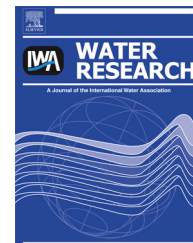




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Effect of organic load on phosphorus and bacteria removal from wastewater using alkaline filter materials

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ABSTRACT

The organic matter released from septic tanks can disturb the subsequent step in on-site wastewater treatment such as the innovative filters for phosphorus removal. This study investigated the effect of organic load on phosphorus (P) and bacteria removal by reactive filter materials under real-life treatment conditions. Two long-term column experiments were conducted at very short hydraulic residence times (average ~5.5 h), using wastewater with high (mean ~120 mg L⁻¹) and low (mean ~20 mg L⁻¹) BOD₇ values. Two alkaline filter materials, the calcium-silicate material Polonite and blast furnace slag (BFS), were tested for the removal capacity of total P, total organic carbon (TOC) and Enterococci. Both experiments showed that Polonite removed P significantly ($p < 0.01$) better than BFS. An increase in P removal efficiency of 29.3% was observed for the Polonite filter at the lower concentration of BOD₇ ($p < 0.05$). Polonite was also better than BFS with regard to removal of TOC, but there were no significant differences between the two filter materials with regard to removal of Enterococci. The reduction in Enterococci was greater in the experiment using wastewater with high BOD₇, an effect attributable to the higher concentration of bacteria in that wastewater. Overall, the results demonstrate the importance of extensive pre-treatment of wastewater to achieve good phosphorus removal in reactive bed filters and prolonged filter life.

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1. Introduction

On-site wastewater treatment is generally based on natural soil infiltration systems, which are reported to exhibit acceptable purification performance with respect to organics,

nutrients and bacteria (Beal et al., 2005). However, this technology has recently been questioned in terms of aspects such as phosphorus (P) removal efficiency and sustainability (Eveborn et al., 2012). Some studies also indicate that soil infiltration systems may pose a bacterial pollution risk to

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groundwater (Stevik et al., 1999). Therefore, on-site filter systems based on specially engineered media have been developed with the aim of removing P from residential wastewaters in rural areas (Renman and Renman, 2010; Drizo, 2012). Extensive research world-wide on different filter materials has confirmed their efficiency in P removal. Comprehensive reviews of suitable filter materials are provided by Johansson Westholm (2006) and, more recently, by Vohla et al. (2011). Some mineral-based and alkaline filter materials have been tested in field trials with real-life wastewater. A few have shown promising results and are now commercially available in Sweden (Renman and Renman, 2010), Norway (Jenssen et al., 2010) and the US (Drizo, 2012), where they are used in small-scale treatment facilities serving single houses or groups of houses.

The main purpose of using alkaline filter materials is to remove P from wastewater to concentrations that comply with statutory effluent criteria such as those applied in Sweden for on-site wastewater treatment (total P < 1 mg L⁻¹ or 70–90% removal) (SEPA, 2008). The alkaline conditions (pH 8–12) also bring about a reduction in bacteria. Conventional soil infiltration systems use sand and rely on physical filtration, straining and adsorption as the main removal mechanisms, with the clogging zone near the infiltration surface playing an important role for bacteria removal (Stevik et al., 1999; Chabaud et al., 2006). In contrast to sand filters, most alkaline filter materials have a coarse, porous structure with high hydraulic conductivity and are operated under saturated flow. Therefore an efficient pre-treatment prior to filtration is suggested to be a necessity to avoid biofilm development and clogging. Organic load, including bacteria, could prevent the removal of P by the filter material, thus reducing its lifetime considerably. Laboratory experiments (Alvarez et al., 2004) have shown that the presence of organic ligands inhibits the precipitation of calcium phosphates, one of the principal mechanisms for P removal in the commercial products Polonite and blast furnace slag (BFS) (Gustafsson et al., 2008).

Contamination of groundwater by on-site wastewater containing pathogenic microorganisms, particularly enteric viruses, has long been reported (Craun, 1985). Contamination of surface water is also common and might involve a human health risk. It is therefore important that the wastewater treatment systems include the immobilization of microorganisms. Furthermore, since the filter materials are intended for recycling to crop production (Cucarella et al., 2012), a product with minimal bacteria content would be of great benefit. A suitable group of bacteria for use as an indicator of contamination are the faecal streptococci, in which the genus *Enterococcus* is a sub-group (Ausland et al. 2002). Enterococci are Gram-positive, facultative anaerobic bacteria that can grow in a wide range of temperatures and withstand freezing and high pH values (Fisher and Phillips, 2009). It is proposed in Sweden that Enterococci can be used as indicator organisms when returning P through wastewater treatment products back to agriculture (SEPA, 2002).

In Sweden, filter traps are constructed primarily for P removal and recycling (Cucarella, 2009; Renman and Renman, 2010). The operation of several full-scale systems shows that the gradual decline in pH that occurs over time, is correlated with a decrease in P removal efficiency. However, there is also

reason to suspect a corresponding effect of organic load, which can vary significantly depending on the pre-treatment system used. Hence it is important to investigate the effects of organic matter on the P removal process and how it affects filter longevity. Bird and Drizo (2010) showed that P removal efficiency and filter life were significantly higher in steel slag filters fed with agricultural effluent with lower organic matter content than domestic sewage.

The aim of this study was therefore to investigate the removal capacity of two filter materials for bacteria (i.e. Enterococci), total organic carbon (TOC) and P in relation to two different loads of organic matter. This was studied in a long-term (duration from 84 up to 136 days) dynamic column experiment using on-site wastewater under real-life treatment conditions.

2. Materials and methods

2.1. Materials

2.1.1. Filter materials – chemical and physical properties

Two different filter materials were used for the investigation, blast furnace slag (BFS) and Polonite (Table 1). The element analysis of the filter materials was performed by ALS Scandinavia AB, Luleå, Sweden, using simultaneous ICP-OES (ARL, model 3560, Thermo Scientific). Radiometer PHM 82 Standard pH-meter was used for determining pH in 100 mL of influent and effluent samples. The pH of filter materials was determined at 1:2.5 (w:V) soil:distilled water suspension after 24 h contact. Density of the materials was determined by the pycnometer method and porosity was calculated from the fluid porosity method. Measurement of the saturated hydraulic conductivity was performed using a permeameter according to the falling head method.

BFS is a by-product from the steel-making industry. Polonite is a manufactured product, prepared from the porous bedrock opoka. Both filter materials are characterised by high contents of calcium (Ca) and silicon (Si) (Table 1). See Renman (2008) for a more detailed description of the materials. For

Table 1 – Chemical and physical properties of blast furnace slag (BFS) and Polonite.

Parameter	BFS	Polonite
Si (g kg ⁻¹)	155	241
Al (g kg ⁻¹)	69.7	27
Ca (g kg ⁻¹)	216	245
Fe (g kg ⁻¹)	3.11	16.5
K (g kg ⁻¹)	6.18	9.15
Mg (g kg ⁻¹)	97.6	4.4
Mn (g kg ⁻¹)	4.69	0.12
Na (g kg ⁻¹)	4.28	1.46
P (g kg ⁻¹)	<0.1	0.34
pH	9.7–10.0	11.2–12.3
Density (g cm ⁻³)	2.2	0.8
Porosity (%)	28	43
Hydraulic conductivity (m day ⁻¹)	255	800
Particle size (mm)	0.5–4	2–6

this experiment, Polonite material was obtained from Biotech AB, Stockholm, Sweden. The BFS was obtained from Merox AB, Oxelösund, Sweden, 2 months prior to the experiment build-up.

2.1.2. Experimental setup

The column experiment was housed in a steel container with the internal dimensions $1.7 \times 1.8 \times 1.9$ m. The whole set-up was placed close to an on-site wastewater treatment facility (WWTF) serving four households in the village Brottbys 35 km from Stockholm, Sweden. The WWTF consists of a septic tank (volume 10 m^3) and a sequencing batch reactor (SBR). Wastewater is pumped from the septic tank to the SBR which is a fill-and-draw, non-steady state activated sludge process in which reactor basins are filled with wastewater during a discrete time period, and then operated in a batch mode. The experiment was divided into two parts, plastic cylinder columns (diameter 10.5 cm, height 67 cm) were used in both parts, with triplicate columns for each of the two filter materials studied. The columns were packed to a height of 50 cm, resulting in a total volume of material of 4 L. Packing was performed in the same way for all columns to minimise variation in permeability and porosity. However, the particle diameter of Polonite was 2–6 mm, while BFS particle diameter was 0.5–4 mm, resulting in pore volumes of 1.8 L and 1.1 L, respectively.

2.2. Methods

2.2.1. Column experiment

During the experiment, the pumping and loading of wastewater was controlled by a system of programmable switches, connected to magnetic valves and electric pumps. A very high dosing rate of wastewater was applied to mimic the most extreme conditions that can arise in a filter well. Three daily doses were applied, at 08:00, 12:00 and 18:00 h. The Polonite columns were fed at a total rate of 4.4 ± 0.1 pore volumes per day, while the BFS columns were fed at 4.5 ± 0.3 pore volumes per day, corresponding to an approximate wastewater load of 8 and 5 L, respectively. The wastewater was pumped in up-flow direction with a pumping rate of 60 mL min^{-1} .

In the first experiment (high organic loading), wastewater was pumped directly from the septic tank, without involving the SBR, to an infiltration trench with crushed bedrock intended for the removal of coarse particles. An underlying plastic liner and drainage pipes collected the pre-treated wastewater and pumped it to the column experiment. The experiment lasted for a total of 320 days, including a break during when the operation was discontinued due to freezing of the pipes feeding the experiment (December–March), and then restarted. The columns were kept saturated with wastewater and under non-frozen conditions during the break. The columns filled with BFS were shut down earlier than those filled with Polonite due to breakthrough in phosphate removal. Thus the columns with Polonite and BFS were active for a total of 119 and 84 days respectively. During this period, the Polonite columns received 8.8 g P (0.074 g day^{-1}), while the BFS columns received 6 g P (0.075 g day^{-1}).

For the second experiment (low organic loading), the columns were filled with fresh Polonite and BFS material and

wastewater was pumped to the columns after treatment in the existing SBR treatment plant. Chemical P removal was omitted during the experiment, but the treatment decreased the BOD_7 value. The experiment lasted for a total of 196 days and the columns with Polonite were active throughout this period. The BFS columns were shut down after 116 days due to breakthrough in phosphate removal but kept saturated and restarted again after a further 60 days, i.e. they were active for a total of 136 days.

In the second experiment, the Polonite columns received a total P load of 7.1 g (0.036 g day^{-1}), while BFS slag received 3.8 g and 0.7 g in the first and second period (0.033 and 0.035 g day^{-1}), respectively. The chemical composition of the wastewater used in both experiments is shown in Table 2.

2.2.2. Phosphorus, TOC, bacteria and BOD – sampling and analysis

Samples for all analyses were taken weekly or bi-weekly during the afternoon pumping event. The pH was measured directly after sampling, using a pH meter of model PHM 95, Radiometer, Copenhagen, all samples were frozen ($-18 \text{ }^\circ\text{C}$) and defrosted prior to analysis.

For analysis of carbon in the incoming wastewater and in the effluents, the samples were defrosted at room temperature and left to rest for the particles to settle. Samples were taken from the liquid and diluted 1- to 5-fold, to a total volume of 5–6 mL. Analysis of total organic carbon (TOC) was performed according to the Combustion-Infrared method, e.g. total carbon in the samples was oxidised into CO_2 , which was subsequently measured. Following this, a second measurement was carried out where only the inorganic carbon was oxidised and measured. Total organic carbon was then calculated as the difference between the total carbon and inorganic carbon. The analysis was performed using a total organic carbon analyser; model TOC-5000 (Shimadzu Corporation, Japan).

After analysis, all samples from the same month were divided into three different groups based on their origin (influent water, effluent from BFS and effluent from Polonite). All samples within the same group were statistically tested for outliers using Grubb's test (GraphPad, 2012), and any outliers detected were removed. A mean value for each month was calculated from the results of the incoming samples. These

Table 2 – Composition of the wastewater used in the column experiments, mean values for the different parameters, with standard deviation in brackets. Number of colony forming units (CFU) given as median values. n = number of samples analyzed for the different parameters in the experiments with high (Exp I) and low (Exp II) biological oxygen demand (BOD) concentrations.

Parameter	n (Exp I)	n (Exp II)	Exp I	Exp II
BOD_7 (mg L^{-1})	5	5	120 (± 11)	20 (± 5)
pH	16	19	7.8 (± 0.4)	7.9 (± 0.4)
Total phosphorus (mg L^{-1})	17	20	8.0 (± 1.7)	5.3 (± 2.6)
TOC (mg L^{-1})	12	14	132 (± 41)	43 (± 9)
Bacteria (CFU mL^{-1})	18	21	11,200	2000

mean values were then used, combined with the individual values from the effluents, to calculate the percentage reduction in TOC in the water after passage through the materials, followed by statistical calculations of the results (described in Section 2.2.3).

Analysis of P in the form of orthophosphate and total phosphorus was performed using Flow Injection Analysis (FIA; Aquatec-Tecator, Sweden). The percentage removal was calculated as the difference between the concentration in the effluent and the concentration in the influent.

In order to determine the reduction in Enterococci in the filter materials, culture and counting of colony forming units (CFU) on specific nutritious agar was performed. In brief, the frozen samples were defrosted at room temperature and pooled, so that all samples originating from the same column and month were merged together into one single sample by mixing 2 mL of each sample in a Falcon test tube, resulting in one bulk sample per specific column and month. Similarly, all samples from the incoming wastewater from the same month were merged together to one bulk sample. All samples were mixed thoroughly by inverting the test tubes. The samples originating from months 2, 3 and 4 of Exp I (high organic loading) were diluted 10-, 100- and 100-fold, respectively (prior to the first cultivation, screening was performed to determine the appropriate dilution and volume). The remaining samples were not diluted. Between 100 and 500 μL of liquid were spread on triplicate plates containing Bile Esculin Agar (manufactured by FLUKA, Sigma–Aldrich). The plates were left to rest for 30 min before being inverted and incubated at 35 °C for 24 h, after which black colonies were counted. The results were recalculated as CFU 100 mL⁻¹.

The mean value for the concentration of Enterococci in incoming water was calculated for each month, based on the results from the incoming samples. These values were then compared against the individual results from the effluents from the same month and the percentage reduction was calculated, after which statistical tests were performed as described in Section 2.2.3.

The overall average TOC, phosphorus and bacteria removal rates for the different materials and organic loadings for the whole experimental period were calculated based on the percentage removal in the individual samples.

Samples of influent and treated wastewater were taken five times during the experiments in 500-mL plastic bottles for BOD₇ analysis. These samples were analysed by the accredited laboratory ALS Scandinavia AB, Täby, Sweden.

2.2.3. Statistical calculations

In order to determine the effect of filter material and concentration of organic material on the removal of phosphorus, bacteria and organic matter, one-way ANOVA was used to test for differences between the set-ups. When differences were detected, Tukey post hoc tests were performed in order to determine the nature and magnitude of these differences. Regression analysis was performed on P and bacteria reduction in order to determine the correlation of these parameters to organic matter and pH. The statistical software used for this was SPSS Statistics 19 (IBM, 2012).

3. Results and discussion

3.1. TOC removal

The change in removal efficiency of TOC by the filter materials over time when loaded with high and low BOD₇ (hereafter BOD) concentrations is shown in Fig. 1. At high concentrations of BOD (120 mg L⁻¹) the monthly average removal of TOC in Polonite ranged between -21.0 and 89.2%, whereas in BFS it ranged from -16.7 and 56.9%. At low BOD concentration (20 mg L⁻¹), the monthly average TOC removal ranged from 11.2 to 82.4% in Polonite and from 0.4 to 31.2% in BFS. The total average removal of TOC when loaded with the high concentration of BOD was 30% in Polonite and 21% in BFS, while when loaded with the low concentration of BOD the total average removal was 46% in Polonite and 19% in BFS. As seen in Fig. 1, the total average TOC removal is not consistent with an average value of the monthly removal rates. This dissimilarity is caused by the variation in number of samples from the different months. The monthly removal rates provide information regarding the change over time whereas the total removal is better represented by an average value based on all of the samples from the whole experimental period.

Statistical analysis using one-way ANOVA showed that Polonite fed with low concentration of BOD had significantly higher removal of TOC than BFS fed with high ($p < 0.05$) and low ($p < 0.01$) concentrations of BOD. No significant difference in TOC removal was observed between high and low concentrations of BOD in Polonite.

The relationship between TOC removal and pH for Polonite and BFS is shown in Fig. 2a and b, respectively. A significant correlation between increasing pH and TOC removal was found for Polonite (Fig. 2a) at low concentration of BOD ($p < 0.01$). This is in agreement with observations by Song et al. (2006) who reported an increase in the removal of humic substances with increasing pH. When using high concentrations of BOD, it is likely that accumulation of organic matter affected the pH. The release of CO₂ during organic matter degradation is suggested to increase the

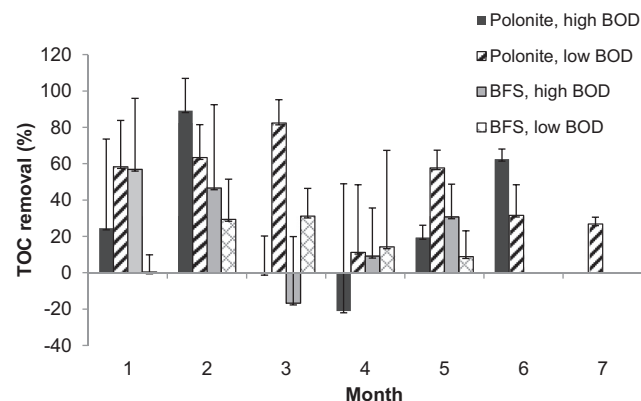


Fig. 1 – TOC removal vs time in Polonite and BFS columns fed with wastewater with high and low concentrations of BOD. Error bars are standard deviation calculated among the individual samples. Missing bar in the chart = no data available.

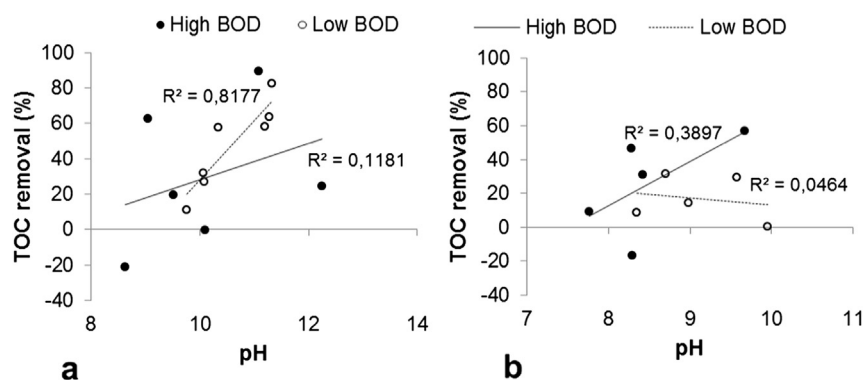


Fig. 2 – Effect of pH on TOC removal capacity. The linear regression represents monthly average pH and removal of TOC vs time in Polonite (a) and BFS (b).

transformation of reactive CaO in the filter materials to less reactive CaCO₃, which is particular the case for the strongly alkaline Polonite. With time, the accumulated organic matter would also cause clogging and preferential flow in the filter thereby causing less interaction with the material, leading to a less TOC removal efficiency. This is further supported by the correlation seen between the removal of P and TOC in Polonite when the filter was fed with wastewater of high BOD concentration ($p < 0.01$).

For the BFS filter (Fig. 2b), a significant correlation was only observed between pH and TOC removal when using wastewater with high concentration of BOD ($p < 0.05$). According to McDowell-Boyer et al. (1986) clogging increases the possibility of straining within porous media, such as BFS in the present case. Organic matter removal in terms of TOC should then increase with time or at least reach a steady state, which was not observed in this experiment. Instead a slightly decline in TOC removal for both filter materials (as seen in Fig. 1) indicates co-precipitation of organic matter with Ca released from the filter material in decreasing amounts with time (cf. Wendling et al., 2012). The pH decline could offer opportunities for biofilm development in a late stage of the filter operation.

Some of the samples from the present study showed negative values of TOC removal, possibly due to release of organic matter, including dead bacteria. This supports findings by Wendling et al. (2012), who attributed the release of DOC in sand columns to the breakdown and release of loosely bound organic matter inside the columns.

3.2. Phosphorus removal

Total phosphorus removal over time in the columns fed a high and low concentration of BOD is shown in Fig. 3. At the high BOD concentration (120 mg L⁻¹), the mean monthly P removal ranged between 47 and 97% in Polonite and -8 and 71% in BFS, with a mean total reduction of 76% and 22% in Polonite and BFS, respectively. With the low BOD concentration (20 mg L⁻¹) the monthly average reduction in P ranged from 76 to 97% in Polonite and -27–77% in BFS, with mean total reduction of 93% in Polonite and 18% in BFS. As seen in Fig. 3, the total average phosphorus removal is not consistent with an

average value of the monthly removal rates. This inconsistency is caused by the same variation in number of samples as discussed in Section 3.1 (Fig. 4).

Statistical evaluation of the results using ANOVA confirmed that the setup with highest removal of P was Polonite fed with low concentration of BOD, followed by Polonite fed with high concentrations. No significant difference could be seen in the performance of BFS when comparing the performance at conditions of high and low BOD concentrations ($p = 0.921$).

The pH in Polonite was consistently higher, decreasing from 12.3 to 9 when using high BOD concentrations and from 11.2 to 10.1 using low BOD concentrations. The corresponding pH values for BFS when using high and low BOD concentrations were from 9.7 to 8.4 and from 10.0 to 8.3 respectively.

The P removal performance of Polonite fed with the low concentrations of BOD in this study (93%) is in accordance with previous studies, showing 97% P removal in Polonite from a synthetic solution (Gustafsson et al., 2008; Renman and Renman, 2010) and wastewater (Renman and Renman, 2010). Renman and Renman (2010) also reported a removal rate of 87% when using Polonite in a filter bed treating the wastewater from a single household. This value is somewhat higher

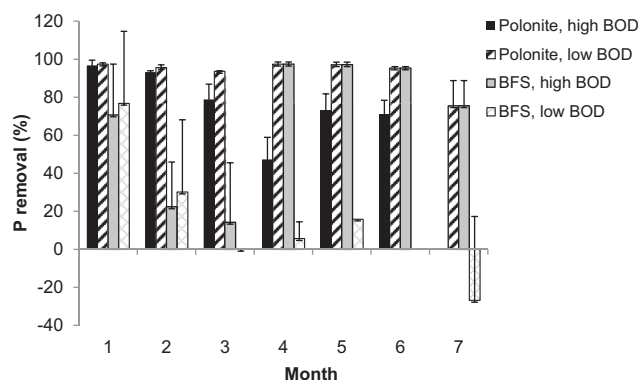


Fig. 3 – Phosphorus removal vs time in Polonite and BFS columns fed with wastewater with high and low concentrations of BOD. Error bars are standard deviation calculated among the individual samples. Missing bar in the chart = no data available.

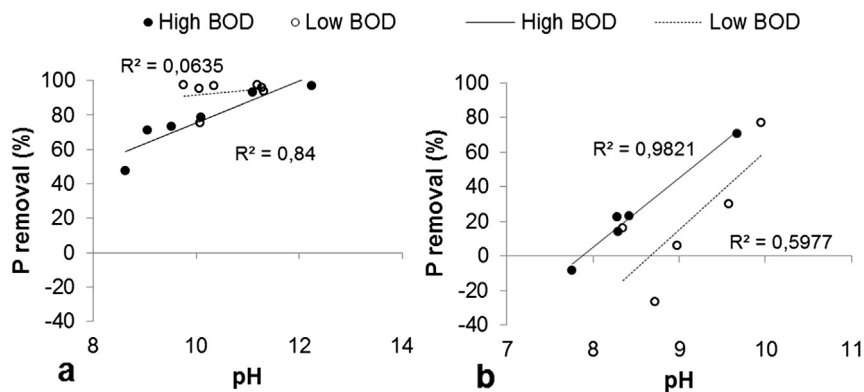


Fig. 4 – Effect of pH on phosphorus removal capacity. The linear regression represents monthly average pH and removal of P vs time in Polonite (a) and BFS (b).

than the removal rate of 76% seen when treating wastewater with a high concentration of BOD (120 mg L^{-1}) in the present study. The difference in P reduction seen in Polonite is further supported by the results reported by Renman and Renman (2010). They ascribed the similar removal rate from synthetic solution and wastewater to the lower water load in the columns fed with wastewater, thus indicating decreased P removal capacity when treating wastewater compared with a synthetic solution. In the present study, P removal rate in BFS using wastewater with low (20 mg L^{-1}) and high (120 mg L^{-1}) concentrations of BOD was lower than reported in previous studies; This can probably be attributed to the unusually short residence time of 0.22 days compared to other studies where hydraulic retention times (HRT) up to 24 h was applied (e.g. Grünberg and Kern, 2001; Oğuz, 2004). The results from this study therefore indicate that BFS material may not be as efficient at short HRT (5.5 h) as at longer HRT (24 h). Furthermore, the initial pH of the slag used in the present study had an average value of 10.0 and 9.7 when using high and low concentrations of organic matter respectively. These values are lower compared to those reported from previous experiments (Johansson, 1999; Gustafsson et al., 2008), which could pose an additional reason for the poorer performance observed here. The different starting pH values of the filter materials can most likely be attributed to the fact that the material were not delivered fresh or was the result of any unknown factor in the production. Both materials showed increased P removal capacity after month 4 (Fig. 3), when the system was in a resting period. When comparing the two months prior to the stop with the two months immediately after, the P removal increase showed no significance for Polonite ($p = 0.564$) as well as for BFS ($p = 0.551$). Regeneration of phosphorus retention sites was expected as demonstrated by previous investigations of metallurgic slags (Drizo et al., 2002).

The difference between the two materials in terms of P removal capacity was confirmed to be significant ($p < 0.01$) by the ANOVA tests. The ANOVA analysis also revealed that Polonite performed better when fed with low concentrations of BOD ($p < 0.05$). This correlation was further established with linear regression, which showed a positive effect of 29.3% on P removal by lowering the BOD concentration.

Furthermore, linear regression analysis showed a positive correlation between removal of TOC and P in Polonite and BFS at high concentrations of BOD ($p < 0.01$). The relationship between pH and phosphorus removal in Polonite and BFS is shown in Fig. 5a and b respectively. A significant correlation between increasing pH and P removal was found for Polonite ($p < 0.01$) and BFS (0.05) at high BOD. No correlations were found between P removal and pH or TOC removal in BFS or Polonite loaded with the low concentration of BOD. The lack of correlation between pH and removal of phosphate in Polonite fed the low BOD concentrations is not consistent with previous work using Polonite, where correlations were obtained using synthetic P solution and in full scale systems treating wastewater (Renman and Renman, 2010). However, in their study the pH of the effluent decreased to 8.2, as compared to 10.1 when using low BOD concentrations in the present study. One possible explanation for the lack of correlation between

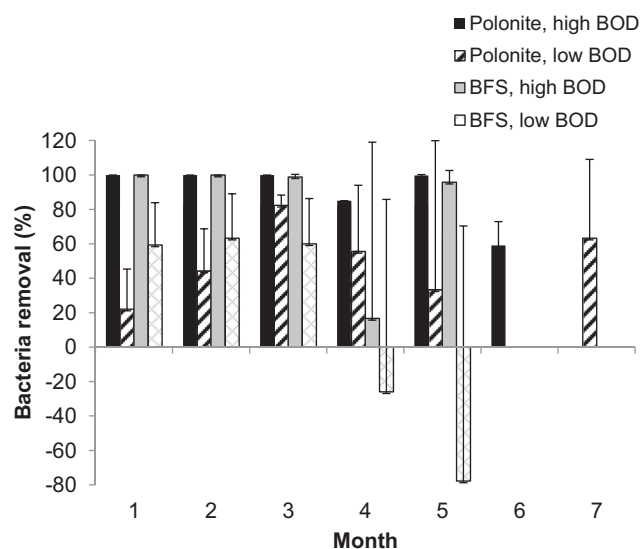


Fig. 5 – Reduction of Enterococci vs time in Polonite and BFS materials fed with wastewater with high and low concentrations of BOD. Error bars are standard deviation calculated among the individual samples. Missing bar in the chart = no data available.

pH and phosphorus removal seen in this study would therefore be an insufficient decrease in pH. It is possible that such a correlation could have been distinguished if the experimental period would have been extended and the pH thereby allowed to further decrease.

From our previous experiments (unpubl. data) it is apparent that adsorption reactions are not wholly responsible for the disappearance of P from wastewater being in contact with reactive filter materials. Rather, the mechanism appears to be solid phase precipitation of various calcium phosphates which is supported by studies of Gustafsson et al. (2008) and Everbom et al. (2009).

Analysis of phosphate removal (results not shown) revealed the same pattern as seen for total phosphorus. Linear regression showed a stronger correlation between reduced BOD concentration and increased removal of phosphate (compared with total P), with a 9.5% increase in phosphate removal for BFS and a 44.5% increase for Polonite.

3.3. Bacteria removal

The change in bacteria removal over time in columns loaded with the high and low BOD concentration is shown in Fig. 5. When loaded with the high concentration of BOD (120 mg L^{-1}) the mean monthly removal of Enterococci ranged from 59 to 100% in Polonite and from 17 to 100% in BFS, with a mean total removal of 91% in Polonite and 81% in BFS. At the low BOD concentration (20 mg L^{-1}) Polonite had a mean monthly removal of Enterococci ranging from 22 to 93% whereas in BFS it ranged from -78 to 63%, with the total removal in Polonite and BFS averaging 52% and 16% respectively. As seen in Fig. 5, the total average removal of Enterococci is not consistent with an average value of the monthly removal rates. This inconsistency is caused by the same variation in number of samples as discussed in Section 3.1.

The difference in bacteria removal between the two concentrations of BOD was established with linear regression analysis, which showed that the decrease in BOD concentration caused a decrease in bacteria removal of 19% in Polonite and 12.6% in BFS ($p < 0.01$). This difference was confirmed to be significant using ANOVA ($p < 0.05$ for Polonite, $p < 0.01$ for BFS). The ANOVA test did not reveal any significant difference in bacteria removal between Polonite and BFS fed with high concentrations of BOD ($p = 0.861$). However, when using low concentrations of BOD, Polonite performed better compared to BFS ($p < 0.05$). The bacteria removal efficiencies in both materials fed the high concentration of BOD is in agreement with previous studies on Polonite and BFS (Renman et al., 2003), as well as other materials (Stevik et al., 1999). However, the removal rates in columns loaded with the low concentration of BOD were considerable lower. According to Stevik et al. (2004), the mechanisms involved in immobilisation of bacteria in wastewater are straining and adsorption. Straining constitutes physical blocking of movement through media and the determining factor is the ratio of the diameter of the pollutants to that of the media particles. The ratio between the small diameter of Enterococci and the large diameter of both Polonite and BFS particles is too large for straining to be the mechanism responsible for retaining bacteria, with clogging zones as possible exceptions (McDowell-Boyer et al.,

1986), leaving adsorption the more probable answer. Bacterial adsorption to material is dependent on the initial bacterial concentration and the contact time between the bacteria and the material and is affected by several different parameters, such as pore size, organic matter, biofilm, ionic strength and pH (Stevik et al., 2004). Since the adsorption of bacteria is dependent on the bacteria concentration, the higher removal of bacteria at high BOD concentrations in the present study was most likely caused by a higher degree of adsorption due to higher concentration of bacteria in that wastewater. It is also possible that when feeding the columns with the higher BOD concentrations, a biofilm formed which would decrease the pore size in the material and thereby enhance the adsorption (Stevik et al., 1999). This theory is further supported by the reported aggregation of bacteria to solids and colloidal particles, which may lead to decreased permeability of the filter (McDowell-Boyer et al. 1986). Furthermore, Polonite showed consistently higher pH compared with BFS for both organic loadings, which is in agreement with results reported by Gustafsson et al. (2008). Enterococci can grow at very high pH (Fisher and Phillips, 2009), it is therefore likely for the higher pH in Polonite compared with BFS to have played a key role in the higher bacteria removal observed in the present study.

Multivariate regression analysis of the results from the present study showed no correlation between the removal of TOC and bacteria in either of the materials or at either BOD concentration. However, the results did show that pH significantly affected removal of bacteria by Polonite loaded with high concentration of BOD ($p < 0.01$) and by BFS loaded with high and low concentrations of BOD ($p < 0.05$). No correlation was seen for Polonite loaded with the low concentration of organic material, which may be ascribed to the greater pore size in Polonite compared with BFS. When passing through the Polonite, the wastewater is more likely to flow through the larger pores, leading to decreased contact time between material and bacteria. In addition, the distance between material and bacteria is greater, resulting in decreased adhesion (Stevik et al., 2004). In BFS, on the other hand, the pores are much smaller, leading to more intimate contact between the bacteria and the material and enabling enhanced bacteria adsorption. Furthermore, the results from the ANOVA suggested the following order of bacteria removal: Polonite loaded with a high concentration of organic material > BFS loaded with a high concentration of organic material > Polonite loaded with a low concentration of organic material > BFS loaded with a low concentration of organic material. This order of reduction agrees with the results presented in Fig. 5. See also Table 3 for a summary of the results.

In the present study, all of the columns except for Polonite fed with a low BOD concentration (20 mg L^{-1}) showed a reduction that spanned over negative values, i.e. for some of the samples for these months there was a higher concentration of Enterococci in the effluent than in the corresponding influent. One possible explanation for this might be that samples from the influent and effluent were taken at the same time and thus did not originate from the same volume of water. Therefore, it is possible that water with a higher concentration of bacteria had passed through the column prior to sampling, which would lead to a higher concentration in the effluent too. Comparing the effluents with an influent with a

Table 3 – Summary of main experimental parameters and main results and comparison with previous studies. Both organic loadings are reported (high organic load/low organic load).

Parameter	Filter material		Comment/comparison
	BFS	Polonite	
Type of filter material	By-product	Natural, processed	See reviews by Johansson-Westholm (2006) and Vohla et al. (2011).
Duration of column experiment (days)	84/136	119/196	Long operation period under real treatment conditions.
Particle size distribution d_{50} (mm)	2/2	4/4	Various particle sizes were used (0–19 mm), see Cucarella and Renman (2009).
Hydraulic retention time, HRT (h)	5.3/5.3	5.5/5.5	HRT of 1–72 h was used in previous column and pilot scale experiments (Renman and Renman, 2010).
Wastewater composition (BOD ₇ , mg L ⁻¹)	120/20	120/20	Normal BOD values for domestic raw and pretreated wastewater.
Effluent pH range	8.4–9.7/8.3–10	9–12.3/10–11.2	Solution pH is a vital factor influencing the sorption behaviour (Gustafsson et al., 2008; Xue et al., 2009)
P removal (%)	22/18	76/93	Synthetic P-solution increase removal efficiency for BFS (Gustafsson et al., 2008).
TOC removal (%)	21/19	30/46	pH dependent removal (cf. Song et al., 2006)
Bacteria removal (%)	81/16	91/52	Influence of straining (BFS) and pH.

lower concentration of bacteria would therefore give a negative value for bacteria removal. Another possible explanation is that bacteria accumulated inside the columns and were flushed out at a later stage of the experiment. This would cause the effluent to contain the previously accumulated bacteria and therefore result in a higher amount of bacteria in the effluent compared with the influent. It is also important to keep in mind that for the experiment with low BOD concentration, the water had been subjected to a pre-treatment process that reduced the content of organic matter, as well as bacteria. Cultivation of bacteria in these samples gave rise to small colony counts, in several cases zero or only a few, both in the effluents and in the influent. All of the samples had also been frozen, which is likely to have reduced the amount of bacteria even further. This is supported by Gao et al. (2009), who found the cell population of Enterococci in water samples to be reduced by around 90% by freezing at between -7 and -30 °C. However, since the influent and effluent samples have been treated the same way, it is probable that the effect of freezing would be similar and the percentage removal rates presented herein should therefore be valid. A low amount of colonies is likely to lead to an increased influence of randomness, so these results should only be regarded as an indication of the effect of the filter materials tested on bacteria removal.

The order of performance in terms of removal of bacteria and P in this study was not coherent. This is most likely due to the difference in bacteria content in the influent with high and low BOD concentration. The amount of Enterococci in the effluent from the high BOD columns was around 6500 and 3500 CFU 100 mL⁻¹ for Polonite and BFS, respectively, while in the effluent from the low BOD columns it was around 250 and 150 CFU 100 mL⁻¹ for Polonite and BFS, respectively. From this comparison, it is clear that even though the percentage removal was higher with the high BOD loading, the pre-treated wastewater used in the low BOD treatment was the better option in respect of removal of Enterococci.

4. Conclusions

This study shows that controlling the concentration of organic material in incoming wastewater is important in achieving a good P removal in treatment using Polonite as a filter material at short HRT. The Polonite material showed increased P removal when loaded with pre-treated wastewater with a lower BOD concentration, whereas for BFS no such correlation was found. The order of P removal in the materials was: Polonite + low BOD > Polonite + high BOD > BFS + high BOD > BFS + low BOD. The results from this study did not show any significant relationship between pH and removal of P when using Polonite fed with low concentrations of BOD.

With regard to reduction in total organic carbon (TOC), Polonite fed with a low concentration of BOD (120 mg L⁻¹) showed higher average removal than the other three treatments. A higher bacteria removal was achieved with a high concentration of BOD in both filter materials, probably due to the higher concentration of bacteria in the wastewater.

Filter particle size, hydraulic retention time and organic loading rate may play an important role in the P and TOC removal efficiency of filter materials. In all aspects examined in this study (removal of P, Enterococci and TOC), Polonite had superior performance compared to BFS. In order to achieve the best possible reduction in P, pre-treatment to reduce the concentration of organic material is of great importance.

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