

1 NOTICE: this is the author's version of a work that was accepted for publication in Water Research. A definitive  
2 version was subsequently published in *Water Research* **64**, 42-52, 2014.  
3 <http://dx.doi.org/10.1016/j.watres.2014.06.034>

## 4 **Phosphorus in soil treatment systems:** 5 **accumulation and mobility**

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## 17 **Abstract**

18 Septic tanks with subsequent soil treatment systems (STS) are a common treatment technique  
19 for domestic wastewater in rural areas. Phosphorus (P) leakage from such systems may pose a  
20 risk to water quality (especially if they are located relatively close to surface waters). In this  
21 study, six STS in Sweden (11 to 28 years old) were examined. Samples taken from the  
22 unsaturated subsoil beneath the distribution pipes were investigated by means of batch and  
23 column experiments, and accumulated phosphorus were characterized through X-ray  
24 absorption near edge structure (XANES) analysis. At all sites the wastewater had clearly  
25 influenced the soil. This was observed through decreased pH, increased amounts of oxalate  
26 extractable metals and at some sites altered P sorption properties. The amount of accumulated  
27 P in the STS were found to be between 0.32 and 0.87 kg m<sup>-3</sup>, which in most cases was just a  
28 fraction of the estimated P load (< 30%). Column studies revealed that high P concentrations  
29 (up to 6 mg L<sup>-1</sup>) were leached from the material when deionized water was applied. However,  
30 the response to deionized water varied between the sites. As evidenced by XANES analysis,  
31 aluminium phosphates or P adsorbed to aluminium (hydr)oxides, as well as organically bound  
32 P, were important sinks for P. Generally soils with a high content of oxalate-extractable Al  
33 were also less vulnerable to P leakage.

34

## 35 **1 Introduction**

36 Phosphorus (P) discharge from anthropogenic sources is a crucial factor for eutrophication of  
37 many inland aquatic systems worldwide (Smith, 2003). In most areas, agricultural activities  
38 are believed to account for the majority of the P discharge on an annual basis (e. g. Smith et  
39 al., 2005; Brandt et al., 2009) . The contribution from onsite wastewater treatment systems

40 (OWTs) is smaller but they can still be a relevant P source, especially in areas such as the  
41 Baltic sea region where the reduction of P loads is of high priority (Boesch et al., 2006; Wulff  
42 et al., 2007)

43 Among OWTs, the use of septic tanks with subsequent soil treatment systems (STS) is the  
44 most predominant treatment technique for domestic wastewater. The use of STS is extensive  
45 in rural parts of Australia, North America, Canada and parts of Europe in rural areas (Butler  
46 & Payne, 1995; USEPA, 2002; Beal et al., 2005; Weiss et al., 2008; Gill, 2011; Motz et al.,  
47 2012). In STS, the unsaturated subsoil beneath the soil trench and above the water table can  
48 be defined as the overall treatment system (Gill et al., 2009). In clay soils (which are not  
49 suitable for infiltration) the STS can be constructed using imported sand. The wastewater then  
50 has to be drained out at the bottom of the system and piped to a surface recipient.

51 Phosphorus removal in STS is attributed to chemical precipitation and sorption processes in  
52 the soil matrix. Formation of Al(III) and Fe(III) (hydr)oxide surface complexes or  
53 precipitation of Al(III), Fe(III) and/or Ca phosphates are all possible attenuation mechanisms  
54 (Robertson, 2003; Eveborn et al., 2012). In addition Fe(II) precipitates may form at low redox  
55 potential (Zanini et al., 1998).

56 From a recipient perspective it has been shown that OWT systems can be a significant factor  
57 for the P status of freshwaters under certain conditions (Macintosh et al., 2011; Withers et al.,  
58 2011); these authors suggested that the observed impacts are attributed to poor design or  
59 insufficient maintenance of the treatment systems rather than general leakage. However, in  
60 the scientific literature there has been observations of both high, variable and low P removal  
61 (e.g. Carroll et al., 2006; Lowe & Siegrist, 2008; Robertson, 2008; Eveborn et al., 2012;  
62 Robertson, 2012).

63 As support within management of decentralized wastewater sources, knowledge regarding  
64 long term P removal in STS and the P immobilization/mobilization mechanisms involved is  
65 important. Eveborn et al. (2012) used a mass balance approach to assess the P removal  
66 capacity of the unsaturated subsoil in a Swedish STS. The study gave evidence for a very poor  
67 P removal (~12%), but was limited to four sites with comparably high P loads. The aim of this  
68 study was to explore the validity of the results by performing additional (simplified) mass  
69 balance calculations and investigate both accumulation and mobility of P in the unsaturated  
70 subsoil of old STS. Specific aims were to:

- 71 1. Investigate the overall removal capacity in the unsaturated subsoil of the systems by  
72 calculating the amount of accumulated P.
- 73 2. Study the P leaching and P removal potential of soil materials from STS through pilot  
74 scale column experiments with reconstructed bed profiles.
- 75 3. Investigate the mechanisms behind the observed P retention and P release by  
76 evaluation of data from batch experiments and physical/chemical characterization  
77 (including X-ray absorption near edge structure (XANES) measurements).

## 78 **2 Materials and Methods**

### 79 **2.1 Investigated sites**

80 Six STS located in various parts of Sweden were investigated: Tullingsås (Tu) near Östersund  
81 N 63° 49.17', E 15° 31.09', Biverud/Glanshammar (Gl) near Örebro N 59° 19.95', E 15°  
82 27.90', Knivingaryd (Kn) near Nybro N 56° 54.45', E 15° 57.44', Luvehult (Lu) near Nybro N  
83 56° 52.59', E 16° 6.95', Ringamåla (Ri) near Karlshamn N 56° 21.94', E 14° 44.26' and  
84 Halahult (Ha) near Karlshamn N 56° 14.05', E 14° 58.06'. Among these sites Lu and Gl were

85 traditional single-household systems whereas the other ones served between 40 and about 200  
86 persons each (Table 1). The age of the sites varied between 11 and 28 years, the hydraulic  
87 load was between 0.9 and 33 cm d<sup>-1</sup> and the estimated P load was between 30 and 540 g m<sup>-2</sup>  
88 yr<sup>-1</sup> (Table 1). At the Kn site a lined pond (open to the air) was used as a pre-treatment step  
89 for particle removal while at all other sites septic tanks or similar sludge removal devices  
90 were used. The Tu, Ri and Ha sites were all constructed using parallel beds, which were  
91 loaded periodically. The areas of the beds were between 30 and 196 m<sup>2</sup> (Table 1) and the soils  
92 used were imported from local gravel pits. The pre-treated wastewater was distributed to the  
93 soil through conventional drainage distribution pipes. However, the Tu and Ha sites were  
94 constructed with infiltration surfaces open to the air (framed by an embankment). At these  
95 sites the wastewater was applied over the soil from a centred (Tu) or mobile (Ha) inflow  
96 device. The sites Tu, Gl and Lu were gravity-fed systems whereas the wastewater at the sites  
97 Kn, Ri and Ha was distributed through pumping. The wastewater was finally discharged to  
98 the groundwater (Gl, Kn and Lu sites) or to a nearby stream through a drainage collection and  
99 distribution system (Tu, Ri and Ha sites). None of the latter systems had any liners, and  
100 therefore the proportion of wastewater that is discharged through the drainage system is  
101 unclear.

## 102 **2.2 Soil sampling**

103 The STS were sampled at five different depths (where the filter bed was sufficiently deep) by  
104 collection of samples from the 0-5, 5-15, 15 – 30, 30 - 60 and 60 –100 cm layers by use of a  
105 spade (in total about 350 kg soil). Sample locations were selected as close to the wastewater  
106 source as possible. However, at the sites Ri and Lu limited accessibility prevented us to  
107 collect samples immediately adjacent to the inlet. Reference samples were also collected at  
108 each site, which represented filter bed material that had not been exposed to P-containing

109 wastewater. At sites where imported sand was used (Ha, Kn, Tu, Ri) one single reference  
110 sample from unused sand was used. However at the other sites, 2 (Lu) or 5 (Gl) reference  
111 samples were selected to be able to consider vertical heterogeneity in soil properties.  
112 Reference samples were collected above the distribution level. Where this was not possible  
113 (Lu 60-100, Gl 0-30, and Gl 30-100) offset samples were collected (offset >2 m). The dry  
114 bulk density was determined by collection of undisturbed soil cores (by use of metal  
115 cylinders) in four replicates. The cores were dried at 105°C before weighing. At all STS soil  
116 sampling was performed at one single location. After collection, all soil samples were placed  
117 in plastic bags and stored in an isolated room protected from freezing ( $T < 12\text{ }^{\circ}\text{C}$ ) prior to  
118 further use.

### 119 **2.3 Soil analyses**

120 Samples larger than 15 kg (all samples excluding reference samples) were homogenized in a  
121 concrete mixer (for at least 15 minutes) before collection of subsamples of volumes relevant  
122 for soil analyses. The subsamples were then stored at 4°C. Field-moist samples from the 0-5  
123 and 5-15 cm depths as well as the reference samples were analyzed for pH in deionized water  
124 (using a liquid to solid ratio of 2) with a combination electrode and a PHM93 Reference pH  
125 Meter (Radiometer A/S, Brønshøj, Denmark). Total C was determined for all samples (dried  
126 at 105°C) using a LECO CNS-2000 Analyzer. Samples for total P analysis were delivered to  
127 the laboratory at ALS Scandinavia AB in Luleå, Sweden, and analysed according to EPA  
128 methods (modified) 200.7 (ICP-AES) and 200.8 (ICP-QMS). This method was also used for  
129 elemental analysis of reference samples and included the elements Al, Fe, Ca, Mg, Mn, K and  
130 Si. Briefly, the soil samples were dried at 105°C and subsequently 0.1 g dried sample was  
131 melted with 0.375 g  $\text{LiBO}_2$  and dissolved in  $\text{HNO}_3$ . The loss of ignition was determined at  
132 1000 °C.

133 Reactive aluminium and iron (hydr)oxides, as well as phosphorus associated with these  
134 fractions, were determined by extraction with ammonium oxalate (0.2 M oxalate buffer, pH 3)  
135 (van Reeuwijk, 1995). However, apart from P associated with aluminium and iron  
136 (hydr)oxides, other P species that are unstable at low pH will also be dissolved by this extract  
137 (e.g. calcium phosphates). Field-moist samples (in duplicates) from the six sites were  
138 extracted using a liquid to solid ratio of 100:1, shaken for 4 hours in the dark in an end-over-  
139 end shaker. Oxalate-extracted Fe, Al and P were determined with inductively coupled plasma  
140 emission spectrometry (ICP-OES) using a Perkin-Elmer Optima 3000 DV instrument.

#### 141 **2.4 Batch experiments**

142 For the reference samples and for the 0-15 and 5-15 cm samples from the sites Lu, Kn, Ri and  
143 Tu, further analysis were made through batch experiments. These sites varied in terms of  
144 loading history as well as in basic chemical and physical properties and were selected also for  
145 the column experiment. Sorption properties were studied through equilibration (5 days  
146 shaking time at 21°C) of 4 g soil in 30 ml phosphate solutions ( $\text{NaH}_2\text{PO}_4$ ) of the following  
147 initial P concentrations: 0, 0.02, 0.05, 0.1, 0.15, 0.2, 0.3 and 0.5 mM. 10 mM  $\text{NaNO}_3$  was  
148 used as a background electrolyte. Another batch experiment was performed to study the pH  
149 dependence of P desorption. This experiment was set up in an identical manner but with  
150 additions of NaOH (0.5 mM) or  $\text{HNO}_3$  (0.5, 1, 2, and 3 mM) instead of P.

151 In both experiments the equilibrations were set up in duplicate. After the equilibration the  
152 samples were centrifuged and the pH value was determined on the unfiltered supernatant  
153 immediately after centrifugation. Subsequently the supernatant was filtered through a 0.2  $\mu\text{m}$   
154 Acrodisc® PF filter and the inorganic  $\text{PO}_4\text{-P}$  concentration was determined colorimetrically

155 with the acid molybdate method using flow injection analysis (Aquatec-Tecator autoanalyser,  
156 Foss Analytical, Copenhagen).

## 157 **2.5 Column experiments**

158 To investigate the P discharge from old or decommissioned STS (at 1 m depth) a column  
159 experiment was set up with reconstructed bed profiles. In the experiments, columns were  
160 loaded either with a reference material (silica sand, SiO<sub>2</sub> content 99,8 %) or with wastewater-  
161 loaded soils from four sites (Tu, Lu, Kn and Ri). The columns were 0.3 m in diameter, the  
162 experiment was reproduced in duplicate and soil columns prepared so that they reached a final  
163 depth of the subsoil equivalent to 1 m (Fig. 1). For the Ri columns where the depth of the real  
164 STS was less than 1 m, additional soil sampled from the bottom layer was added to reach a  
165 depth of 1 m soil in the column. Distribution and draining layers (height ~0.15 m) of  
166 macadam (16-32 mm) were put at the top and at the bottom of the columns, and a piece of  
167 geotextile was used to separate the subsoil from the drainage layer (Fig. 1 (a)). The leachate  
168 was collected in polyethylene containers (40 L) which were arranged so that they could be  
169 weighed *in situ*.

170 During the first 12 weeks of the experiment, the columns were fed with domestic wastewater  
171 (from a community with about 2500 inhabitants). A mechanical treatment (2 mm drum  
172 screen) was applied before transfer to a 1 m<sup>3</sup> buffer tank. The columns were fed from the  
173 buffer tank (Fig. 1 (b)), which was completely refilled with fresh wastewater once a week.  
174 The hydraulic loading rate was adapted to the design hydraulic loading rates given in the  
175 Swedish guidelines (Swedish EPA, 2003), which implies a value between 3 and 6 cm d<sup>-1</sup>  
176 depending on the grain size distribution of the soil (Fig. 1 (b)). The columns were fed  
177 intermittently with 3 hours interval. Detailed characterisation of the influent wastewater



178 quality was performed at weeks 1, 5 and 9 through spot-checks directly from the buffer tank.  
179 The samples were analysed at the laboratory at Uppsala Vatten och Avfall AB, Uppsala,  
180 Sweden, according to standardised methods (Table S1, supporting information). In short, the  
181 concentration of BOD, NH<sub>4</sub>-N and alkalinity varied between 120 and 160, 42 and 50 and 517  
182 and 569 mgL<sup>-1</sup> respectively (Table S1). Effluent water from each sample container (including  
183 untreated wastewater, see Fig. 1 (b)) was weighed (in order to follow mass flows) and  
184 sampled regularly (typically once a week) for analysis of pH and total P. At week 5 and 9 (24  
185 hours after characterization of the influent wastewater) also BOD<sub>7</sub>, PO<sub>4</sub>-P, NH<sub>4</sub>-N, NO<sub>3</sub>+NO<sub>2</sub>-  
186 N and NO<sub>2</sub>-N were determined in the leachate. The pH value was determined instantly after  
187 collection with a combination electrode and a PHM93 Reference pH Meter (Radiometer A/S,  
188 Brønshøj, Denmark). Samples for BOD<sub>7</sub> analysis were instantly delivered and analysed at the  
189 laboratory at Uppsala Vatten och Avfall AB according to international standards (ISO  
190 5815:1989). An additional sample volume was frozen (-18°C) for later analysis of remaining  
191 parameters. Inorganic forms of nitrogen were analysed using flow injection analysis (FIA,  
192 Aquatec-Tecator autoanalyser, Foss Analytical, Copenhagen, Denmark). PO<sub>4</sub>-P was  
193 determined as for the batch experiments and for total P unfiltered samples were first digested  
194 in acid potassium persulfate solution before subsequent analysis.

195 After the end of the first column experiment, the buffer tank was cleaned and filled with  
196 deionized water. An identical column experiment (but with deionized water instead of  
197 wastewater) was then started. This experiment was carried out for a period of 9 weeks. The  
198 buffer tank was recharged with fresh deionized water every third week. The hydraulic loading  
199 scheme, as well as the sampling scheme, was identical to that of the first experiment with  
200 wastewater (Fig. 1 (b)). However, characterization of effluent water was performed at week 5  
201 and 9 and included (in addition to total P and pH) only PO<sub>4</sub>-P. Analyses were conducted as

202 above. As a consequence of hydraulic failure (clogging) in the reference columns at the  
203 beginning of the deionized water experiment, the ref 1 and ref 2 columns had to be run with a  
204 decreased loading rate. The ref 2 column even had to be closed for most part of the  
205 experiment. However there was always sufficient leachate available for sampling.

## 206 **2.6 XANES analysis**

207 Soil samples from the 5-15 cm layer of the Lu, Ha and Tu site (dried at 105°C and ground in a  
208 mineral grinder) were mounted on caption tape and analysed using P *K*-edge XANES  
209 spectroscopy on beamline BL8 of the Synchrotron Light Research Institute, Thailand  
210 (Klysubun et al., 2012). The beamline were operated in fluorescence mode and the  
211 fluorescence signal was measured using an solid state Ge detector. The scans ranged from  
212 2100 to 2320 eV with a smaller energy step near the absorption edge (down to 0.2 eV  
213 between 2144-2153eV). The counting time was constantly 3 s. Between 3 and 9 scans per  
214 each sample were collected depending on the level of noise in the data, and subsequently  
215 merged.

216 The XANES data processing was performed by means of the Athena program in the Demeter  
217 Software Package (v 0.9.18) (Ravel & Newville, 2005). All samples and standards were  
218 calibrated to a common energy scale by setting the maximum of the first derivative of the  
219 spectrum of variscite to 2149.0 eV. Correction of any shifts on energy scale caused by  
220 monochromator drift could be performed since validation data for variscite periodically were  
221 collected. Merged spectra were normalized using a consistent procedure. In brief, a linear  
222 baseline function was subtracted from the spectral region below the edge (typically between -  
223 45 to -6 eV relative to  $E_0$ ), and spectra were normalized to unit edge step and quadrature

224 removed across the post-white-line region (typically between 26 to 170 eV relative to  $E_0$ ) to  
225 obtain normalized XANES spectra.

226 By means of a linear combination fitting (LCF) approach (Tannazi & Bunker, 2005) a set of  
227 spectra of known standards were combined and fitted to the sample spectra. All standards  
228 used in the evaluation have been characterized by XRD (Eriksson et.al., manuscript in  
229 preparation), and XANES data were collected at the same beamline as the samples. The  
230 standard compounds included amorphous calcium phosphate, octacalcium phosphate,  
231 hydroxyapatite, brushite, monetite, amorphous aluminium phosphate, phosphorus adsorbed to  
232 aluminium hydroxide, variscite, amorphous iron phosphate, phosphorus adsorbed to  
233 ferrihydrite, strengite, struvite, potassium tarankite, lecithin and phytate. In the fitting  
234 procedure no energy shifts were permitted and the sum of the weighting factors were not  
235 forced to one. With support from earlier studies (Eveborn et al., 2009), the first derivative was  
236 chosen as the fitting space. At most three standards were accepted in each fit and the fitting  
237 range was constrained to between -5 to 30 eV relative to  $E_0$ .

## 238 **3 Results**

### 239 **3.1 Characterization of reference soils**

240 According to element analyses there were no dramatic differences in the elemental  
241 composition of the soil between the six sites (Table S2). The somewhat higher Ca content at  
242 the sites Ri and Ha (17 and 30 mg g<sup>-1</sup> dw<sup>-1</sup> compared to around 10 mg g<sup>-1</sup> for the other sites)  
243 coincides with a higher pH value (around 8.9 for Ri and Ha compared to between 5.9 and 6.8  
244 for the other sites). This may be caused by the presence of calcite (CaCO<sub>3</sub>) at the Ri and Ha  
245 sites. The initial P content ranged from 0.15 to 0.31 mg g<sup>-1</sup>. Between 6 and 100 % of the total

246 P was oxalate-extractable (Table S2). According to the grain size distribution analysis the  
247 GD<sub>50</sub> values for the soils ranged from 0.21 to 3.93 mm.

### 248 **3.2 Phosphorus accumulation and correlation to soil properties**

249 The amount of accumulated P (calculated as the difference between total P in a sample and  
250 total P in the corresponding reference sample) typically varied between 0.15 and 0.6 mg g<sup>-1</sup>  
251 (Fig. 2 and Fig. S1). However, the Lu site showed evidence for stronger (up to 1.2 mg g<sup>-1</sup>) P  
252 accumulation (Fig. 2). An even stronger P accumulation (2.28 mg g<sup>-1</sup>) was observed for the  
253 Tu 0-5 sample. However, this result was associated with an extreme (and abnormal) amount  
254 of organic content (Fig. S1) and this particular layer visually resembled sewage sludge.

255 A distinct relationship between oxalate-extractable P and oxalate-extractable Al was observed  
256 for wastewater loaded samples ( $r^2 = 0.92$ ,  $p < 0.001$ ), whereas the correlation between  
257 oxalate-extractable P and oxalate-extractable Fe was much weaker ( $r^2 = 0.6$ ).

258 As evidenced by the pH-dependence experiment, the solubility of the bound P was generally  
259 lowest at pH values ranging from 4 to 6 (for the Kn, Lu, Ri and Tu soils, see Fig. S2). All  
260 sites showed an increasing P solubility when the pH value was decreased further (pH < 4).  
261 Except for the Tu site (for which no high-pH data were available), an increased P solubility  
262 was observed at higher pH starting at pH 5.5 for Lu and at around 6 for the sites Kn and Ri.

263 If the layer thickness and the density of the soil are considered, the total P accumulation on a  
264 volume basis in each bed can be summed up to 0.32, 0.32, 0.46, 0.66, 0.73 and 0.87 kg m<sup>-3</sup> for  
265 the Gl, Ri, Ha, Kn, Tu and Lu sites respectively (Fig. 3). Among the studied sites no  
266 relationship could be established between the estimated P load and the amount of accumulated  
267 P (Fig. 3).

### 268 **3.3 XANES Analysis**

269 Of the 15 standard compounds included in the evaluation, 8 standards (Fig. 4 (a)) were  
270 represented in any of the ten best fits at weights above 10 % (Table 2). In both the Ha, Lu and  
271 Tu samples the XANES analysis indicated significant amounts of P bound as amorphous  
272 aluminium phosphates or as P adsorbed onto aluminium (hydr)oxide surfaces. The best fits  
273 resulted in weights between 24 and 62% of these phases (Table 2, Fig. 4 (b)). Organically  
274 bound phosphates were also well represented in all analyses (weights between 35 and 43%).  
275 Evidence for the importance of calcium phosphates was found only in the Ha sample where  
276 calcium phosphates predominated with 43% weight in the best fit (Table 2, Fig. 4 (b)).

277 For the best fits (Fig. 4 (b)) the Athena software reported R factors between 0.022 and 0.045.  
278 In general the distribution of P between Al, Ca and organically bound P was stable among the  
279 10 best fits (Table 2). The differences between the fitting results from the Lu site was caused  
280 by a small amount (weight less than 10%) of a third component, the identity of which differed  
281 in the fits. The weight of iron phosphates never exceeded 16% at any site.

### 282 **3.4 Soil properties as affected by wastewater**

283 When comparing reference samples and wastewater-loaded samples, several differences in  
284 soil properties were observed. In the wastewater-loaded samples, the pH of the top layer was  
285 typically between 1 and 2 pH units lower than in the reference samples. Further, oxalate-  
286 extractable metals had increased considerably in the deeper layers at many sites (Fig. 2, Fig.  
287 S1). However, there were no distinct patterns in the depth distributions of oxalate extractable-  
288 metals (Fig. S1). The sorption experiment revealed that for the Lu site, the sorption capacity  
289 was higher in reference samples than in wastewater loaded samples (Fig. S3). In fact, the  
290 opposite was true for the Tu site while no significant differences were observed at the Kn and

291 Ri sites. According to the sorption data of the reference samples, the soils at the Tu and Lu  
292 sites were superior to the Kn and Ri sites in terms of the P removal capacity (Fig. S3). At an  
293 equilibrium concentration of  $1 \text{ mg P L}^{-1}$  the Lu and Tu reference samples both removed more  
294 than  $50 \text{ mg P kg}^{-1}$  whereas the Kn and Ri sites removed less than  $30 \text{ mg kg}^{-1}$ . However, as  
295 evidenced from the mass balance calculations, the Tu, Lu, Kn and Ri sites had removed  
296 between 213 and  $680 \text{ mg P kg}^{-1}$ . Thus, the laboratory-established P removal capacities were  
297 about an order of magnitude lower than those obtained through mass balances.

### 298 **3.5 Phosphorus leaching in the column experiment with wastewater**

299 After five weeks of wastewater load biological processes in the columns were active and  
300 performed well in terms of nitrification and organic degradation. More than 97 % of the N  
301 ( $\text{NO}_3\text{-N} + \text{NO}_2 + \text{NH}_4\text{-N}$ ) was present as  $\text{NO}_3$  and the BOD concentrations were below the  
302 detection limit of  $3 \text{ mg L}^{-1}$  (data not shown). In week 9 the results indicated 100% nitrification  
303 and the reduction of BOD was still complete. The fraction of total P that was inorganic  $\text{PO}_4\text{-P}$   
304 (as evidenced by the acid molybdate method), varied between 13 and 80 % with a mean value  
305 of 50 % (data not shown).

306 The total P concentrations were generally low in the effluent waters during wastewater load  
307 (Fig. 5). The Kn and Ri sites had relatively weak P removal (the effluent P concentrations  
308 ranged from  $0.8$  to  $3 \text{ mg L}^{-1}$ , which corresponded to between 74 and 85 % P removal on mass  
309 basis). By contrast the Lu and Tu sites had very strong P removal, with effluent P  
310 concentrations always being  $<0.3 \text{ mg L}^{-1}$ , corresponding to 97 % P removal (Fig. 5). For the  
311 silica reference a small and relatively constant P removal (18 % on mass basis) was observed  
312 and the effluent concentrations varied between 3 and  $5 \text{ mg L}^{-1}$  (Fig. 5). The amount of P

313 accumulated during the loading period was around  $8.3 \text{ mg P kg}^{-1}$  for the Lu and Ri sites and  
314 around  $9.6 \text{ mg P kg}^{-1}$  for the Kn and Tu sites.

315 The pH value in the effluent was variable (Fig. S4). The Ri and Silica reference columns were  
316 following the pH of the influent wastewater closely (around pH 8), whereas the other sites  
317 generally had a lower and more variable pH (5.7- 7.7). The Kn site had a rising pH trend  
318 (from about pH 7 to pH 7.7) during the experiment and was approaching the pH value of the  
319 influent wastewater. However, the Lu and Tu sites did not follow any distinct pattern and the  
320 pH varied between 5.7 and 7.7.

### 321 **3.6 Phosphorus leaching in the column experiment with deionized water**

322 When the influent to the columns was shifted from wastewater to deionized water a dramatic  
323 colour shift occurred in the effluent, i.e. from transparent and clear to yellowish or brownish  
324 with high turbidity. However, this observation did not bring about a consistently different  $\text{PO}_4$   
325 : total P ratio. The concentration of dissolved  $\text{PO}_4$ -P in the effluent was in the range from 20  
326 to 80 % of the total P with a mean value of around 50 %, i.e. similar as in the first column  
327 experiment with wastewater. The P concentration in the effluent varied between the different  
328 sites, but effluent P concentrations were consistently higher than during wastewater load (Fig.  
329 5). The Kn columns generally had the highest effluent P concentrations which were initially  
330 up to  $6 \text{ mg L}^{-1}$  (Fig. 4). After week 4 the Kn column effluent concentrations stabilized at  
331 around  $3 \text{ mg L}^{-1}$ . The silica reference columns and the Ri site columns followed a similar  
332 pattern with initially high effluent P concentrations (up to 4 and  $3 \text{ mg L}^{-1}$  respectively) and  
333 then the P concentrations decreased to about  $2 \text{ mg L}^{-1}$  at the end of the experiment (Fig. 4).  
334 The P leaching patterns for the Ri and Lu site columns were different and the P discharge  
335 from these columns were significantly lower. In the latter two columns the dissolved P

336 concentration was quite stable from week 6 onwards, with dissolved P ranging from about 1  
337 to 1.5 mg L<sup>-1</sup> (Fig. 4).

338 The shift from wastewater to deionized water generally resulted in a pH increase in the  
339 effluent waters except for the Ri site columns and the silica reference columns, for which pH  
340 value decreased a little (Fig. S5). At the end of the experiment the pH value in all columns  
341 ranged from 6.7 to 7.7.

## 342 **4 Discussion**

343 The decrease in soil pH and the increase in oxalate-extractable metals that we observed (Fig.  
344 2, Fig. S1) is consistent with other studies (Robertson, 2003; Eveborn et al., 2012). In most  
345 cases (neutral to strongly acid soils) a pH decrease will probably favour the P removal, as  
346 sorption and precipitation processes that involve iron and aluminium (hydr)oxides are usually  
347 more efficient at low pH. According to the pH-dependence results (Fig. S2), the lower pH  
348 limit (below which P solubility rapidly increases) are between pH 3.5 and 4. In calcareous  
349 soils the pH decrease might prevent the formation of calcium phosphates as these  
350 precipitation processes is most effective at pH >9 (Eveborn et al., 2009).

351 The increasing levels of oxalate extractable aluminium and iron over time (Fig. 2, Fig. S1)  
352 may enhance the P removal. It is likely that both precipitation of aluminium phosphates and  
353 surface complexation reactions occur. As concluded earlier (Eveborn et al., 2012), the P to Al  
354 ratio in the oxalate extract is larger (0.73 according to linear regression of oxalate-P vs.  
355 oxalate-Al) than would be expected if surface complexation reactions alone would be  
356 responsible for the P removal. The XANES analysis confirms the importance of aluminium  
357 chemistry (Table 2) but the spectral differences between amorphous aluminium phosphates  
358 and phosphorus adsorbed to aluminium (hydr)oxides is small (Fig. 4 (a)). Thus, it is not



359 evident that any of these two mechanisms predominates at any of the sites. Aluminium-rich P  
360 phases have been identified as important sinks for P in similar environments (Zanini et al.,  
361 1998; Arai & Livi, 2013). According to the XANES analysis (Table 2), even organic P may  
362 play an important role in the P accumulation. However, the long term stability of this P pool is  
363 unclear. Degradation of the organic substances will result in the release of mineralized P.

364 Several mechanisms have been proposed that may cause the transfer of surface-bound P into  
365 stable P pools (Robertson, 2008), which may in the long run increase the P sorption capacity  
366 of soils. In this study we observed clear discrepancies (up to over tenfold) between laboratory  
367 sorption data on reference samples and the long term P accumulation in the field (based on  
368 mass balances). Changes in pH and in oxalate-extractable metals during the wastewater load  
369 can partly explain these discrepancies and therefore the results do not necessarily imply that  
370 any P will be immobilized into inactive, “insoluble” forms. In fact Lookman et al. (1995)  
371 found that all oxalate- extractable P was reversibly fixed in a selection of acid sandy soils  
372 ( $3.9 < \text{pH}_{\text{KCL}} < 5.7$ ).

373 It is clear from our P load estimates and P accumulation calculations that the P removal  
374 capacity in the subsoil can be easily exceeded. From this we can conclude that the long term  
375 performance of STS are much dependent on the wastewater load. Let us assume that the  
376 phosphate concentration in a septic tank effluent is on average  $10 \text{ mg L}^{-1}$ , that the long term  
377 hydraulic loading rate is  $0.6 \text{ cm d}^{-1}$  and that the available soil volume for treatment in the  
378 unsaturated subsoil is  $1 \text{ m}^3$  per meter of drainage tube. In such a scenario (close to that of  
379 Robertson (2012)), the STS studied here will theoretically (as estimated from mass balance)  
380 be able to accumulate P during 15 to 40 years of wastewater load, the exact value depending  
381 on the soil properties (Fig. 3). However, if the hydraulic loading rate is increased to  $3 \text{ cm d}^{-1}$   
382 (close to the maximum load according to USEPA guidelines, but still common with Swedish

383 design specifications) the time it takes to saturate the system will decrease to between 3 and 8  
384 years. Accordingly, the hydraulic loading rate is crucial for the P mass balance calculations.

385 We expected poor P removal when the already overloaded soil (according to P mass  
386 balances) was subject to further wastewater application but the results were contradictory  
387 (Fig. 5). The P discharge during the experiment was even lower than that reported from  
388 several short-term field studies (Nilsson & Stuanes, 1987; Aaltonen & Andersson, 1996;  
389 Nilsson et al., 1998). Accordingly, the results obtained do not seem to reflect the actual P  
390 leakage from old and heavily loaded STS. One possible explanation to the contradictory  
391 behavior could be that the collected soil layers were mixed (separately) during the setup of  
392 column replicates. This homogenization procedure may eventually expose new or hidden  
393 sorption or precipitation agents in the soil and eliminate macro pore pathways that might be  
394 present in an undisturbed soil profile.

395 The release pattern observed during the deionized water load is not fully understood. It is  
396 surprising that much higher concentrations of P were found in the leachate during deionized  
397 water load than during wastewater load (Fig. 5). Zurawsky et al. (2004) partly explained  
398 similar observations (leachate concentrations up to  $9 \text{ mg L}^{-1}$ ) from subsoils of STS by  
399 reductive dissolution of Fe-P phases. However, according to the XANES analysis, iron  
400 phosphates were not present to any considerable extent in the studied soils and the Fe  
401 concentrations in the leachate during wastewater load were usually very low ( $< 0.25 \text{ mg L}^{-1}$ ;  
402 data not shown). The evidence for substantial amounts of aluminium phosphates in the soil  
403 (Table 2) indicates that dissolution of these compounds is a possible P release mechanism.  
404 However, the P concentrations in the leachate were unreasonably high to be explained only  
405 through this mechanism. We hypothesize that the dramatic shift in ionic strength might  
406 destabilize particulate material and increase the mobilization of non-reactive P. A substantial

407 part of the P is organically bound (Table 2) and may be a source for such mobile forms of P.  
408 Destabilization mechanisms has been proved to be important elsewhere (Laegdsmand et al.,  
409 2005). Although no significant changes in the ratio of PO<sub>4</sub>-P:total P could be observed in the  
410 experiments (when comparing data from the periods of deionized water load and wastewater  
411 load) this hypothesis might still be possible.

412 The result from the column study using deionized water load, indicates that certain STS bed  
413 material cause substantial wash-out of P. The P wash-out could be caused by e.g. ground  
414 water inflow, diluted wastewater application or long-term drainage after decommissioning.  
415 However, the sites Lu and Tu, which had the highest oxalate extractable aluminium contents  
416 (Fig. 2 and Fig. S1), showed much less P discharge during both wastewater application and  
417 deionized water application (Fig. 4) in comparison to the other sites. These observations  
418 emphasize the role of aluminium chemistry for efficient P removal and are supported by other  
419 desorption studies on acid sandy soils (Lookman et al., 1995). The findings also show that  
420 despite favourable conditions for strong P fixation, significant amounts of P can be released.  
421 In terms of groundwater quality even a discharge of 1 mg P L<sup>-1</sup> is substantial. Hinsby et al.  
422 (2008) suggested 0.08 mg P L<sup>-1</sup> as threshold value for P in Danish ground water systems (for  
423 the protection of dependent ecosystems).

## 424 **5 Conclusion**

- 425 • Phosphorus removal in the unsaturated subsoil of STS is limited, and the risk for P  
426 leakage will be dependent on the long term magnitude of the P load. Thus, STS in  
427 close proximity to water bodies will pose a risk for significant P leakage.

- 428       • It is not safe to assume that P accumulated in STS is immobilized irreversibly. The  
429       vulnerability to wash-out of P through groundwater through-flow or atmospheric  
430       precipitation could be high.
- 431       • In the investigated sandy soils both the P accumulation and the vulnerability to wash-  
432       out are correlated to the amount of oxalate-extractable Al. In the most P-retaining STS  
433       P is accumulated mainly as aluminium phosphates or as P associated with aluminium  
434       oxyhydroxide surfaces, although organically bound P was also an important phase  
435       according to the XANES analysis.

## 436   **Acknowledgements**

437   The authors would like to acknowledge Mirsada Kulenovic and her colleagues at the  
438   department of Soil and Environment at the Swedish University of Agricultural Sciences in  
439   Uppsala, Agnieszka Renman, KTH and Wantana Klysubun, SLRI for technical support. We  
440   are also thankful to the municipalities of Nybro, Karlshamn and Krokomb as well as to Lars  
441   Eveborn and Thomas Molin (private homeowners) who put their treatment systems at our  
442   disposal. Thanks to The Swedish Agency for Marine and Water Management and to The  
443   Swedish Research Council Formas (project no. 2006-632) for financial support of this  
444   research.

445

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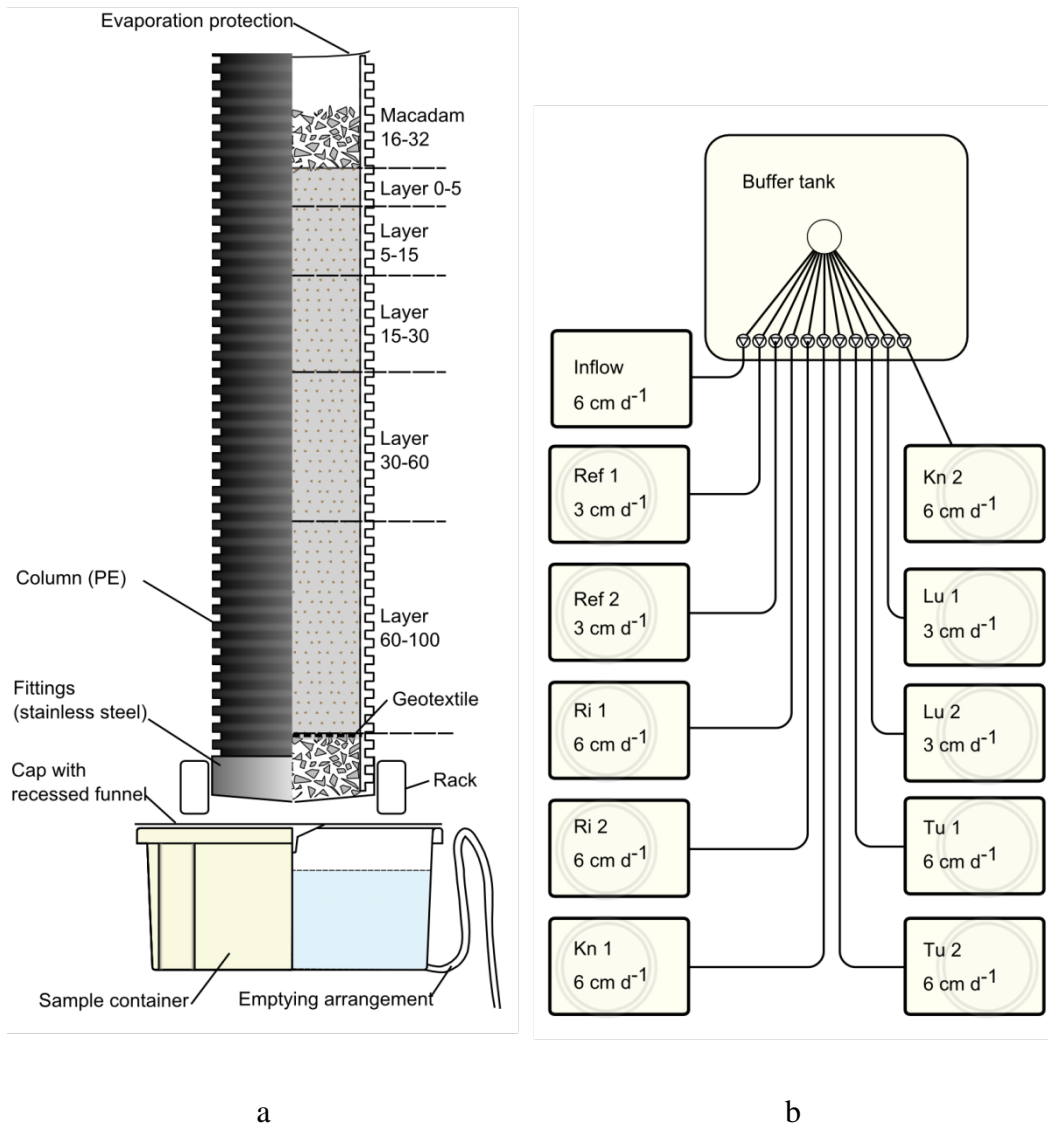
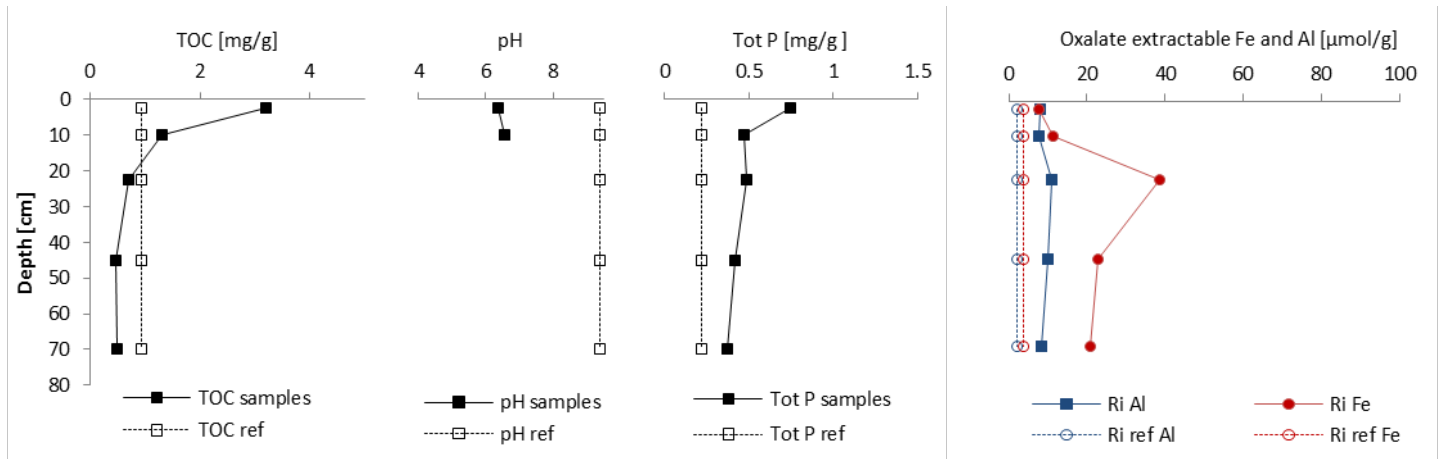
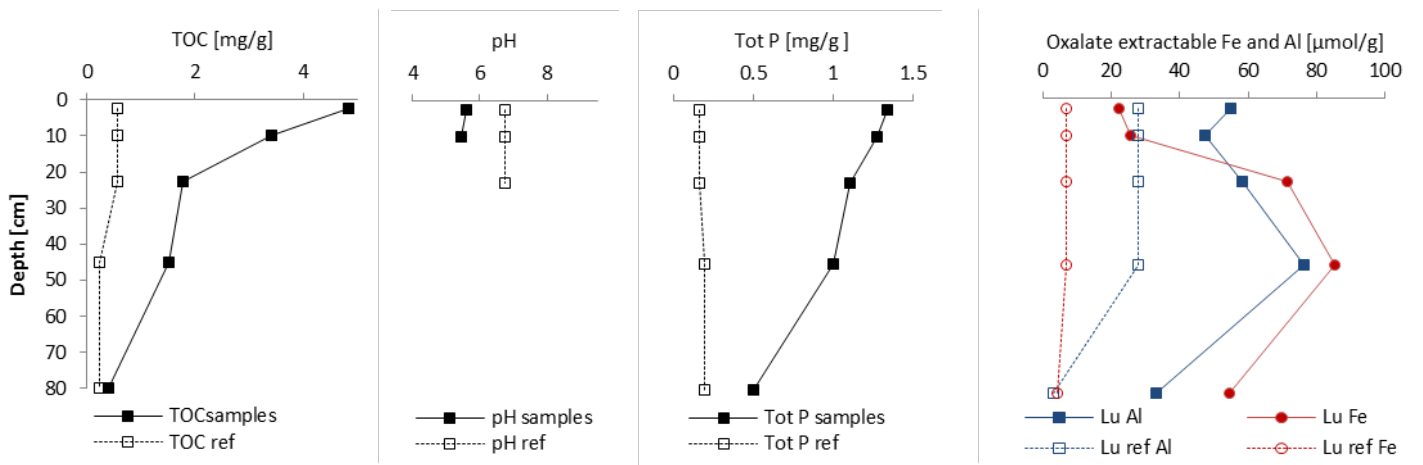


Figure 1. Layout of columns and sample containers (a) and schematic view over the column experiment including soils and loading rates (b).



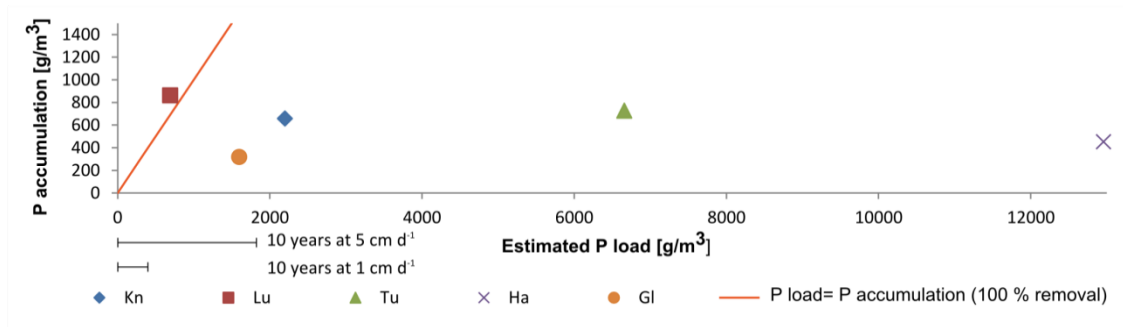


a) Ri site



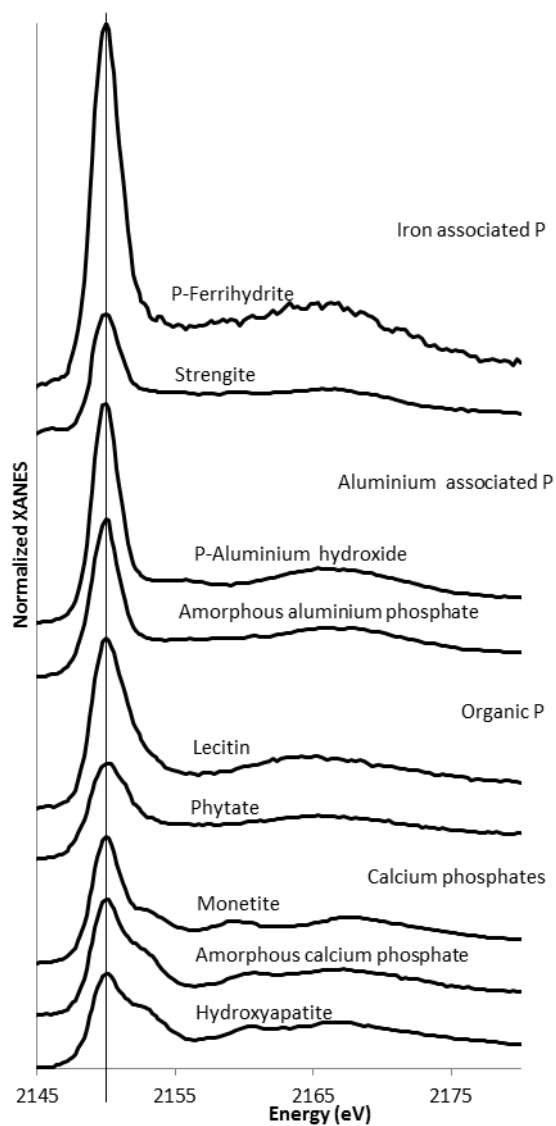
b) Lu site

Figure 2. Depth distribution of different soil properties for samples and reference samples at a) site Ri and b) site Lu.

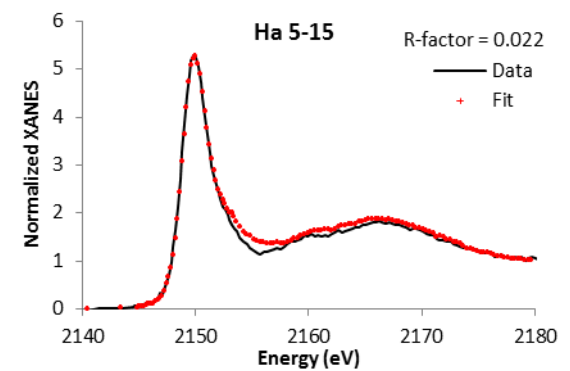
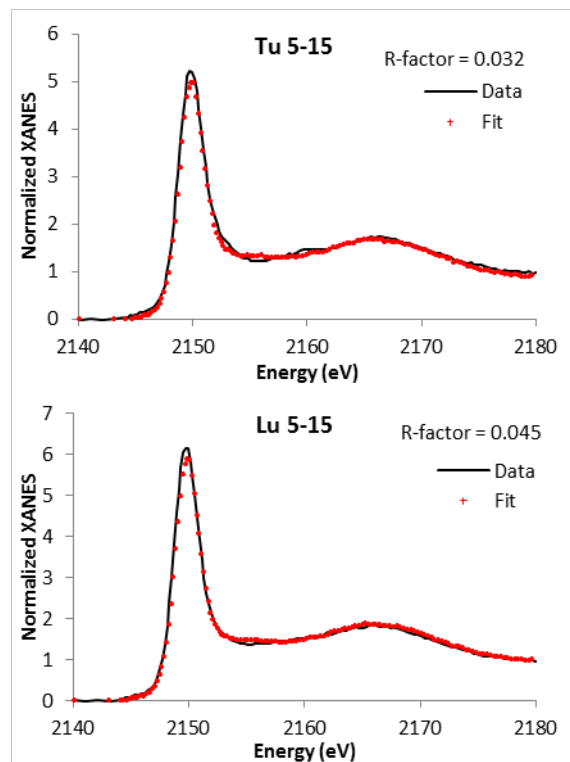


557 Figure 3. P accumulation at the studied sites in relation to the estimated P load. P load  
 558 estimations and the basis for these are given in Table 1.

559



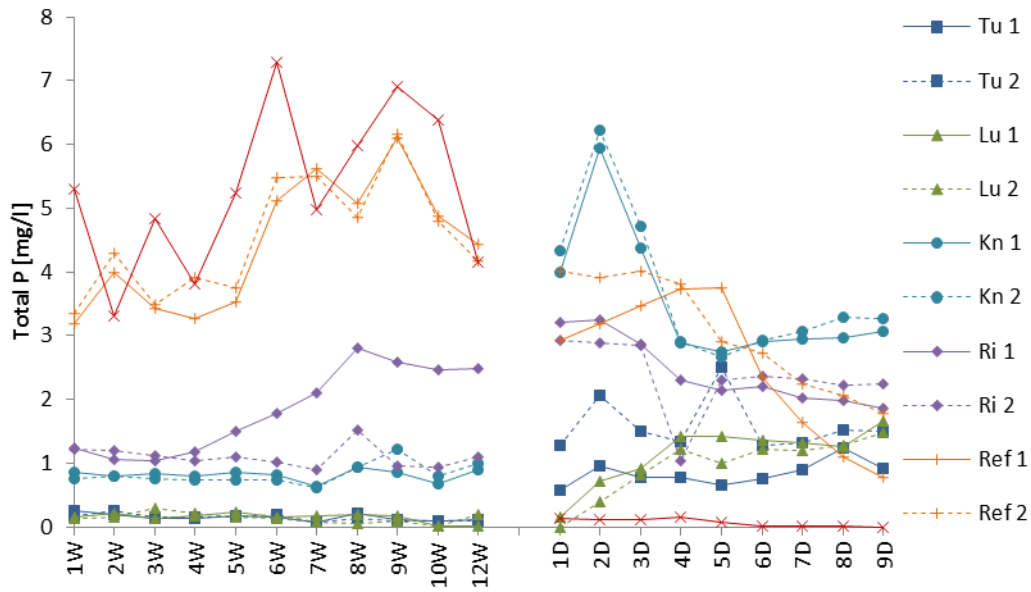
a



b

560 Figure 4. Spectra for standards represented (with weights >10%) in the 10 best linear  
 561 combination fits for the samples (a) and overlaid plots of sample data and the best (lowest R-  
 562 factor) linear combination fits (b).

563



564

565 Figure 5. Total P concentrations in inflowing water and leachates from the Tu site column  
 566 replicates (Tu 1, Tu 2), Lu sites column replicates (Lu 1, Lu 2), Kn site column replicates (Kn  
 567 1, Kn 2), Ri sites column replicates (Ri 1, Ri 2) and silica reference replicates (Ref 1, Ref 2)  
 568 during 12 weeks of wastewater water load (W1-W12) and 9 weeks of deionized water load  
 569 (D1-D9).

570

Table 1. Description of studied soil treatment systems

	<b>Tu</b>	<b>GI</b>	<b>Lu</b>	<b>Kn</b>	<b>Ri</b>	<b>Ha</b>
<b>Design base (pe)</b>	225	5	5	75	150	100
<b>Connected (pe)</b>	n.a.	6	4	40	n.a.	n.a.
<b>Surface area(s) (m<sup>2</sup>)</b>	2x196	30	50	80	2x160	2x50
<b>Hydraulic load<sup>a</sup> (cm d<sup>-1</sup>)</b>	33	2.2	0.9	25	n.a.	30
<b>P load (g m<sup>-2</sup> yr<sup>-1</sup>)</b>	370 <sup>c</sup>	80 <sup>b</sup>	30 <sup>b</sup>	200 <sup>b</sup>	n.a.	540 <sup>c</sup>
<b>Age (year)</b>	18	20	23	11	28	24
<b>Pre treatment</b>	Septic tank	Septic tank	Septic tank	Lined pond	Septic tank	Septic tank
<b>Wastewater distribution</b>	Gravity fed, open surface distribution	Gravity fed, drain field	Gravity fed, drain field	Pump fed, drain field	Pump fed, drain field	Pump fed, open surface distribution
<b>Thickness of soil bed (m)</b>	>1	>1	>1	0.9	0.8	0.9
<b>Discharge</b>	Drained to surface water	Ground-water	Ground-water	Ground-water	Drained to surface water	Drained to surface water

<sup>a</sup> Based on annual mean flows and active infiltration areas (where several beds are shifted). Mean flows for the sites GI, Lu and Kn have been calculated based on a water usage equivalent to 180 L person<sup>-1</sup> d<sup>-1</sup> and 60 % home attendance. Mean flows at other sites taken from Bylund (2003).

<sup>b</sup> An estimation based on mean flows (as described above), total infiltration area and a 10 mg L<sup>-1</sup> P concentration in the wastewater (Jönsson et al., 2005).

<sup>c</sup> Calculated from a dataset of ~50 inflow and P concentration measurements (Bylund, 2003) and total infiltration area.

571

572

Table 2. Fitting results for linear combination fit performed in first derivative space with an energy range between -5 and 30 eV. Standards represented with weights <10% not reported.

Category	Standard component	Weight, best fit	Presence in 10 best fits										Mean weight
Ha 5-15													
Al-P	Amorphous aluminium phosphate				x	x	x	x					0.29
	P adsorbed to Al(OH) <sub>3</sub>	0.24	x	x						x	x	x	0.21
Ca-P	Apatite				x		x			x			0.41
	Amorphous calcium phosphate	0.43	x		x		x	x	x		x	x	0.47
Organic P	Phytic Na	0.35	x	x				x	x				0.32
	Lecithin				x	x				x		x	0.27
Fe-P	P adsorbed to ferrihydrite						x		x		x		0.13
Lu 5-15													
Al-P	Amorphous aluminium phosphate	0.3	x										0.3
	P adsorbed to Al(OH) <sub>3</sub>	0.32	x	x	x	x	x	x	x	x	x	x	0.45
Organic P	Phytic Na	0.36	x	x	x	x	x	x	x	x	x	x	0.4
Ca-P	Monetite				x								0.11
Tu 5-15													
Al-P	Amorphous aluminium phosphate	0.28	x	x	x	x			x		x		0.37
	P adsorbed to Al(OH) <sub>3</sub>	0.2	x					x	x		x	x	0.27
Ca-P	Monetite									x	x		0.23
	Amorphous calcium phosphate						x		x	x			0.19
Organic P	Phytic Na	0.43	x	x	x		x	x	x	x	x	x	0.4
Fe-P	P adsorbed to ferrihydrite				x		x	x					0.11
	Strengite				x						x		0.26