



**KTH Land and Water
Resources Engineering**

BED FILTERS FOR PHOSPHORUS REMOVAL IN ON-SITE WASTEWATER TREATMENT:

Removal mechanisms and sustainability

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SAMMANFATTNING

De enskilda avloppen i Sverige beräknas bidra med ca 12 % av den totala antropogena fosforbelastningen på östersjön (från svensk sida). Som ett led i att minska näringsbelastningen och begränsa effekterna av övergödning i Östersjön så är det angeläget att minska belastningen från enskilda avlopp. Nya riktlinjer för rening av avloppsvatten från enskilda avlopp utgavs av Naturvårdsverket 2006. Dessa ställer betydligt högre krav på fosforrening än de tidigare. I många fall duger inte de konventionella reningsteknikerna (infiltrationer och markbäddar) för att uppfylla de nya kraven. Det finns dock flera nya avloppslösningar som utvecklats för att klara av höga krav på fosforrening, däribland reaktiva filterbäddar och filterbäddar med kompletterande kemisk fällning. Reaktiva filterbäddar bygger på fastläggning av fosfor i ett sorberande filtermaterial.

De nya kraven medför att man på många fastigheter kommer att tvingas uppgradera de befintliga avloppsanläggningarna. Det rör sig om en upprustning i stor skala. Vad som dock riskerar att åsidosättas är den miljöpåverkan och den resursförbrukning som kommer att uppstå i samband med att upprustningen genomförs.

Syftet med denna licentiatavhandling har varit att undersöka hur implementering av förbättrad fosforavskiljning från enskilda avlopp kan ske på ett så miljövänligt och uthålligt sätt som möjligt. Vidare har avsikten också varit att leverera kunskap för att bättre förstå fosforavskiljningen i olika typer av infiltrationsapplikationer (reaktiva filter såväl som infiltrationer och markbäddar). Avhandlingen undersöker vilken total miljöpåverkan man kan förvänta sig från några olika alternativa tekniker för fosforavskiljning i enskilda avlopp men inriktar sig också i hög grad på att förbättra underlaget för sådana uppskattningar genom att studera de kemiska mekanismerna bakom fosforavskiljning. I den första delstudien användes livscykelanalys för att jämföra miljöpåverkan från högpresterande fosforavskiljningstekniker (två olika reaktiva filterbäddslösningar samt en applikation med kompletterande kemisk fällning) med konventionell infiltrationsteknik. I den andra delstudien undersöktes uttjänt filtermaterial med hjälp av röntgenabsorptionsspektroskopi för att undersöka i vilken kemisk form som den fastlagda fosfor i materialen uppträdde. I den tredje studien gjordes en undersökning av fyra öppna infiltrationsanläggningar för att få klarhet i vilken fosforfastläggningskapacitet som på lång sikt uppnåtts och vilka kemiska mekanismer som är avgörande för avskiljningsförmågan. Slutsatserna från avhandlingen summeras i punktform nedan:

- Livscykelanalys är ett bra verktyg för att bedöma den totala miljöpåverkan som uppstår från olika typer av enskilda avloppssystem. Det finns dock behov av bättre underlag och metodutveckling för att göra en tillräckligt god bedömning av eutrofieringspotentialen från olika reningstekniker.
- En viktig uppgift för att minska den totala miljöpåverkan från reaktiva filtermaterial är att minska energibehovet per avskild mängd fosfor genom exempelvis effektivare tillverkningsprocesser, förbättrad avskiljningsförmåga eller helt nya typer av material.
- I de flesta reaktiva filtermaterial tycks utfällning av kalciumfosfatmineral vara den dominerande avskiljningsmekanismen. Fastlagd fosfor föreligger vanligen i form av kristallina kalciumfosfater (exempelvis oktakalciumfosfat eller hydroxyapatit), men även amorft kalciumfosfat påträffas i stora mängder i flera material. I vissa material återfinns en del av fosfor bunden till järn- och aluminiumfaser.
- Den långsiktiga avskiljningskapaciteten för fosfor i infiltrationssystem kan ha varit kraftigt överskattad i äldre svenska studier. Det krävs ytterligare forskning för att ta fram pålitliga bedömningar av fosforavskiljningskapaciteten i dessa system och sådan forskning måste även kunna förankra empiriska resultat vetenskapligt. På denna punkt brister i flera fall äldre undersökningar.

- I infiltrations- och markbäddar tycks och aluminiumföreningar vara viktigare för avskiljningen av fosfor än järn och kalcium.

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Finally, it's time for the classical family phrase (however, it would be a shame to avoid it!) To make it simple – all my nearest, you are such glorious creatures!

Uppsala 7/2 2010

David Eveborn

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LIST OF PAPERS

This thesis is based on the following papers which are referred to in their corresponding Roman numerals and can be found in Appendix 1-3.

Papers included in the thesis:

- I. Weiss, P., Eveborn, D., Karrman, 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. (2010) Wastewater treatment by soil infiltration – long-term phosphorus removal. Manuscript.

Relevant reports not included in the thesis:

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

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ABSTRACT

For many surface waters, phosphorus (P) leaching is a serious problem that should be minimized to prevent eutrophication. In Sweden there is a demand for physical and technical development of high-performance P removal techniques to reduce phosphorus leaching from on-site wastewater treatment systems to the Baltic Sea. However, although these systems are designed to reduce eutrophication there are also other environmental impacts to be considered when implementing them in on-site systems; energy use and global warming potential are two examples. This study has investigated several bed filter materials (reactive media and natural soils) for their total environmental impact (in commercial applications) as well as for the predominating chemical phosphorus removal mechanisms. The use of life cycle assessment revealed that several reactive bed filters are relatively energy-consuming due to the material manufacturing process. Characterization of phosphorus compounds in used reactive media provided evidence for calcium phosphate precipitation as the predominating P removal mechanism in alkaline filter materials. However, in soil treatment systems with noncalcareous soils, batch experiments and extractions suggested that aluminium compounds were important for P removal. According to mass balance calculations that compared accumulated P with the estimated P load in a soil treatment system, the long term P removal capacity was very low; only 6.4 % of the applied phosphorus had been removed during 16 years of operation.

Keywords: On-site wastewater treatment, Eutrophication, Environmental impact assessment, Phosphorus removal mechanisms, Soil infiltration, Reactive bed filters.

INTRODUCTION

Eutrophication of aquatic environments is a serious environmental issue in many parts of the world (Smith, 2003). In Sweden and nearby countries the Baltic Sea is a special concern. Reduction of the phosphorus (P) load to the Baltic Sea is particularly important since it is the limiting nutrient at several coastal regions (Boesch *et al.*, 2006). To fulfil the national environmental objective *Zero Eutrophication* (Environmental Objectives Council, 2009) there is need for measures in several sectors of the society. From on-site wastewater treatment systems (small scale systems with private owners), the contribution of P to the Baltic Sea has been estimated to be 12 % of the total anthropogenic P load from Sweden (Brandt *et al.*, 2009). This is similar in magnitude to the total discharges from municipal wastewater treatment plants (WWTPs). The corresponding percentage of nitrogen (N) that originates from on-site treatment systems is several times lower (2 %).

In 2006, the Swedish Environmental Protection Agency (SEPA) adopted new guidelines that put on-site wastewater treatment systems under pressure. Many

conventional treatment systems are not likely to meet the new stricter guidelines. Hence the guidelines should create an impetus for development of high-performance P removal techniques. However, the strong focus on eutrophication may overshadow other environmental impacts (such as climate change, energy use, etc.) that may occur when high-performance P removal techniques are implemented in a large scale. Compared to most high-performance P removal methods, the use of reactive bed filters (treatment by filtration through P-sorbing materials) represents a young and largely untested technique. One of its advantages is that it is easy to upgrade existing on-site systems to include reactive bed filters.

The aim of this project is to investigate how implementation of high-performance P removal techniques in on-site wastewater treatment can be made in an as sustainable way as possible. Furthermore it aims to increase the knowledge on the processes involved in phosphorus removal in bed filters. The study is therefore relevant for several Baltic countries in which P leaching

from on-site wastewater treatment systems are relevant to reduce.

A goal with this work is to develop a method for life cycle assessment that can be used to compare the relative environmental impact of different on-site wastewater treatment solutions. To achieve this goal and provide reliable input data for life cycle assessments much effort has to be put into understanding chemical P removal mechanisms. Bed filter systems (with reactive media as well as soil as the filter agent) are the applications in focus in this study.

TRENDS IN ON-SITE WASTEWATER TREATMENT: A SWEDISH PERSPECTIVE

On-site wastewater disposal as an environmental and health related problem has followed the evolutionary pattern of many other waste-associated issues. It began with the view that this was a local disposal problem, but with time and increasing knowledge this view evolved to something that concerned also larger regions and which therefore constituted a complex environmental impact chain.

The health aspect

In Sweden, the foundation for on-site wastewater treatment systems was laid in the middle of the 1940s (SOU, 1955). Health concerns had been raised due to the increased use of running water and water borne toilets in areas not supported by municipal wastewater treatment systems. As a response, the government involved a committee of wastewater experts to evaluate the situation in the country and to make suggestions for improvements. Even if the knowledge has evolved since then, the technical solutions highlighted in 1955 are still common and in fact many questions discussed in the SOU evaluation are still relevant.

In 1969 the first regulations with explicit relevance for private domestic wastewater treatment were published (SFS 1969:387). All permanent households were expected to have an infiltration unit after the septic tank.

However, the primary objective for treating on-site wastewater was still health protection although the knowledge on the environmental threats associated with wastewater had advanced considerably.

During the 1970s and 1980s a large amount of research and development was performed in Sweden and the focus shifted towards removal of nutrients and organic matter (see Nilsson, 1990; Pell, 1991). Swedish EPA published their first guidelines for on-site wastewater treatment in 1987 (AR 87:6); these were strongly focused on the proper design of soil treatment systems.

The eutrophication aspect

Eutrophication of aquatic environments can lead to ecological disturbances. Eutrophication may have negative effects on human resources such as food production and recreation values (Ronnbäck *et al.*, 2007) as well as biodiversity (Weijters *et al.*, 2009). However, the eutrophication issue is widely spread throughout the world (Smith, 2003).

In Sweden and nearby countries the Baltic Sea is a special concern. With the foundation of the Helsinki Commission (HELCOM) 1974, international cooperation concerning the protection of the Baltic Sea was initiated (Rheinheimer, 1998). The Baltic Sea has been subject to different types of pollutions (*e.g.* organochlorines and heavy metals) as well as high nutrient loads (Elmgren, 2001). A priority issue for HELCOM has been to reduce eutrophication. Moreover, Sweden has a national environmental objective *Zero Eutrophication* that additionally stresses the importance of measures to reduce the nutrient load on the Baltic Sea.

Among the eutrophying substances P is particularly important since it is the limiting nutrient in several parts of the Baltic Sea (Boesch *et al.*, 2006). Phosphorus originates from several anthropogenic sources such as industries, agriculture, forestry and wastewater (on-site and municipal treatment plants). The most recent compilation of pollution loads from Sweden estimates that on-site wastewater treatment systems accounts for 12 % of the anthropogenic P load to the Baltic Sea (Bothnian Bay,

Bothnian Proper and Baltic Proper) (Figure 1). This percentage, given by Brandt et al. (2009), is similar in magnitude to the amount of P discharged from WWTPs. Much work has already been done to reduce P from WWTPs in Sweden. However, for on-site systems there is still a huge potential for enhancements. On the other hand the nitrogen emissions from on-site wastewater treatment systems do not account for more than 2 % of the Swedish anthropogenic nitrogen load to the Baltic Sea. Thus P reduction has become a key issue for the management of on-site wastewater treatment systems.

The sustainability aspect

Since the Environmental Code was adopted in 1999, on-site wastewater treatment systems also need to deal with the concept of sustainability. One of the general objectives of the Environmental Code is to further a sustainable development (SFS 1998:808). Sustainability is a broad concept that was first defined in the Brundtland report (Rebitzer *et al.*, 2004; McConville, 2008). However, note that the environmental code refers to sustainability in an environmental perspective, leaving social and economical aspects at least partly outside its scope. This thesis does not claim to be able to quantify sustainability in absolute terms. Instead it is assumed that the degree of sustainability is related to the fulfilment of environmental objectives and guidelines set up by society.

Since sustainability is such a vague and broad concept it is not a straightforward task to apply it for on-site wastewater treatment in practice. However, it is clear that there is a need for several aspects to be introduced in assessments of sustainability, such as phosphorus recovery and resource- and energy savings. Phosphorus recovery is emphasized in the Swedish Environmental Objective *A good built environment* adopted by the Swedish parliament 1999 (Environmental Objectives Council, 2009). According to the objective, 60 % of the phosphorus compounds present in wastewater should be recovered for use on productive land by 2015. At least half of this amount should be

returned to arable land. Energy and resource conservation is also connected to *A good built environment* as well as for example the objective *Reduced Climate Change*.

Current regulation and practice in Sweden

Long ago the responsibility for the local management of Swedish on-site wastewater treatment systems was given to the health authorities, which gradually also got responsibilities for environmental issues. Today these authorities handle the authorization of on-site systems with the help of guidelines drawn up by SEPA. The latest SEPA guidelines from 2006 meant that a conceptual change was made by moving from a technical guideline (AR 87:6) to new and target-oriented guidelines (NFS 2006:7). The guidelines from SEPA can be seen as an interpretation of the current legislation (mainly the Environmental Code). On-site treatment systems today need to have 90 % COD removal, 70-90 % P removal and up to 50 % of N removal depending on the status of the local environment. This reflects the focus on eutrophication and the goals are often not likely to be met by conventional treatment systems (at least not in sensitive areas where the strongest reductions of P and N are required).

It is important to note that the current SEPA guidelines (NFS 2006:7) address other relevant sustainability aspects (such as nutrient recovery or resource management) only in a very general way. A contributing factor may be that it is difficult to formulate clear guidelines that take into account these aspects.

PHOSPHORUS REMOVAL TECHNIQUES

At many sites in Sweden conventional treatment systems will not meet the P removal requirements of SFS 2006:7; indeed they were not designed to meet any P-recovery goals. However, several new applications focused on enhanced P removal are available on the market. Commonly these applications rely on direct chemical precipitation (typically by aluminium

polymers) or filtering to reduce P. A different approach of reducing P is to separate urine or black water for further treatment locally or externally. However, this requires reconstruction of the existing sewage disposal systems.

Principles of reactive bed filters

P removal by chemical precipitation has been used for a long time in WWTPs. Thus, using it in on-site waste water treatment is only a new field of application for a well-known technique. On the other hand, the use of engineered filter materials for P removal purposes is a novel technique. The idea behind reactive bed filters is to capture P from wastewater in a sorbing filter material. An ideal reactive filter application should be as robust and simple to maintain as a conventional soil infiltration system and provide a straightforward way of recycling P to agriculture.

Limitations and advantages

It would be fair to say that a reactive bed filter at least potentially is a very robust technique. This is due to its simple design, to the minimum of automatic control equipment needed and to the fact that no dosage of chemicals is required. Despite the simplicity of the technique it can reduce P to outflow concentrations less than 1 mg/l (Heistad *et al.*, 2006; Renman & Renman, 2010). An ideal filter material will also capture P in a way that makes it available for plant uptake and at such a high concentration that it will be suitable as fertilizer. However, this target is not fully reached by the systems available on the market today. The fertilizer effectiveness of spent filter materials was demonstrated in greenhouse pot experiments (Hylander *et al.*, 2006). In the field however, it was more difficult to prove the fertilizer value of a certain filter medium, probably due to its low P concentration (Cucarella *et al.*, 2009). On the other hand, as most spent filter materials are alkaline, they can be used as liming agents with some fertilizer value. However, the experience of practical recycling of exhausted filter materials (in agriculture or for other purposes) is still very limited.

Phosphorus removal chemistry

Removal of P by infiltration can be enforced by several processes (including biological, chemical and physical mechanisms). Biological processes are probably not important for long-term P removal. Unless significant amounts of P can be removed through harvesting of plants, the biological P will be degraded and released when the load of wastewater is paused or finished. On the other hand chemical adsorption and precipitation mechanisms will depend mostly on the speciation in the equilibrium solution and could be of more interest over a longer time frame. Favouring P removal through chemical precipitation is all about providing a chemical environment that will keep the equilibrium speciation in an oversaturated state with respect to any phosphate mineral phase. Removing P through adsorption requires a reactive mineral surface with a large number of surface groups capable of forming surface complexes with P. However, P adsorption is still an equilibrium reaction. The equilibrium is heavily pH-dependent and also affected by the P concentration and by competition from other complex-forming substances (i.e. organic acids).

Many high-performing engineered filter materials are alkaline. They operate at high pH and are rich in soluble calcium (Cucarella & Renman, 2009). Under such conditions, several calcium phosphates are known to be stable and several studies indicate that calcium phosphate precipitation is an important removal mechanism in these materials (Drizo *et al.*, 2006; Gustafsson *et al.*, 2008; Bowden *et al.*, 2009). However, even precipitation and/or adsorption driven by Fe and Al phases can be present to some extent despite a high operational pH (Drizo *et al.*, 2006). Furthermore Rietra *et al.* (2001) showed that the presence of calcium will favour phosphate adsorption onto Fe oxides (goethite) particularly in alkaline environments.

In soil treatment systems the chemical conditions are different. The pH is typically between 4 and 8, and the calcium concentration is much lower than in the materials mentioned above. In the lower part

of the pH range P can be strongly adsorbed onto Al and Fe mineral surfaces (Arai & Sparks, 2007). These minerals are usually (hydr)oxides. Aluminium and iron could be important for removing P through chemical precipitation too: under acid conditions the solubility of some Fe and Al phosphate minerals is low and in particular variscite, $\text{AlPO}_4 \cdot 2\text{H}_2\text{O}(\text{s})$, could be an important phase that controls P solubility (Roncal-Herrero & Oelkers, 2008). Robertson (2003) found that soil treatment systems performed better on non calcareous soils than on calcareous ones which is reasonable according to what is stated above. However, the knowledge about the P removal processes actually taking place in soil treatment systems is very limited. Typically, studies have been carried out as “black-box” investigations using an inflow/outflow measurement approach (see *e.g.* Aaltonen & Andersson, 1996). Some work has also been done to follow the development of P-enriched groundwater plumes (Robertson *et al.*, 1998; Robertson & Harman, 1999). However, the net P removal seems to be dependent on processes in the unsaturated zone (Zanini *et al.*, 1998; Robertson, 2008).

TARGETING MULTIPLE OBJECTIVES

The P leaching from on-site waste water treatment must be reduced to prevent eutrophication. However, when implementing high-performance P removal techniques it is also important to prevent that this implementation induce any significant negative impacts with respect to other environmental issues. In other words, to favour sustainable development of on-site treatment systems there are several targets to consider simultaneously.

Principles of Life Cycle Assessment

To elucidate the total environmental impact of a particular activity or scenario several types of environmental system analysis (ESA) approaches are available. One frequently used method for ESA is life cycle assessment (LCA) (Ness *et al.*, 2007). The basic idea of an LCA is to consider a certain product or

service from a life cycle perspective (cradle to grave). The procedure of a life cycle assessment can be broken down into two phases: a life cycle inventory (LCI) and a life cycle impact assessment (LCIA). In the LCI phase, activities appearing during the life cycle are methodically studied and emissions and resource consumption induced by these activities are estimated (Rebitzer *et al.*, 2004). In the next step (the LCIA phase) the potential environmental impacts that the emissions and consumptions will give rise to are modelled (Pennington *et al.*, 2004). It is important to keep in mind that results from impact modelling show the potential but not the actual impacts arising from a particular study object (Baumann & Tillman, 2004).

By means of LCA it is possible to provide a more comprehensive view of a particular activity. Such a view is needed to evaluate on-site wastewater treatment systems with respect to the complex set of regulations and objectives set up by society.

There is a limited number of published scientific papers on the total environmental impacts of different options for on-site wastewater treatment (Brix, 1999; Dixon *et al.*, 2003; Tidåker *et al.*, 2007b). Furthermore the quality and scope of these studies are different. Most studies have been concerned with larger wastewater treatment systems (*e.g.* Lundin *et al.*, 2000; Tidåker *et al.*, 2006; Ortiz *et al.*, 2007; Tidåker *et al.*, 2007a). The reason is probably due to the differences of the potential impacts between these two groups of wastewater treatment systems. Moreover, WWTPs have a stronger principal.

Managing priority issues

Certain impact categories are of particular interest when evaluating on-site wastewater treatment systems. Eutrophication impacts and P recycling potential are two issues stressed in the Swedish environmental objectives. Indirectly, through the nutrient removal requirements in the SEPA guidelines (NFS 2006:7), eutrophication is also managed by legislation.

Modelling eutrophication impacts

Proper management of eutrophication requires not only high-quality emission data but also a well-aimed impact model. There are a number of methods available for modelling of potential eutrophication impacts using LCA. Typically, they have been developed for inclusion in larger frameworks for impact modelling such as EDIP2003 (Hauschild & Potting, 2005), IMPACT2002+ (Jolliet *et al.*, 2003), LIME (Itsubo & Inaba, 2003), TRACI (Bare, 2002) and LUCAS (Toffoletto *et al.*, 2007). Most of the present models take their starting point in the formula of an average alga ($C_{106}H_{263}O_{110}N_{16}P$), also termed the Redfield ratio (Huijbregts & Seppala, 2001). N and P emissions are assumed to contribute to eutrophication if the algae were limited by the nutrient in question. Thus the highest possible impact by the emissions can be estimated. Carbon, hydrogen and oxygen are not involved since these elements are never a limiting growing factor. The manuals stated above use this theory but have added methods to: consider the differences between emissions to different media (Huijbregts & Seppala, 2001), consider the concept of limiting nutrient (Jolliet *et al.*, 2003), increase the spatial resolution by compiling retention factors for different regions (Bare, 2002; Potting *et al.*, 2005; Toffoletto *et al.*, 2007) and consider the contribution of substances causing oxygen depletion (Kärman & Jönsson, 2001).

Managing P recycling

P recycling is normally not treated separately in an LCA. However, when applied to wastewater treatment systems it is clear that many LCA contributors give this issue special attention (*e.g.* Lundin *et al.*, 2000; Tidåker *et al.*, 2006; Tidåker *et al.*, 2007). In Sweden, the environmental objectives are clear about the goal of 60 % P recycling from the wastewater sector, which is probably a reason for the attention. However, treating the P recycling issue correctly is difficult. The P removal techniques for on-site wastewater treatment result in a range of different recycled products. The nutrient content as such does probably not reflect the potential of the product as fertilizer since the plant



Figure 1. Overview of the Baltic Sea area and the sites studied in Paper III.

availability may differ. Moreover, other properties of these products (such as physical structure and pH) will impact spreading practices as well as the suitability of the product for different soils.

MATERIALS AND METHODS

To provide a better understanding of how to implement enhanced P removal in on-site wastewater treatment systems, this project has used a variety of different methods and approaches. Initially a screening LCA was performed (Paper I). During this work a number of issues critical to reliability and usability of the results of the LCA as well as to development of reactive bed filters were identified. Two important aspects were then further highlighted and treated in Paper II and Paper III. This chapter gives a brief description of the various approaches and methods used. The different bed filter materials included in the project are also described.

Filter materials (Paper I, II and III)

As regards reactive bed filter materials, the intention has been to consider all filter materials currently available on the Swedish market. In addition some materials that so far only have been studied at a research level have been included. To enhance the knowledge and LCI data for soil infiltration, studies have also been performed on sand and gravel-like materials used in soil

Table 1. Description of filter media investigated in this study

Name	Provider	Description	Major elements (g/kg) ^a				pH ^b	Reference ^c
			Si	Al	Ca	Fe		
Filtralite P (FTE)	Maxit Group, Norway	Expanded clay aggregates doped with limestone during the thermal expansion process.	-	20.3	30.5	5.8	7.5-9.6	Adam et al. (2007)
Filtra P (FAP)	Nordkalk Oyj Abp, Finland	Manufactured granules made up of heated limestone, gypsum and iron.	15	11.1	312	41.3	7.8-12.5	Gustafsson et al. (2008)
Polonite (PTE)	Biotech AB, Sweden	Thermally treated and crushed calcium-silica bedrock	241	27	245	16.5	8.2-8.5	Gustafsson et al. (2008)
Absol (AOL)	Yxhult/Svesten AB, Sweden	Mixture of sand, crushed concrete and heated lime-stone. Marketed as sorbent for oils, paint spills etc.	232	10	194	8.2	9.3	^d
Water cooled blast furnace slag (BFS)	SSAB Merox AB, Sweden	By-product from the steel industry	155	69.7	216	3.11	8	Gustafsson et al. (2008)
Wollastonite (WTE)	Aros Mineral AB, Sweden	Residues from mining of wollastonite, a calcium-silicate mineral.	276	54.6	151	21.5	9.3	Gustafsson et al. (2008)
Soil (Al)	Municipality of Krokomb, Sweden	Collected from a soil treatment system in Alsen	-	0.26 ^e	-	1.46 ^e	7.1-7.6	Paper III
Soil (Rö)	Municipality of Krokomb, Sweden	Collected from a soil treatment system in Rötiviken	-	0.21 ^e	-	0.61 ^e	7.3-7.7	Paper III
Soil (Tu)	Municipality of Strömsund, Sweden	Collected from a soil treatment system in Tullingsås	-	0.49 ^e	-	1.29 ^e	5.0-6.9	Paper III
Soil (Ha)	Municipality of Karlshamn, Sweden	Collected from soil treatment system in Halahult	-	0.30 ^e	-	0.31 ^e	6.6-8.2	Paper III

^aTotal concentrations if nothing else stated.

^b Range of pH values in studied samples, measured as the pH in the equilibrium solution at the end of the P-loading procedure. In the case of the Tullingsås site, values are from location 5.

^c Reference for detailed material description

^d Renman *et al.*, manuscript in preparation

^e Extracted by oxalate buffer at pH 3.

treatment systems. These samples consisted of soil collected from four well-documented wastewater infiltration plants. One site was located near Karlshamn (southern Sweden) and the other three near Östersund (northern Sweden) (Figure 1). Table 1 summarizes the complete set of materials studied within the project.

Life cycle assessment (Paper I)

As a tool for comparison of the use of natural resources and of the relative environmental impact of different on-site treatment systems, LCA was chosen. In paper I, the county of Stockholm was chosen as a case study area to evaluate suitable options for on-site wastewater treatment.

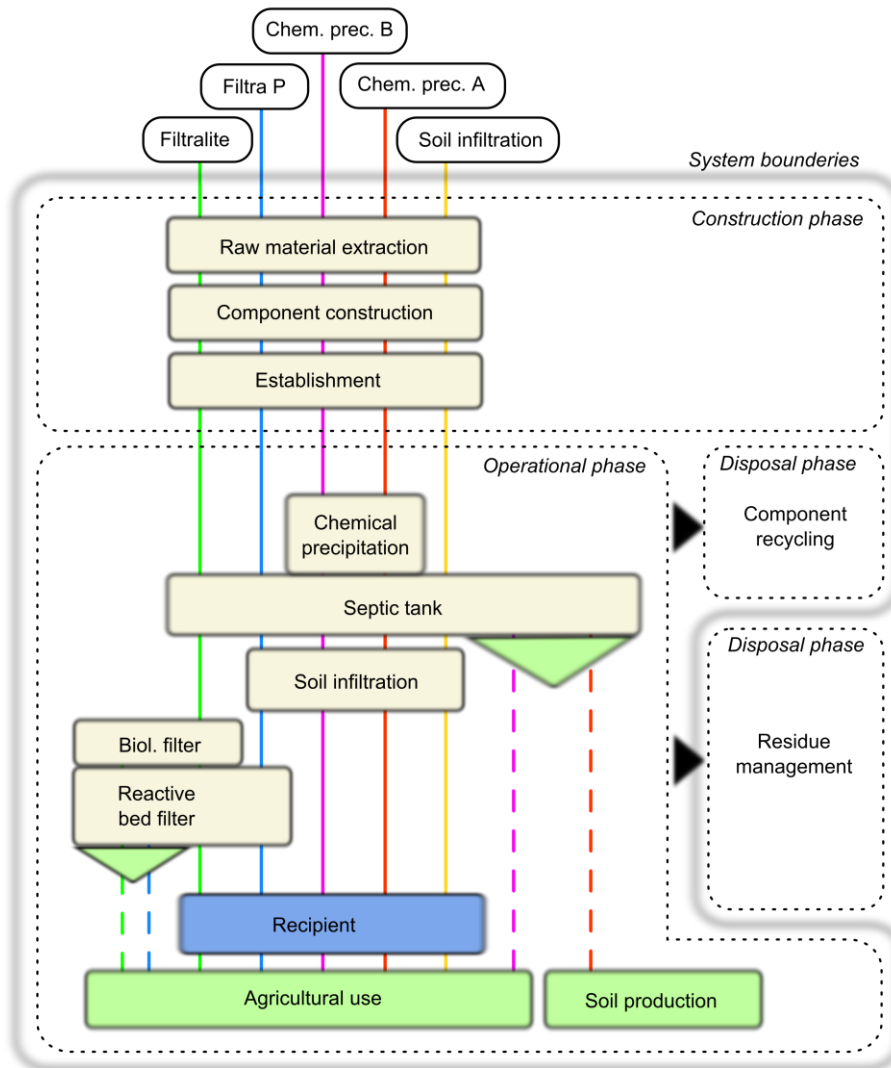


Figure 2. Simplified system scheme for the different wastewater treatment options included in the life cycle assessment in Paper I. Each line colour represents a particular wastewater treatment scenario. Dashed lines are pathways for nutrient recycling.

The region has a growing population with an increasing number of summer houses being turned into permanent housing, where sanitation systems are not satisfactory. Therefore, there is a strong need for effective and easy-to-implement on-site wastewater treatment solutions (RTK, 2003).

Scenarios

Three on-site wastewater treatment systems with high P removal performance were considered: Chemical precipitation, P removal using the reactive filter media Filtra P and Filtralite® P. These three systems were adopted as hypothetical replacements for the currently predominating system in the area (soil infiltration). A prerequisite when selecting the alternatives was the possibility to easily upgrade existing on-site wastewater

treatment facilities. The soil infiltration system was also used as a reference system for evaluation of the scenario of keeping the current level of technology.

The functional unit was defined as treatment of wastewater for one person equivalent (pe) during 1 year. The fertilizer value of plant nutrients in wastewater treatment residuals was also included in the system. The chemical precipitation scenario was divided into two sub-cases A and B. In case A the sludge is handled in a WWTP (the same as for the septic tank sludge produced in all other systems). In scenario B the sludge is hygienized by urea and then directly used in agriculture. The potential for nutrient recycling was limited by the heavy metal content in the sludge, which was estimated

based on measurements by Hellström *et al.*, (2003).

Inventory

In the inventory analysis, data concerning energy use, resource use, as well as emissions to air and water, were collected for each of the flows that were considered to be relevant. The main activities included in the studied life cycle were: raw material extraction, fabrication, establishment, operation, nutrient recycling (including hygienization if necessary) and disposal (Figure 2). Data were collected mainly from scientific papers, evaluation reports, national statistics and using information from suppliers of treatment facilities.

Impact assessment and normalization

The impact categories chosen for the impact assessment were: Depletion of abiotic resources, global warming potential, and eutrophication. The environmental loads from the inventory analysis were classified, i.e. assigned to the chosen impact categories. The impact assessment was performed using the equivalency factors given by Guinée (2002).

A better understanding of the magnitude of the different environmental impacts can be gained using normalisation, in which characterisation results are divided by the actual magnitude of each impact category for a certain geographic area (Baumann & Tillman, 2004). In this study, global average data, expressed in magnitude of intervention per capita (Guinée, 2002), were used to perform the normalization.

Phosphorus characterization techniques (Paper II)

In paper II, various characterization techniques were used to identify the chemical forms (species) of accumulated P in filter media used for on-site treatment of wastewater. Phosphorus speciation in exhausted filter media provides insights into the mechanisms behind P removal and provides guidance to the use of these media as fertilizer when recycled to agriculture. These aspects are relevant for the future development of bed filter solutions and

useful as a basis for realistic and reliable estimates during inventory and model design in LCAs.

XANES spectroscopy

X-ray absorption near edge structure (XANES) spectroscopy is a useful technique for characterization of chemical species of phosphorus in complex environmental samples (Kelly *et al.*, 2008). Spectra are derived from changes in the X-ray absorption coefficient around the X-ray absorption edge of the element of interest (the absorber). Edge energies are unique for different chemical elements; hence XANES is element-specific. Spectral features near the edge depend on the average local coordination environments of all absorber atoms in the sample (Kelly *et al.*, 2008).

XANES analysis was performed on all materials listed in Table 1 except soil samples. Interpretation of XANES data was carried out by a linear combination fitting (LCF) approach (Tannazi & Bunker, 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 phosphate adsorbed to aluminium oxide (boehmite) (Alox-P) or ferrihydrite (Feox-P). All data were analyzed using the Athena software (v0.8.056) (Ravel & Newville, 2005). The XANES data were collected at Beamline X-15B at the National Synchrotron Light Source, Brookhaven National Laboratory, Upton, 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.

ATR-FTIR and XRPD

X-ray powder diffraction (XRPD) and attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy were used as complementary techniques. In most filter samples, P species were undetectable by both ATR-FTIR and XRPD, probably because the

concentrations of any P mineral phases were too low to be detected (one of the Polonite samples was an exception). However, both techniques were used to verify the purity of the standards used for XANES data interpretation.

Soil investigations, mass balances and speciation modelling (Paper III)

To improve the knowledge basis for a proper treatment of soil infiltration systems within LCAs, a study on long term P-removal efficiency and the chemical mechanisms behind the removal process was carried out (Paper III).

Soil sample collection

The treatment systems included in this part of the project were uncovered soil treatment systems constructed as vertical-flow filter beds with an embanked surface. The wastewater was pretreated by a septic tank (Tullingsås and Halahult sites) or by ponds for particulate removal (Alsen and Rötviiken sites).

The soil treatment systems were sampled at five different depths: 0-5, 5-15, 15-30, 30-60 and 60-100 cm. For each site a reference sample was also collected. The reference sample was intended to represent filter bed material that had been unexposed to P-containing wastewater. At the Tullingsås site, sample collection was carried out at five different locations within the filter area for an in-depth analysis of the long-term removal capacity of the bed.

Mass balance calculations

A mass balance approach was used to determine the long-term phosphorus removal at the Tullingsås site. By determining the accumulated P (defined as total P in samples minus total P in a relevant reference sample) and by considering the volumes and bulk densities of the material, the total amount of P removed by the system could be estimated. Representative volumes were defined by means of Thiessen polygons and by use of the predefined sample depths (as shown in Figure 3). Samples and reference samples were analyzed for total P by nitric acid digestion and P determination with

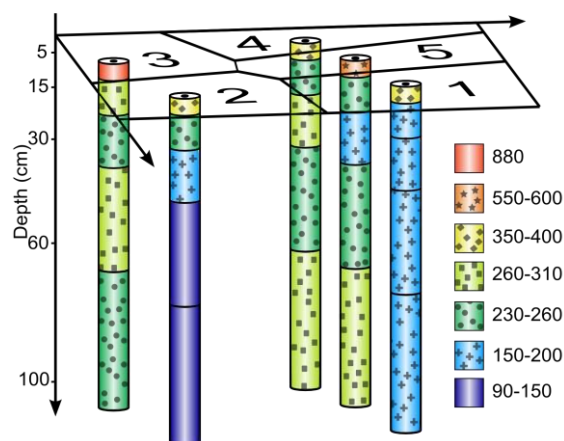


Figure 3. Accumulation of P (mg/kg) within different locations in the bed filter at the Tullingsås site.

inductively coupled plasma emission spectrometry (ICP-OES). The mass balance calculation was carried out by comparing the calculated P accumulation to the estimated P load during the operational life-span of the soil treatment system. The P load estimation was based primarily on a large number of inflow measurements carried out during 1992 and 1993.

Soil sample tests

Reactive aluminium and iron (hydr)oxides, as well as phosphorus associated with these fractions, were determined by ammonium oxalate (0.2 M oxalate buffer, pH 3) extraction (van Reeuwijk, 1995). The oxalate extraction was performed to investigate the importance of Fe and Al (hydr)oxides for the P fixation.

On reference samples and samples from the depth of 0-5 cm a series of batch experiments were performed at room temperature (21°C). In a *pH-dependence experiment*, field-moist samples were suspended in solutions (wet weight L/S~9) in polypropylene centrifuge tubes in duplicates and shaken for 5 days in an end-over-end shaker. The solutions consisted of 0.01 M NaNO₃ with different additions of acid (from 0 to 2.6 mM HNO₃) or base (from 0 to 1.7 mM NaOH).

In a *sorption experiment*, field-moist samples were suspended in solutions consisting of background electrolyte (0.01 M NaNO₃) to which different PO₄ additions (as NaH₂PO₄)

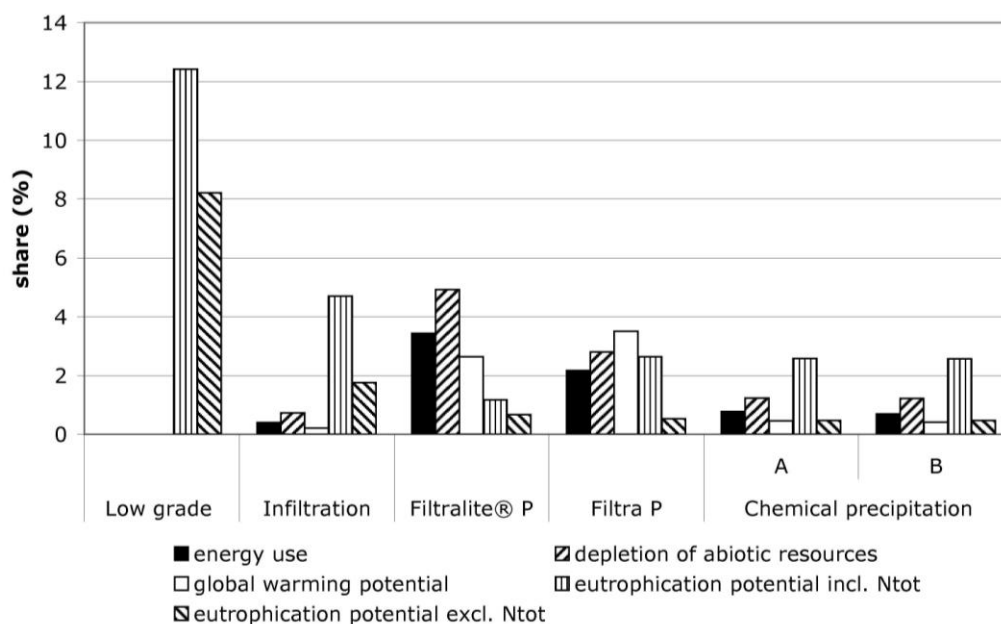


Figure 4. Environmental impacts (normalized to global average values) caused by the four studied wastewater treatment systems.

had been added (final concentration: 3.1, 6.2 and 12.4 mg P/l). These samples were also prepared in duplicates and shaken for 5 days in an end-over-end shaker.

The batch experiment samples were centrifuged after equilibration. The pH value and the alkalinity were then determined on unfiltered samples. After filtration through a 0.2 μm Acrodisc PF filter, the inorganic $\text{PO}_4\text{-P}$ concentration was determined colorimetrically whereas Fe, Al, Ca, Mg, K and Na were determined by ICP-OES and F^- , Cl^- and SO_4^{2-} by ion chromatography.

Chemical speciation modelling

The chemical equilibrium modelling software Visual MINTEQ (Gustafsson, 2009) was used to evaluate the conditions for precipitation of solid calcium phosphate phases. Activities of Ca^{2+} and PO_4^{3-} were calculated and used to investigate the conditions for precipitation. The following P phases were included in the investigation: amorphous calcium phosphate (ACP), octacalcium phosphate (OCP), dicalcium phosphate (DCP), dicalcium phosphate dihydrate (DCPD) and hydroxyapatite (HAp). Equilibrium constants and heats of reaction for soluble complexes were taken from the default thermodynamic database of Visual MINTEQ, whereas for mineral phases

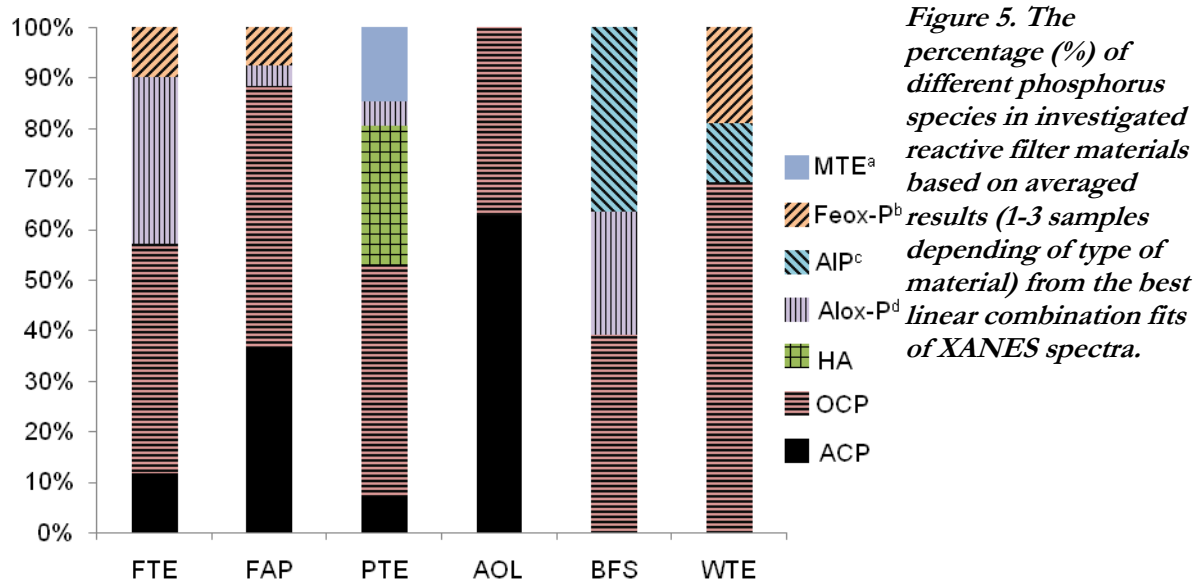
they were taken from Gustafsson et al. (2008).

RESULTS

Environmental impacts

In paper I the environmental impacts obtained using four treatment options were analyzed for the County of Stockholm case study (Figure 4). The conclusion was that the chemical precipitation system obtained the most favourable results from an environmental and resource conservation perspective. Both Filtralite® P and Filtra P produced large environmental impacts in energy-use-related areas, which could be primarily explained by energy-demanding material manufacturing processes.

Let us assume that P is the limiting nutrient for the surface waters that receive the wastewater and look at Figure 4 without considering the eutrophication potential caused by N compounds. Then it would be reasonable to state that in a global perspective the eutrophication potential is low compared to the energy use and global warming potential of the Filtralite P and Filtra P systems. By comparison, the infiltration system performed relatively well; however it needs to be pointed out that this system possessed no nutrient recycling



a. Monetite

b. P adsorbed to aluminium oxide (boehmite)

c. Hydrated aluminium phosphate

d. P adsorbed to ferrihydrite

potential. Another important conclusion was that the inventory showed that there is a lack of accurate data and knowledge regarding many important features of both new and conventional treatment techniques. Examples are the extent of phosphorus removal in infiltration systems, the life span of reactive filters and management of different phosphorus-enriched by-products.

Phosphorus characterization in reactive filter materials

By use of phosphorus K-edge XANES, Paper II examined the chemical forms of P present in a set of used filter materials (Figure 5). All six investigated filter media contained significant amounts of crystalline calcium phosphates. Furthermore significant amounts (14-60 %) of amorphous calcium phosphate were identified in all types of media for at least one of the differently treated samples, except for the media wollastonite and blast furnace slag. In Filtralite P and blast furnace slag, more than 35 % of the accumulated phosphorus was associated with Fe or Al.

Phosphorus removal in soil treatment systems

By means of the mass balance approach in Paper III (Figure 3) the P removed by one of the four investigated soil infiltration systems was estimated to be 6.4 % of the P load through wastewater during 16 years. A strong relationship between oxalate-extractable Al and P (Figure 6) indicated that AlPO_4 precipitation and/or sorption to poorly ordered aluminium compounds was the dominating P removal mechanism at most of the studied sites. As was evidenced by the batch experiments with acid and base addition in combination with chemical speciation modelling, calcium phosphate precipitation was likely not an important long-term P removal mechanism, with the possible exception of the Halahult site. However, in the sorption experiment with P solutions, saturation with respect to ACP was reached for all but the Tullingsås site. This indicated that ACP can work as a temporary pool for P when the P concentrations in the influent wastewater are high.

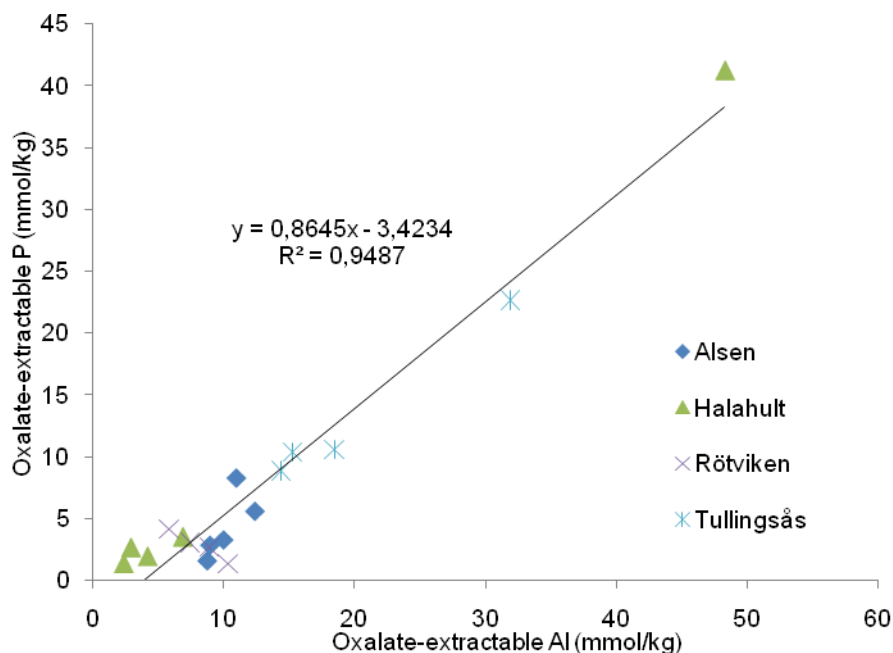


Figure 6. Relationship between oxalate-extractable Al and P for filter bed samples from all investigated soil treatment sites.

DISCUSSION

As already pointed out, a prerequisite for sustainable development is that the implementation of high-performance P-removal techniques also considers negative impacts with respect to other environmental issues than eutrophication. Useful decision support must provide indicators targeted to fulfil current regulations and additional objectives set up by the society. The results of Paper I show that LCA is suitable for compilation of such indicators. However, obviously the reliability of the results obtained are dependent on the quality of the methods used. Thus, the required resolution of the output must be backed up by high-quality input data, and system boundaries and result categories must be carefully chosen. Paper I provided insights into some issues that should be given priority to increase the reliability of future LCAs of on-site wastewater treatment systems. These issues and their connection to other activities in the project are discussed below.

Life-span of reactive filters

The life-span of reactive bed filters is a key to assessing the total environmental impact of these systems (Paper I). Development of materials with a less energy-consuming

production process or with a longer life span and higher P-removal capacity should reduce the energy-related impacts greatly. For this development knowledge regarding P removal mechanisms is essential. From a removal mechanism point of view the results in Paper II was consistent with other investigations (Drizo *et al.*, 2006; Gustafsson *et al.*, 2008; Bowden *et al.*, 2009) and it confirms that Ca phosphate precipitation is important for removal of P from wastewater by alkaline filter materials. However, the results from the study do not show the pathway by which the final P phases were formed and what species that actually is the solubility-controlling phase in a short-term perspective.

Long term P removal in soil treatment systems

In wastewater treatment prevention of eutrophication is a very central objective. Thus, the quality of the background data for nutrient emissions is of great importance. A particular weakness in current LCAs of on-site wastewater treatment systems is that the data quality on the long term phosphorus removal in conventional soil treatment systems is unreliable. However, including these systems in LCAs is of great interest since they are the most common systems in Sweden. In addition, they are required to

describe the current situation and a hypothetical “passive” scenario in which no advanced P removal techniques are introduced.

In the past, interpretations and decisions about long-term P removal in soil infiltration beds have been heavily dependent on inflow/outflow (I/O) measurements. In comparison to most I/O studies our result of 6.4 % P removal (Paper III) is extremely low. Aaltonen & Andersson (1996) reported between 38 % and 60 % P removal for 12 monitored beds in Sweden. According to the Swedish EPA (SEPA, 2008), 25-90 % P removal is obtained in soil treatment systems. These figures are heavily based on the work by Nilsson (1990). USEPA (USEPA, 2002) claims that between 85 and 95 % P removal is obtained (septic tank removal included) and refers to Sikora & Corey (1976).

An important limitation of I/O investigations is that they only provide a “snapshot” of dissolved P in different sampling points. However, without extensive data collected over a long period of time it is not possible to determine the net P removal with any degree of confidence. Furthermore a shortcoming of many studies in the past is that a scientifically based analysis of empirical results of P removal in relation to underlying chemical mechanisms has been lacking. The results in Paper III were consistent with several observations showing quick migration of phosphorus in septic system plumes (Robertson *et al.*, 1998; Robertson, 2008). However, variations in chemical, physical and operational properties between the applications investigated in Paper III and those studied earlier could not be completely ruled out as possible reasons to the discrepancy between this mass balance study and earlier I/O measurement studies.

In the light of the results in Paper III, the assumption of 70 % P removal in soil infiltration systems in Paper I was probably unrealistic. The choice of 70 % reflected the current guidelines (NFS 2006:7) but the results show that this assumption may not be justified in future LCAs.

Modelling eutrophication impacts

The analysis of how the current impact models for eutrophication work has revealed the need for improvements. Two main issues that must be handled by a proper eutrophication impact model for on-site wastewater treatment systems can be identified:

1. Retention of P during transport to the target recipient
2. Potential impact of leached P on the target recipient

The target recipient, i.e. the water body in focus for the study could be *e.g.* a nearby lake, a bay or the Baltic Sea as a whole.

Basically the limitation of the current eutrophication impact models when evaluating on-site wastewater treatment systems is the spatial resolution. The current models can treat the second issue to some extent but fail to address the first issue properly. The most detailed method provides retention factors on a country basis. Since LCA typically is handled on a quite a high level of abstraction this is not always a problem. However, the environmental sensitivity to eutrophication is very much dependent on local conditions and both the geographical location and the recipient type (groundwater or surface water) of the on-site system will have a large influence on the potential retention of P during transport to the target recipient. Thus, since there is a high demand for detailed indicators of potential eutrophication impacts for the management of on-site wastewater treatment, enhanced impact modelling is needed.

Paper I did not deal with eutrophication at a reasonable spatial resolution but future LCAs should try to handle this issue on a more detailed level. Appropriate estimates of retention are particularly important for the correct handling of systems that dispose of treated wastewater to the soil environment instead of discharging it directly to surface water.

Managing P recycling

The P removal techniques for on-site wastewater treatment result in a range of

different recycled products. Chemical precipitation systems will produce sludge similar to that from municipal wastewater treatment plants (with possible exception for the heavy metal content). On the other hand, the recycled product from a reactive bed filter consists of a P-enriched filter medium. Paper II gave insights into the chemical forms of phosphorus present in a number of different filter materials. From a P recycling perspective it would be preferable to capture phosphorus from wastewater in an easily soluble fraction readily available to plants. ACP would then be preferable to the more crystalline calcium phosphates HA or OCP. The Al- and Fe-associated P species are likely to be less soluble than the Ca phosphates, especially in acidic soils. Based on this simplified reasoning Absol would be the most efficient fertilizer while blast furnace slag and Filtralite P would be less beneficial (at least on acid soils). However, plant availability is a complex issue that cannot be assessed from the phosphorus species in the recycled material only. In essence, plant availability of P from filter materials will depend on properties of the agricultural soil, any material pre-processing (such as grinding or mixing with other soil amendments), and chemical and biological processes that influence plant uptake. Furthermore there is currently a large number of practical questions that remain unanswered regarding P recycling of filter materials, which make realistic recycling scenarios difficult.

In Paper I a simplified approach to P recycling was used that assumed that all nutrients from the material were plant-available. This approach is certainly not perfect but it might be argued that it is reasonable. In a longer time perspective the sorbed P might be available. Probably, this strategy will overestimate the benefits of P recycling. However, this error is probably not large compared to other existing errors in the LCA (which in any case is only a simplified model of reality). For example, it should have been fair to consider the benefits of systems that actually recycle P to the food chain compared to systems that recycle P to

applications that do not have any essential functionality (*e.g.* when producing soil for construction purposes).

CONCLUSIONS

Some important conclusions drawn from the work are listed below:

- LCA is a suitable tool for compilation of different environmental impacts of on-site wastewater treatment systems, but there is need for improved input data and model design for estimates of potential eutrophication.
- A key issue for the environmental performance of reactive bed filters is the manufacturing process of the filter material. Decreased energy demand per removed amount of P by *e.g.* more efficient production processes, increased removal capacity or completely new materials is desirable.
- In most reactive filter materials calcium phosphate precipitation seems to be the predominant removal mechanism. Trapped P commonly consists of crystalline calcium phosphates species such as octacalcium phosphate and hydroxyapatite. Several materials also accumulate significant amounts of amorphous calcium phosphate and some aluminium and iron bound P.
- The long-term phosphorus removal capacity in soil treatment systems might have been highly overestimated in past assessments in Sweden. Future research is needed to provide a reliable estimation of the P removal capacity in these systems. Such research should provide a scientifically based analysis of any empirically obtained results. In several past I/O studies such an analysis has been missing.
- Aluminium compounds rather than iron and calcium seem to be important for P removal in soil treatment systems.

FUTURE RESEARCH

To produce an improved LCA with high-quality input data and improved impact

models could prove to be a never-ending story. However, some aspects need to be further developed to obtain satisfying resolution and quality in the output. Further investigation of chemical P removal mechanisms (in both reactive materials and soil) is a priority since it will provide information useful for both P removal predictions and process optimization. A model for prediction of the changes in P removal efficiency over time should be developed. Hopefully such a model could be applied for both prediction of the P removal capacity of soil infiltration systems, the life span of reactive filters and P retention in soil during transport to surface water. However there is need for more research on a mechanistic level before such a model can be developed.

Studies of the dissolution pathways and equilibrium speciation would be valuable since such work would provide better insight into chemical speciation and dissolution kinetics. It can be hypothesized that a large part of the removal capacity is lost early during the operation of reactive bed filters due to fast initial leaching of Ca. By achieving

a more uniform Ca dissolution from the filter material over time it might be possible to increase the life span of the application. Another chemical issue is how the phosphate mineral crystallization and dissolution processes are affected by the presence of complex-forming organic substances in the wastewater. It is known that the crystallization process may be inhibited or delayed by the presence of complex-forming organic substances (van der Houwen *et al.*, 2003; Alvarez *et al.*, 2004; Song *et al.*, 2006), but it is unclear to what extent.

Another question is in what way LCA can be used as a decision support tool for on-site wastewater treatment. At present it is unclear on what level of decision-making sustainability aspects will be treated. An investigation of the opinions among stakeholders and in the on-site wastewater community should probably be of great value to the process of designing a well-aimed LCA. In future LCAs it would also be interesting to include other kinds of on-site wastewater treatment solutions than those treated in Paper I, such as black water and urine separation solutions.

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