}_4 \cdot 2\text{H}_2\text{O}$) were observed to be close to the saturation index in the Tullingsås samples (Fig. 9). In the desorption experiment saturation with respect to amorphous calcium phosphate (composition

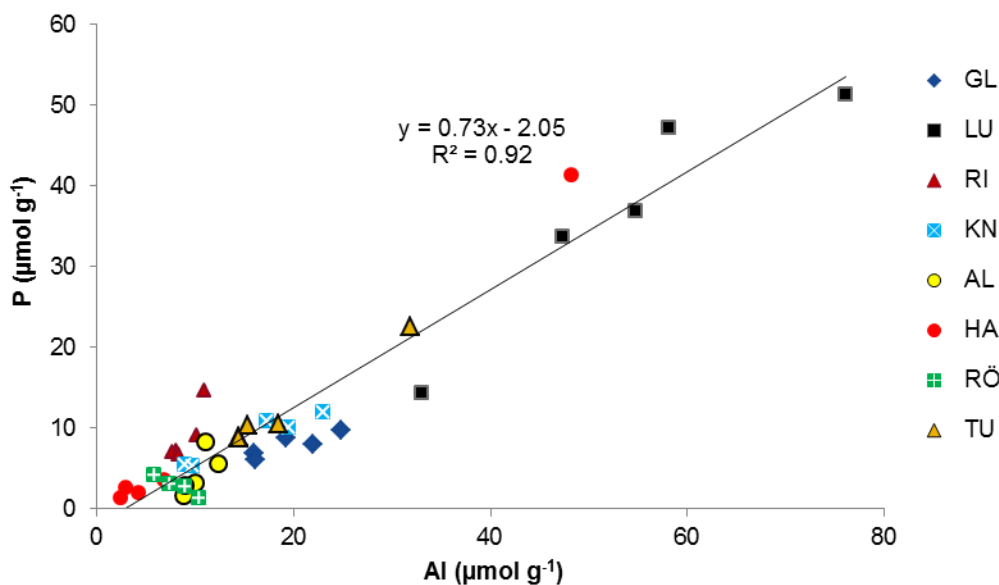


Figure 8. Oxalate-extractable P as function of oxalate-extractable Al. The data set includes wastewater loaded samples from all investigated sites.

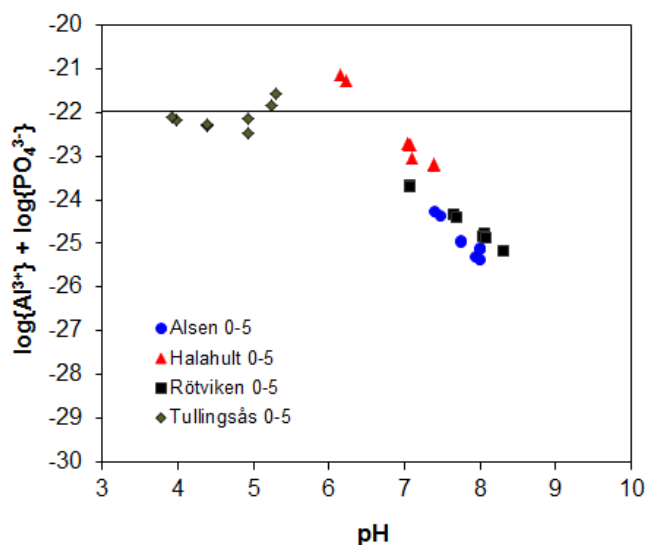


Figure 9. Modelled ion activity products and solubility line for variscite in equilibrated extracts from the pH-dependence experiment in Paper III, see Table 2 for constants used.

$\text{Ca}_3(\text{PO}_4)_2$ was only observed at the Halahult site. However, at high pH saturation could be observed for amorphous calcium phosphate at several sites in the batch experiment when phosphate had been added.

4.3. Phosphorus accumulation and mass balances

The amount of P accumulated in the soils varied considerably between different sites; the range was from 0.15 to 1.2 g P kg⁻¹, corresponding to 0.32-0.87 kg P m⁻³. No correlation between the observed P accumulation and the estimated P load could be observed (Fig. 10). High removal rates

(> 70 %) were observed in the column study when wastewater was applied. Even better P removal was obtained in the columns with soils from the Tu and Lu sites (> 95 % removal). However, the reference material (silica sand) showed very limited P removal (~20%). The detailed mass balance study at Tullingsås site indicated 12±4% P removal.

4.4. Phosphorus mobility

When the columns were loaded with deionized water, considerable amounts of P was mobilised and started to leach from the columns (Fig. 11). The leaching was greater in the columns with soil from the Ha and Ri sites than for the columns with soil from the Tu and Lu sites.

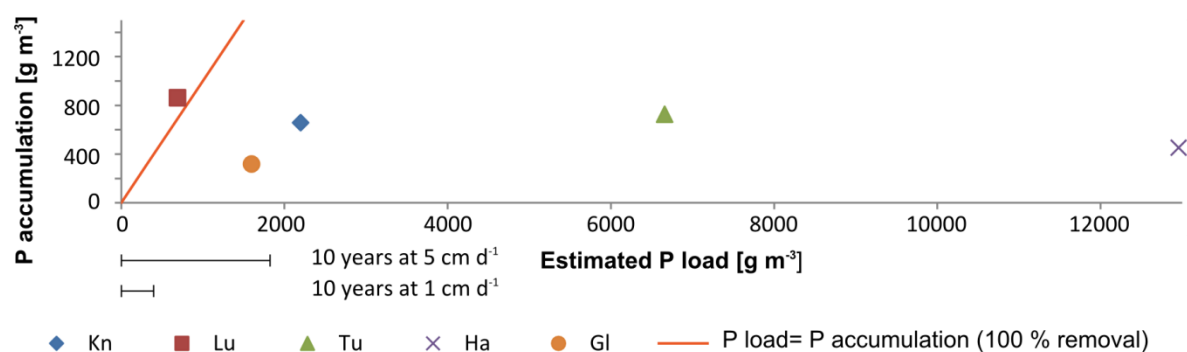


Figure 10. Phosphorus accumulation (y-axis) in comparison to estimated P load (x-axis) at the sites Kn, Lu, Tu, Ha and Gl. Red line indicate a theoretical maximal accumulation (100 % P removal). Distance indicators for x-axis express the time elapsed at hydraulic loads corresponding to 1 and 5 cm day⁻¹ and P concentrations corresponding to 10 mg L⁻¹.

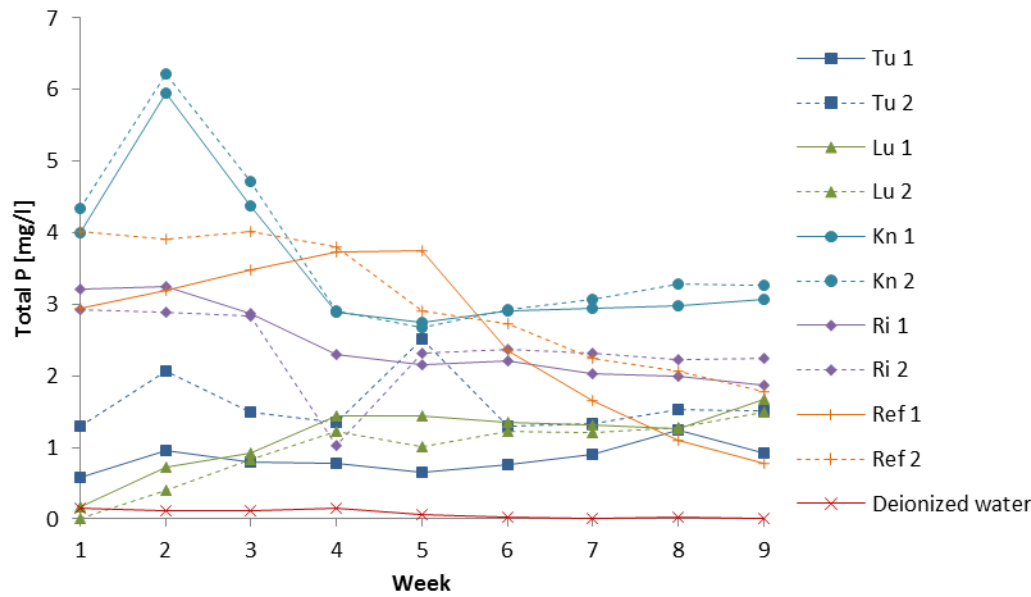


Figure 11. Total P concentrations in inflowing water and leachates from the Tu site column replicates (Tu 1, Tu 2), Lu sites column replicates (Lu 1, Lu 2), Kn site column replicates (Kn 1, Kn2), Ri sites column replicates (Ri 1, Ri 2) and silica reference replicates (Ref 1, Ref 2) during 9 weeks of deionized water load.

4.5. Environmental impacts

The differences between the outcomes of our two studies to evaluate the environmental performance of conventional OWT systems and enhanced P removal systems were significant in certain respects. In general, the eutrophication potential scored a significantly higher relative share in Paper V than in Paper I (compare Fig. 12a and Fig. 12b). In paper I no effect on abiotic depletion potential can be observed from the use of P fertilizer (compare e.g. *Filtra P*, a system that recovers P, with *Infiltration*, a system without P recovery in Fig. 12a). On the other hand the losses of P are clearly recognized in the TPL category introduced in Paper V and apparently the relative share is generally high for TPL in comparison to most other impact categories (Fig. 12b and Fig. 13).

Some patterns occur consistently throughout both studies. The emissions of GHG are in general higher for the enhanced P removal techniques (in particular the reactive filter systems) than for conventional systems (Fig. 12a, Fig. 13). The reverse is true for emissions of eutrophying substances (Fig. 12a, Fig. 13). From a Baltic Sea perspective (when natural retention is

considered) the relative share of the eutrophication impact was shown to be dramatically decreased (compare Fig. 12b and Fig. 13, note that the scale differs between the graphs). In Paper V, the P accumulation capacity of the soil as well as the half-life parameter in the groundwater retention model was observed to be crucial for the eutrophication impacts caused by the two STS (Table 4). The factors that had most effect on GHG were related to transports. The degree of usage was important to all systems in terms of GHG emissions. However, in contrast to the conventional STS, the enhanced P removal systems were not sensitive in terms of EP (Table 4).

5. DISCUSSION

5.1. Impacts of wastewater

In reactive filter media, a pH decrease after long term load of wastewater (Table 1) is expected as a consequence of chemical dissolution and consumption of reactive calcium oxide and other alkaline minerals such as Ca silicates. However, the pH changes in the natural soil samples (Table 1) are not as obvious and need a different explanation. Similar pH decreases in soils exposed to long term wastewater have been

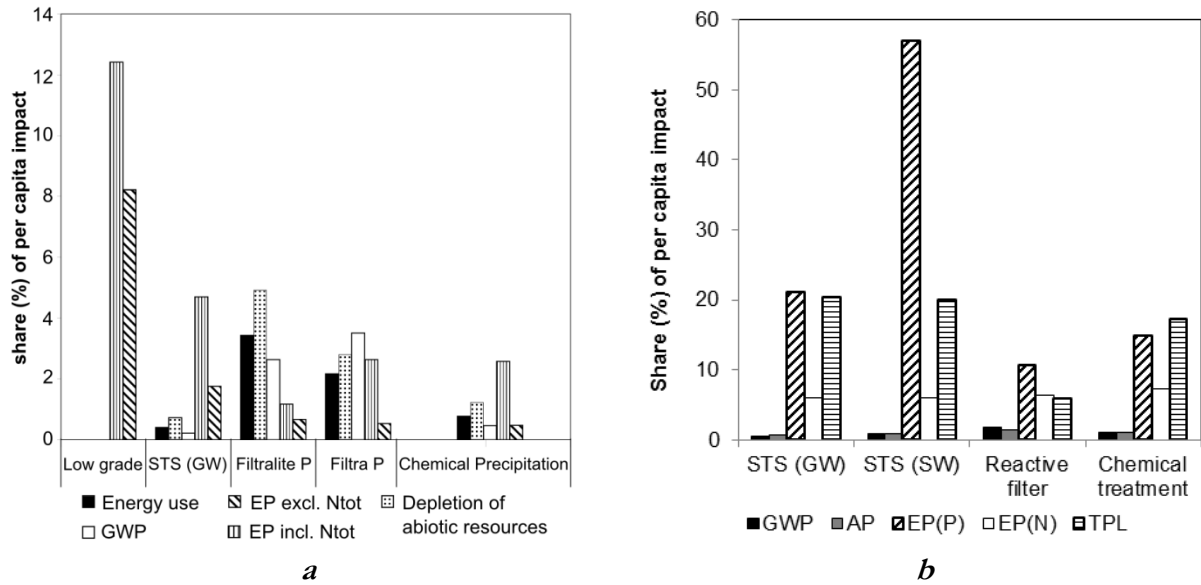


Figure 12. Relative shares of annually per capita contributions (in a set of impact categories) from different OWT systems acquired from LCA modeling in Paper I (a) and Paper V (b). The graphs show results from baseline scenarios.

reported by others (e.g. Robertson, 2003) and have been explained as an effect of acidifying organic degradation processes. The reason for the increased amounts of oxalate-extractable Al and Fe (Fig. 6) could be due to accelerated weathering of easily weatherable minerals brought about by the more acidic conditions. Lookman et al., (1994) suggested a P-induced weathering mechanism to explain P sorption on amorphous aluminium hydroxides at high P concentrations. However, the wastewater itself could also be a source of iron and aluminium, as suggested by Robertson (2012).

5.2. Phosphorus removal mechanisms and accumulation

The results from the XANES characterization indicate that calcium phosphate formation is an important P removal mechanism in all reactive media investigated (Fig. 7). From a chemical equilibrium perspective, it is reasonable that the high pH and Ca concentrations caused by dissolution of reactive forms of calcium in these materials will produce equilibrium conditions that induce calcium phosphate precipitation. Evidence for the presence of such conditions in experimental applications have been presented elsewhere through

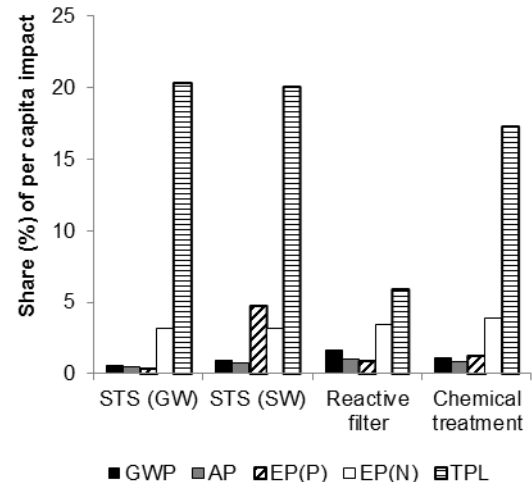


Figure 13. Relative shares of annually per capita contributions (in a set of impact categories) from different OWT systems acquired from LCA modeling in Paper V. The chart expresses results from a scenario with a Baltic sea perspective. It includes assumptions of high natural nutrient retention, low population density and a distance to surface water corresponding to 200 m. Eutrophication potentials are normalized to Baltic sea region per capita contributions.

chemical speciation modelling (e.g. Johansson & Gustafsson, 2000; Gustafsson et al., 2008). From the XANES results (Fig. 7), it is possible to further divide the calcium-bound P into different species. However, we observed rather small differences in spectral patterns between different calcium phosphate species at the P K-edge. Certainly a clear difference could be observed between non-crystalline phases (such as amorphous calcium phosphate) and crystalline phases (such as hydroxyapatite) but not among crystalline phases in general. Accordingly, detailed interpretations in this regard should be made with caution.

In Filtralite® P and blast furnace slag not only Ca phosphates but also aluminium phosphates and/or adsorption onto aluminium or iron (hydr)oxide surfaces seem to be important sinks for P (Fig. 7). Evidence for P associated with iron or aluminium in both Filtralite® P and metal slags have been presented earlier on basis of scanning electron microscopy (Pratt et al., 2007) or sequential extractions (Adam et al., 2006). Both these materials are rich in aluminium and many of the associated samples have been operated at a pH below 8 (Table 1).

The correlation between P and Al in the oxalate extracts (Fig. 8) indicates that AlPO_4 precipitation and or sorption to poorly ordered aluminium compounds are important removal mechanisms in many of the studied soils. The results from the chemical speciation modelling supported this hypothesis (Fig. 9). Since these removal mechanisms are favoured by low pH we can expect that the natural processes that decrease the pH in the soil and increase the amount of soluble aluminium will be beneficial for the removal of phosphate in the STS under most circumstances (acid to neutral soils). However, on the basis of the results from chemical speciation modelling on data from equilibrations from the batch experiments with P additions, we cannot exclude that also calcium phosphate formation may play a role under certain circumstances. However, such mechanisms should more likely appear in calcareous soils,

where the soil can buffer the pH to high levels, thus resisting the wastewater-induced acidification processes and at the same time be a source of calcium ions.

The P accumulation at the sites varied. The observations fell within an interval of 320–870 g P m⁻³ (as site averages). Theoretically the accumulation can be very low as almost no P removal was observed in our SiO₂ reference material (Paper IV). Moreover ongoing column studies at JTI have indicated negligible P removal rates in crushed bed rock material (Ljung et al., 2013).

It may appear surprising that that the P accumulation in reactive media may not necessarily be higher than in soils from STS. Renman & Renman (2010) reported P concentrations from full scale applications with Polonite® in the order of 600 g m⁻³. However, when comparing these figures to the results from our STS (320 to 870 g m⁻³), we need to remember that the hydraulic loads on the compact reactive filters are very high and that the material is considered as outdated when the P removal of the treatment system is lower than 90 %. Accumulation of P up to 7400 mg kg⁻¹ has been reported in identical materials from column studies with synthetic P solution (Renman & Renman, 2010).

The variations in P removal potential between soils should mainly be seen as a consequence of differences in inherent soil properties. The oxalate-extractable aluminium seems to be the most distinct indicator on the P removal capacity of the soils that we have investigated (Fig. 8), whereas physical parameters such as grain size distribution and specific surface area appears to be of minor importance (Elmefors, 2011). According to our accumulation studies (Fig. 10) there is no correlation between P load and P accumulation, which means that the traditional way of expressing P removal in STS by means of percentage estimates (e.g. USEPA, 2002; Ek et al., 2011) has fundamental shortcomings. Instead, the P accumulation capacity (g kg⁻¹ soil), the P load, and the amount of unsaturated subsoil involved in the treatment are essential for

correct estimates of the long term P removal. The crucial role of the P load and the P accumulation capacity for the performance of STS is illustrated by the observed difference between the P removal observed in Paper III and that of another recently published study (Robertson, 2012). The site in our study, with an age of 16 years, a P accumulation of up to 880 mg kg^{-1} , a hydraulic load of 16 cm d^{-1} and a subsoil depth of about 1 m, proved to achieve a removal rate of only $12 \pm 4 \%$. On the other hand Robertson's site with an age of 20 years, a P accumulation of up to 3340 mg kg^{-1} , a hydraulic load of 0.6 cm d^{-1} and a similar depth of the soil proved to achieve almost complete P removal.

The loading history of the soils in Paper IV (see also Fig. 10) suggests that we should expect poor P removal performance if they were subject to further wastewater application. However, the column studies did not deliver results that were consistent with these expectations. High removal rates (consistently above 70%) were achieved during 12 weeks wastewater load. These results are difficult to explain and in fact the columns performed better than reported for STS from several short-term field studies (Nilsson & Stuanes, 1987; Aaltonen & Andersson, 1996; Nilsson et al., 1998). One possible explanation to these results could be that the collected soil layers were mixed (separately) during the setup of column replicates. This homogenization procedure could expose new or hidden sorption or precipitation agents in the soil. Even though the effect of similar treatments have been reported to be very limited for other kinds of filter materials (Pratt et al., 2009) we assume that it can be a critical factor in this particular case where the hydraulic load was limited ($3\text{-}6 \text{ cm d}^{-1}$), the columns were deep (1 m) and the experimental period was short (12 weeks).

The possible mobility of accumulated P is an aspect of considerable importance for the long-term environmental impacts of STS. According to our column studies with deionized water, there is obviously a risk for leakage of P (Fig. 11). Soils with low

concentrations of oxalate-extractable aluminium (site Ha and Kn) which have poorer affinity for P also appear to have a greater risk of P leakage (up to 4 and 6 mg P/l was observed in the leachate from these sites). In contrast the P at the sites Lu and Tu proved to be less mobile (Fig. 11). However, even though the P leakage was substantially lower at the Lu and Tu site ($\sim 1 \text{ mg/L}$), the leakage is still substantial and relevant from an eutrophication perspective. The high leachate concentrations (up to 6 mg/l), which is higher than those obtained during wastewater load, are not trivial to explain. However, similar leachate concentrations (up to 9 mg/L) have been observed in a similar study (Zurawsky et al., 2004). Compared to those results, the P leakage from our sites is comparable to the leakage from some calcareous sites studied by Zurawsky et al. (2004) (leachate concentrations up to 4 and 9 mg P/L). An acid site studied by Zurawsky et al. (2004) showed almost no potential for P mobilization.

5.3. Environmental impacts

Several enhancements were implemented in the second LCA model (Paper V) compared to the earlier one (Paper I), and this resulted in dramatic differences in the relative share of eutrophication (compare Fig. 12a with Fig. 12b). Neither a change of system boundaries nor a refinement of the input data is responsible for this. Instead, the reason is mainly a change of data sources for normalization. In Paper I the eutrophication impacts were normalized to world averages (Guinée, 2002) whereas in Paper V regional averages for Sweden and the Baltic Sea region were used. It is noteworthy how important this change of the base for normalization is for the general conclusions in the two studies. In Paper I we found that the increased GHG emissions caused by some enhanced P removal techniques were critical to the results (Fig. 12a). This was not as obvious in Paper V, in which the relative impact of eutrophication became more important (Fig. 12b). However, we must also remember that environmental challenges such as global warming and eutrophication may not always be regarded as equivalent.

The introduction of the TPL concept in Paper V allowed for a more uniform representation of the environmental impacts. In Paper I this aspect was treated separately and could not be reviewed in the summary figures (compare Fig. 12a and Fig. 12b). Apparently, dispersion of P sources is significant (20 % of the relative per capita share) for conventional STS. However, only a limited fraction of the population in Sweden (~10%) uses OWT systems on a regular basis. Hence, because the flow of P within the sector is limited, the need of P recycling from OWT systems may be discussed.

From a local point of view, the eutrophication of STS with surface water discharge causes large eutrophication impacts, especially if considering that P is the limiting nutrient (Fig. 12b). In fact the eutrophication impact seems to be considerable compared to other impacts also from a Baltic Sea perspective when the nutrient retention is lower than 80 % (Fig. 13). Thus, enhanced P removal systems may be a better choice in many cases. However, if P losses are not considered important, our model indicates that it is undesirable to substitute GW discharge STS as long as the hydrogeological conditions are appropriate and the distance to surface water is sufficient. Such replacements would cause increased emissions of GHG and acidifying substances without any positive effects on EP (Fig. 12).

In Paper I we concluded that one of the most distinctive uncertainties in the LCA model was the estimation of nutrient removal in the STS. In lack of reliable long-term performance data we assumed a removal rate of 70 %, which reflected the requirements in the Swedish legislation (Swedish EPA, 2006). Considerable efforts have been made to address the P removal issue in this thesis (Paper III, Paper IV) and a new approach for the estimation of the P removal of STS has been implemented. However, despite these efforts P removal estimates are still uncertain. As shown in the sensitivity analysis shown in Table 4, the parameters introduced for the estimation of

P removal are crucial to the end result. Our baseline assumptions adheres to the precautionary principle, since the model is based on a P removal capacity of 350 g P m^{-3} in unsaturated soil (a poorly P-retaining soil according to Paper IV) and a half-life of 30 years during ground water flow, which is similar to an aquifer with poor P removal properties (Robertson, 2008). Moreover the groundwater flow is assumed to be directed straight towards a surface water recipient and no infiltration is assumed to occur in the SW discharge STS. This is probably not true in most cases as these systems rarely are constructed with impermeable liners (Ridderstolpe, 2009; Kiessling, 2013).

The precautionary principle approach taken in the new model may lead us to believe that eutrophication impacts of STS could not have been underestimated. However, the model has a limitation in time. What happens if the P discharge to the groundwater continues for an unknown number of generations of treatment systems? What happens with the temporarily bound P in the soil when a treatment system is decommissioned? These aspects are not handled in any of our studies and are very difficult to tackle. Problems with migrating P plumes have been reported in literature (e.g. Robertson & Harman, 1999; Parkhurst et al., 2003) and might become more common in the future since STS, from a groundwater perspective are a young phenomenon (groundwater velocities are generally small and P transport greatly retarded). Consequently, there should be reason to be cautious with the use of groundwater aquifers for wastewater treatment purposes, especially in areas with relatively high population densities and proximity to surface waters.

6. IMPLICATIONS FOR SOCIETY AND RESEARCH

This section discusses the result from this thesis from a legal, administrative and technical viewpoint. It also gives recommendations for future work in these fields.

Table 4. Baseline scenario results and outcomes (% change) of various parameter changes (sensitivity tests) performed in Paper V.

Sensitivity test	Impact category	Outcome			
		STS (GW)	STS (SW)	Reactive Filter	Chemical treatment
Baseline scenario	GWP (kg CO ₂ eq)	39	57	120	76
	EP(P) (g PO ₄ eq)	230	610	120	160
	TPL (kg P)	350	350	100	230
Increased system usage from 2.6 to 5 persons	GWP (%)	-47	-47	-48	-44
	EP(P) (%)	29	25		1
decreased biogas exchange from septic tank sludge (50 % of baseline value)	GWP (%)	5	3	2	6
Increased distances for delivery and recycling of system components (150 % of baseline value)	GWP (%)	6	11	1	1
Increased fuel consumption for septic tank emptying (130 % of baseline value)	GWP (%)	8	5	3	8
Decreased halftime in groundwater retention model from 30 to 15 years	EP(P) (%)	-55			
Increased P removal in unsaturated subsoil from 350 to 700 g m ⁻³	EP(P) (%)	-43	-53		
	TPL (%)			1	

6.1. Authorization

In the authorization process of OWT systems there is need for a uniform assessment of STS as well as other techniques. This might be promoted by providing tools or methods for estimation of environmental impacts.

6.1.1. Estimation of P removal in soil treatment systems

A simple strategy for assessment of the P discharge from STS would be to follow the basics used in Paper V. This approach can be broken into four parts:

- Estimate the P load on the system
- Determine the amount of soil available for P accumulation
- Estimate the P accumulation capacity in the subsoil
- Assess the potential for P retention in the groundwater zone

The P load on the STS will in general be critical for the predicted P discharge from SW discharge STS. According to Swedish regulations (Swedish EPA, 2006), the design hydraulic load should typically be based on 5 persons who spend all their time at home

independent of the circumstances of the individual case. In practise this design hydraulic load will usually overestimate the P-load to the system. Hence, a more realistic load estimate may be considered for use in P removal estimations. If permissions were given for a reasonable period of time it may be appropriate to base the P-load on the actual housing situation. That would be even more motivated if permissions were personal (instead of bound to property) alternatively associated with usage restrictions.

The amount of subsoil available for treatment varies. In GW discharge STS it is recommended to pay particular attention to get a sure indication of the highest seasonable groundwater table in order to get reliable estimations of subsoil volume. It is also noteworthy that the soil available in SW discharge STS may differ substantially depending on design. Systems with prefabricated infiltration modules may have significantly less soil volumes than traditional ones.

According to the P accumulation capacity of soils the advice should be to have a conservative view (in Paper V a

P accumulation capacity of 350 g m^{-3} soil was assumed). This approach is further motivated by the fact that accumulated P might be easily mobilized. According to our findings, oxalate-extractable Al may be a possible indicator for the P accumulation. However, to use oxalate-extractable Al in unused soils as a basis for assumptions is unreliable for several reasons. In particular because of the observed increase of oxalate-extractable Al in the soils as a result of wastewater loads. More over this type of information is generally not available in the authorization process and grain size distribution that usually are available does not seem to be correlated to oxalate-extractable metals (Elmfors, 2011). Thus, simple and precise estimates of the P accumulation of local soils are probably not possible.

The saturated ground water zone could without doubt be a huge sink for P. Hence, for STS with groundwater discharge (fully or partially) the P retention in ground water must be considered. In Paper V a simple exponential decay function with a half-life of 30 years was used to estimate the amount of P that could reach nearby surface waters. It has been verified that P plume development (Spiteri et al., 2007) can be fitted reasonably to such functions and there are examples where discussions about half-lives appears in scientific literature (e.g. Robertson, 2008). However, an exponential decay function will not describe the processes involved in P removal and predictions performed with this method should not be considered reliable.

6.1.2. Sustainable authorization

This thesis (Paper V) clearly shows that local conditions (e.g. hydrogeology) as well as the recipient perspective (a local or a regional) will influence on the predicted environmental impacts of different treatment options. Consequently, regulations and other policy instruments need to be smart and flexible designed in order to direct to the objective of sustainable development. In this regards, current legislation and policy instrument may need a review. The LCA model designed for the studies in the thesis could with some further development be

used as an interactive instrument in decision support at any level of decision-making. It would then be useful to expand the model to include other relevant types of treatment systems.

6.2. Enhanced treatment designs

No considerable efforts have been put to design or enhancements of treatment systems but some concluding remarks can be done.

6.2.1. Soil treatment systems

In Sweden the hydraulic load recommendations for STS are in the range of $3 \text{ to } 6 \text{ cm d}^{-1}$ (Swedish EPA, 2003). These recommendations are substantially higher than e.g. the American ($0.8\text{-}3 \text{ cm/d}$) and Norwegian ($0.6\text{-}5 \text{ cm/d}$) standards (USEPA, 2002; Jenssen et al., 2006). Accordingly, the P removal with the Swedish design may be poorer.

One reason for the high hydraulic loads promoted in Swedish guidelines is that coarse-textured soils are required for the authorization of STS. This is however a reasonable and safe approach since prevention of hydraulic failure should be a first priority in the design guidelines. In summary, a revision of the current guidelines for STS could be relevant but further investigations are then needed.

6.2.2. Phosphorus removal techniques

An important note regarding reactive filter systems is that the P recycling potential will be dependent on the choice of pre-treatment technique. Therefore, in situations when the ambition is to recycle P, reactive filter systems should always be combined with biological treatment units with minimal P removal. In combination with traditional STS, there is probably no need for any P removal units (either reactive filters or chemical treatment) until a few years after installation.

The high relative impact of eutrophication and P losses compared to most other environmental impacts (Paper V) indicate that even more radical measures (e.g. source separation techniques) to reduce nutrient leakage and to promote P recycling from

OWT systems may be justified from an environmental perspective. In my opinion an important development field for these solutions is technical design of innovative and appealing bathroom-close techniques that can create a demand for source separated techniques driven by the consumer market.

6.3. Sustainability research needs

This thesis is dealing with sustainability in a narrow environmental perspective and it could be argued that the results are not comprehensive even from a strict environmental viewpoint. Thus might an expanded approach be desirable which includes a broader selection of environmental aspects and incorporates evaluation of socio-economic aspects (e.g. an approach similar to the one described by Hellström et al. (2000)). Within the subject of OWT systems should microbiological risks, technical robustness as well as user and maintenance aspects be regarded important. Criteria and methods to evaluate these aspects need to be developed.

It is still many potential improvements that can be done even in the current model framework used in this thesis. For example more fine-tuned impact models are needed for eutrophication. This need can be motivated by the significant eutrophication impact that arises from OWT systems. It is particularly urgent for evaluation of local impacts. There exist many data resources from e.g. Swedish River Basin District Authorities that might be valuable for use in localized eutrophication impact models. However, there are no suitable methods available that can take advantage of these resources. Such models would give greater precision at evaluations of OWT systems in the local context.

6.4. Geochemical research needs

This thesis leaves several important geochemical questions regarding STS unanswered, question that needs an answer in order to take wise decisions about management of OWT systems. Some of the most important should be:

- How is the P discharge from the subsoil distributed over time?
- What P retention can we expect in the groundwater zone and which parameters are crucial?
- What are the risks associated with mobility of P in the subsoil as well as in the groundwater?

To answer these questions one would benefit from have a better mechanistic knowledge of P sorption and P release processes in the soil. To characterize P compounds in wastewater-loaded soils to get direct evidence for the forms of P that are accumulated would provide valuable knowledge. The use of XANES spectroscopy or similar techniques is recommended for such purposes. Another priority issue should be to clarify whether the limited P removal observed in calcareous soils (Robertson, 2003) is a direct result of the high pH value or rather the result of absence of oxyhydroxide surfaces. In general, calcareous soils are not common in Sweden but does this mean that our soils should be regarded as strong P sorbents? Our results indicate the opposite as the highest P accumulation observed among the investigated soils (an acid one) was 1200 mg kg^{-1} whereas concentrations up to 3340 mg kg^{-1} has been observed in acid soils in Canada (Robertson, 2012). However, further research is needed to clarify this.

The generally slow movement of P in groundwater makes modelling approaches valuable tools in evaluating long-term aspects regarding P mobility. As suggested by this research, biological acidification and weathering processes may impact the chemical speciation and should ideally be considered in such models. Moreover, hydraulics and reaction kinetics is critical components of process-oriented transport models. Examples on advanced modelling approaches exist (Parkhurst et al., 2003; Spiteri et al., 2007) and one attempt was recently performed within this project (Yu, 2012). To verify model accuracy as well as getting direct data for risk assessment there is also need for long-term monitoring of

discharge from STS and P movement in groundwater.

7. CONCLUSIONS

- Sorbed P in alkaline reactive filter media occurs primarily as calcium phosphates. In some materials (particularly in Filtralite® P and BFS) some P is also bound to iron and aluminium.
- In STS with sandy soils, precipitation of aluminium phosphates and/or adsorption onto aluminium (hydr)oxide surfaces are common and important mechanisms for P immobilisation.
- Significant differences are to be expected between the P accumulation capacities of different soils. The maximum capacity can easily be exceeded during the life-span of conventional STS.
- The P removed in STS is not irreversibly fixed but can be mobilized. The risk for mobilization differs between different soils.
- The eutrophication potential caused by SW discharge STS is generally high. Thus, substitution with enhanced P removal techniques may be justified in many cases even though the latter systems contribute to higher emissions of GHG and acidifying substances.
- If P recycling is not considered important, GW discharge STS may be appealing from an environmental point of view. Under suitable hydrogeological conditions and when the distance to surface water is sufficient they do not necessarily contribute more to eutrophication than enhanced P removal techniques. However, the long-term sustainability of using such systems may be questioned, as it is uncertain how P will behave in the groundwater zone in a long-term perspective.

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