Sveriges lantbruksuniversitet
Swedish University of Agricultural Sciences

Havs och Vatten myndigheten

Institutionen för energi och teknik

Capacity of Biochar Filters for Onsite Wastewater Treatment: Phosphorus and Nitrogen Removal – Technical Report

Biokol i små avloppsanläggningar: rening av övergödande ämnen– teknisk rapport

Sahar S. Dalahmeh

Institutionen för energi och teknik
Sveriges lantbruksuniversitet
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Sahar S. Dalahmeh
Sahar.Dalahmeh@slu.se

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Keywords: Biochar filters; impregnated biochar; Hydrus modelling; nitrogen removal; phosphorus removal; two-stage filters.

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SAMMANFATTNING

Cirka 15% av Sveriges befolkning bor i områden utanför tätorter där fastigheterna inte är anslutna till kommunala ledningsnät. Där sker reningen av deras avlopp i enskilda och småskaliga avloppssystem. Slamavskiljare och infiltration eller markbädd är det vanligaste teknikerna för småskalig avloppsrening i Sverige. Reningssteg för fosfor saknas helt i de flesta anläggningarna. De flesta anläggningarna som används idag byggdes på 80-talet och många av dem har börjat tappa funktionen och renar avloppsvattnet allt sämre. Utsläppen från enskilda anläggningar bidrar till förorenat grundvatten samt övergödning i hav, sjöar och vattendrag. Detta innebär att små och enskilda avloppsanläggningar kopplades till 15% av befolkningen står för 15% av den antropogena nettobelastningen av fosfor medan kommunala reningsverk som renar avloppsvattnet från 85% av Sveriges befolkning står för 18 % av denna belastning (Hav- och vattenmyndigheten, 2016).

Enligt Havs- och vattenmyndighetens allmänna råd (HVMFS 2016:17) om små avloppsanläggningar för hushållsspillvatten, bör reningen av totalfosfor (Tot-P) nå minst 70% för områden med normal miljöskyddsnivå och 90% för områden identifierade som hög nivå. Vidare bör reduktionen av organiskt material, BOD_7, vara minst 90% för alla anläggningar (Havs- och vattenmyndighetens, 2016). Dessutom bör reningen av totalkväve (Tot-N) vara minst 50% i områden som är känsliga för kväve. Lösningar för att förbättra fosfor- och kvävereningen i små enskilda anläggningar samt för att nå Havs- och vattenmyndighetens allmänna råd för små avlopp finns på marknaden men många har visat sig ha brister i rening och robusthet.

Biokol är pyrolyserat organiskt material som kännetecknas av hög porositet och stor specifik yta med många ytaktiva bindningsplatser. Biokol har låg vikt och är lätt att transportera. Behandling av avloppsvatten med biokol bygger på fysiska och biologiska processer, d.v.s. adsorption samt biologisk nedbrytning och omvandling av organiska föroreningar och kväve. Biokolets stora specifika yta och höga andel av mikro- och makroporer möjliggör effektiv avskiljning av partiklar, adsorption av organiska och oorganiska ämnen och påväxt av biofilm för biologisk nedbrytning.

I flera projekt vid Institutionen för energi och teknik vid Sveriges lantbruksuniversitet genomfördes ett antal försök som syftade till att utveckla biokol som filtermaterial för att förbättra reningen av olika föroreningar i avloppsvatten t.ex. kemisk syreförbrukning (COD), ammoniumkväve (NH_4-N), nitratkväve (NO_3-N) och totalkväve (Tot-N). I projektets första försök undersöktes reduktion av fosfatfosfor (PO_4-P) och totalfosfor (Tot-P) i biokol impregnerat med: 1) kalk, 2) järn eller 3) biokol blandat med polonit. Biokolet var gjort av tallbark som först impregnerades med kalciumoxid eller järnklorid och sedan pyrolyserdes i 3-4 timmar vid 350-400°C. Kalciumoxid och järnklorid är två kemikalier som används för fosforbindningen. Ett tredje biokol blandades med det fosforbindande materialet polonit, som innehåller mycket kalk. De impregnerade biokolen och polonitkolet jämfördes med

obehandlat biokol av tallbark. Biokolen packades i kolonner och belastades med 50 L m^{-2} $dygn^{-1}$ riktigt avloppsvatten från Kungsängsverket i Uppsala. Detta försök pågick under ett år.

I ett annat försök studerades biokolets kapacitet att rena avloppsvatten från ammonium (NH_4-N), nitrat (NO_3-N) och totalkväve (Tot-N) i ett tvåstegsbiokolsfilter; ett steg med vertikalt flöde följt av ett vattenfyllt steg med horisontellt flöde. Detta skapade ett filter med en syresatt del följt av en syrefattig del, vilket är gynnsamt för de bakterier som renar bort kväve. Tvåstegsbiokolsfiltret belastades med 23 L m^{-2} $dygn^{-1}$ riktigt avloppsvatten ett drygt år.

I det tredje försöket simulerades biokolets hydrauliska beteende vid avloppsvattenrening samt dess transport och rening av COD, fosfor och kväve med hjälp av HYDRUS-mjukvara kopplad till ”Constructed wetland module 3D” (CW3D). De simulerade resultaten jämfördes med observationer från laboratoriefilter av aktivt biokol som belastades med artificiellt avloppsvatten. Aktivt biokol (kallas också aktivt kol) är biokol som behandlats med syre för att höja porositeten och den specifika ytan hos materialet. Filtermaterialens hydrauliska egenskaper bestämdes experimentellt och genom invers (baklänges-) simulering med hydrauliska belastningar på 32 och 64 L m^{-2} $dygn^{-1}$. Concentrationerna av COD i utflödet från filtrer simulerades för olika belastningar (32 och 64 L m^{-2} dag^{-1}, 13-16 och 28 g BOD_5 m^{-2} dag^{-1}).

Studien av impregnerat biokol visade att det kalciumoxid- och järnkloridimpregnerade biokolet gav högst fosforreduktion, mer än 90 %. Inget av de två impregnerade biokolen visade tecken på minskad fosforreningsförmåga under studien. Det polonitblandade biokolet hade en fosforreduktion på ca 42 % medan det obehandlade biokolet renade bort ca 15 %. Både biokolsfiltret med polonit och det obehandlade biokolsfiltret tappade i effektivitet under försökets gång. Resultaten från försöket med kväverening i tvåstegsbiokolsfiltret visade på en reningsförmåga på 75 ± 12 % för Tot-N och 84 ± 10 % för NH_4-N . Steget med vertikalflöde i systemet var viktigt för nitrifikation (omvandling av ammonium till nitrit och nitrat) och 40 ± 26 % av Tot-N i inflödet omvandlades till nitrat. Den horisontella delen av systemet var viktigt för denitrifikation (omvandling av nitrit och nitrat till kvävgas eller lustgas) och mycket kväve renades bort (52 ± 41 % Tot-N) i den delen. Ammoniumreningen i tvåstegsbiokolsfiltret var effektiv (84 ± 10 % an NH_4-N). Mekanismerna för reningen var dels nitrifikation/denitrifikation och dels adsorption. Simuleringen av reningen av organiskt material (COD) visade på 94 % reduktion av COD vid belastningen 32 L m^{-2} dag^{-1} och 15 g BOD_5 m^{-2} dag^{-1} vilket stämde väl överens med uppmätta värden på 95 ± 2%. Däremot underskattade CW3D-modulen COD-reduktionen vid högre belastning och kväve- och fosforreduktionvid alla simulerade belastningar. Detta berodde på att simuleringen bara inkluderade bionedbrytning utan att inkludera fysisk adsorption av organiskt material, ammonium och fosfat.

Sammanfattningsvis kunde kalk- och järnimpregnering av biokol förbättra dess förmåga att binda fosfor, vilket innebär att impregnerat biokol passar väl som filtermaterial i små avloppsanläggningar för fosforrening. Trots att det kalk- och järnimpregnerade biokolet inte mättats under försöksperioden (ca 1 år) förväntas det mättas över tid och därmed minska dess fosforreningsförmåga. Biokolet kan kanske också mättas av andra föroreningar (t. ex organiskt material, ammoniak, läkemedel och spårmetaller). Därför rekommenderar vi att

impregnerat biokol används som en separat filtermodul för fosforrening. Denna bör komma efter huvudreningen (där organiskt material och ammonium renas från avloppsvattnet). Filtrets fosforrening vid olika hydrauliska belastningar undersöks just nu och resultaten kommer att redovisas i en kommande rapport.

Biokolsfilter i två steg med först vertikal- och sedan horisontallflöde var effektivt för reduktion av organiskt material och hade lovande prestanda för reduktion av total-kväve från avloppsvatten. Systemet har dock bara undersökts under fasta belastningsförhållanden. Systemets prestanda under varierade (dynamisk) belastning undersöks just nu och resultaten kommer att rapporteras i en kommande rapport. Filtersystemets utformning behöver dock undersökas och optimeras med avseende på djup, bredd och längd på det horisontella flödessystemet, liksom rekommenderade belastningsförhållanden.

Simuleringarna i Hydrus med CW3D-modulen visade lovande resultat vad gäller simulering av filter med biokol med avseende på hydrauliska egenskaper och reduktion av COD, men underskattade kväve- och forforreduktion i biokolfiltren. Adsorptionsförmågan hos biokol behöver inkluderas i modellen. Detta skulle förbättra simuleringarna och göra det möjligt att använda simuleringar för att optimera filterdjup, identifiera bästa belastningsförhållanden, utvärdera risken för igensättning av biokolet vid kontinuerlig belastning och därmed biokolets effektiva funktionstid.

INTRODUCTION

BACKGROUND

There are around 700 000 onsite wastewater treatment systems (OWTSs) in Sweden, half of which have a treatment process that goes beyond primary sedimentation. Most of the existing systems were constructed in the 1970s and 1980s (Ridderstolpe, 2009), and are now ageing and lack sufficient performance for pollution reduction. The phosphorus from onsite wastewater treatment systems in Sweden represents 15% of the total net anthropogenic load of total phosphorus (Tot-P) to the Baltic Sea, despite the fact that only 15% of the Swedish populations uses an OWTS (Hav- och vattenmyndigheten, 2016). This can be compared with the net anthropogenic load originating from wastewater treatment plants (WWTPs) treating wastewater from the remaining 85% of the population, which are responsible for only 18% of the Tot-P load (Hav- och vattenmyndigheten, 2016). Reducing Tot-P loads is a high priority for the Baltic sea region (Boesch et al., 2006; Hav- och vattenmyndigheten, 2016; Heinonen-Tanski & Matikka, 2017) and strict regulations on quality of the treated wastewater are applied to protect water bodies, especially sensitive waters. According to the Swedish Agency for Marine and Water Management (Havs- och vattenmyndigheten), the effluent from OWTS should achieve a minimum reduction of 90% for phosphorus in areas classified as sensitive to wastewater discharge and a minimum reduction of 70% at other sites (Havs- och vattenmyndigheten, 2016). Furthermore, the reduction is organic material should be at least 90% for all sites. To meet these regulations, efficient treatment with enhanced capacity for phosphorus removal is required.

A soil-based wastewater system is the most common form of OWTS used in Sweden. A fully functioning soil infiltration system has the capability to remove 30 ± 10 %, 70 ± 20 % and 80 ± 10 % of influent total nitrogen (Tot-N), Tot-P and chemical oxygen demand (COD), respectively (SMED, 2015). One problem with soil infiltration beds is that many existing systems have not been constructed according to the recommended guidelines. A common mistake is to locate the soil filter too close to the groundwater (<1 m). If the distance is too short, the water is not treated. Moreover, Tot-N and Tot-P removal show large variations in soil-based OWTSs, depending on soil, placement and load.

Natural sand is considered a fossil source and there is growing move to protect it from excavation and excessive exploitation. Thus there is a need to investigate another medium for onsite wastewater treatment as a complement or replacement for sand. One cost-effective treatment method for onsite wastewater treatment is use of biochar filters. Biochar is a material of organic origin charred at high temperatures (300-800 °C) in the absence of oxygen (Downie et al., 2009) and is characterised by large specific surface and high porosity. Forest residues and other organic waste products (e.g. agricultural waste and sewage sludge) can be utilised as a resource for biochar production, thereby contributing to sustainable management and re-utilisation of waste products (He et al., 2016). Previous studies have

demonstrated the efficiency of biochar in removing organic matter, surfactants and nitrogen from wastewater and greywater (Berger, 2012; Dalahmeh, 2016; Niwagaba et al., 2014). The main purpose of the biochar filters in those studies was to optimise the removal of organic matter, and thus little attention was paid to increasing the capacity of the biochar to remove phosphorus and nitrogen. Thus there is a need to investigate whether biochar filter bed material and bed structure can be modified to enhance removal of phosphorus and nitrogen from wastewater under conditions similar to those prevailing in onsite wastewater treatment systems.

AIMS AND OBJECTIVES

The overall aim of this report is to document and disseminate our latest findings in a project examining the potential of biochar filters for treatment of wastewater in onsite and small-scale systems, with special emphasis on removal of phosphorus and nitrogen. In the long term, the project is expected to contribute to the development and optimisation of biochar filters as a new technology for purification of wastewater in onsite, small systems, thus reducing the eutrophication of lakes, streams and coastal waters.

Specific objectives of this report are to:

1- Demonstrate and assess the performance of filters made of biochar impregnated with calcium oxide, iron chloride and Polonite in removal of phosphate-phosphorus (PO_4-P) and Tot-P from wastewater, and compare their performance with that of unimpregnated biochar.
2- Demonstrate and assess nitrification and denitrification in a two-stage biochar filter with combined vertical and horizontal flow regimes in the same bed.
3- Present and interpret results of computer simulations of flow dynamics and pollutant removal in activated biochar filters using Hydrus and CW3D simulation software.
4- Describe and assess the performance of household filters in removal of organic matter, nitrogen, phosphorus and *Escherichia coli* from a household biochar wastewater treatment system after three years of operation.

This remainder of this report is divided into four sections dealing with these four objectives. The first and second sections summarise our research findings regarding phosphorus and phosphate removal in impregnated biochar filters and nitrogen removal in combined vertical-horizontal flow biochar filters, respectively. The third section summarises the results of Hydrus and CW3D simulations on flow dynamics and pollutant removal in biochar filters. The fourth and final section describes current progress in a study examining the long-term performance of biochar filters constructed to serve single-family households.

PHOSPHORUS REMOVAL IN IMPREGNATED BIOCHAR FILTERS

BACKGROUND

In a soil infiltration system, the capacity to bind PO_4-P, and consequently Tot-P, depends on pH and on the availability of surfaces rich in aluminium (Al), iron (Fe) and calcium (Ca) (Arias et al., 2001; Hylander et al., 2006). The PO_4–P adsorbed to the surface of the material can also react with Fe, Al or Ca minerals to form strong precipitates or surface complexes. The pH in the medium affects the reaction. At low pH, PO_4–P reacts with Fe and Al more easily, forming e.g. $FePO_4 \cdot H_2O$. At higher pH, PO_4^{3+} forms complexes with Ca ions more easily, such as $CaHPO_4 \cdot 2H_2O$ and $Ca_4H(PO_4) \cdot 3H2O$ (US EPA, 2002).

The objective of this section of the report is to present our latest results regarding the potential of biochar impregnated with calcium oxide, iron chloride or Polonite in removing PO_4-P and Tot-P from wastewater and to compare their performance with that of unimpregnated biochar. Impregnated biochar filters have been developed for use as an advanced filtration step to be implemented after the biological treatment step (e.g. after a sand infiltration beds), in order to enhance the removal of phosphorus and phosphate from wastewater in onsite systems. In this section, we present and discuss the initial performance of freshly impregnated biochar filters in removing PO_4-P and Tot-P from wastewater during one year of operation. In addition, we describe the leaching of PO_4-P and Tot-P from impregnated biochar due to re-start of the filters after a pause in operation. This pause of operation was intended to mimic discontinuity of wastewater flows due to vacation by household members or technical problems leading to stops in filter operation. The impregnated biochar filters were assumed to be implemented as an advanced treatment stage, e.g. a treatment step that comes after the septic tank and soil infiltration system. Thus, the concentrations of PO_4-P and Tot-P tested in this project mimicked the concentrations of PO_4-P and Tot-P found in partially treated wastewater.

MATERIALS AND METHODS

Preparation of impregnation of biochar

Four forms of biochar were tested for PO_4-P and Tot-P removal. These were: (i) unimpregnated biochar (UBC); (ii) biochar impregnated with ferric chloride ($FeCl_3$; FBC); biochar impregnated with calcium oxide (CaO; CBC) and biochar mixed with Polonite (PBC). Impregnation of the biochar was performed according to the following method: Pine bark with particle size 1-5 mm was saturated with a solution of $FeCl_3$ (VWR, Stockholm) or CaO (VWR, Stockholm) before pyrolysis. The $FeCl_3$ and CaO had a purity of 97% and 95%, respectively, and the $FeCl_3$ or CaO to bark ratio was 0.3w/w. After being immersed in the solution for 24 hours at room temperature, the bark was dried at 100 °C for another 24

hours. Finally, the impregnated bark was pyrolysed in a muffle stove at 400 °C for 4 hours according to recommendations by Agrafioti et al. (2014). Unimpregnated biochar was produced by pyrolysis of the pine bark without impregnation. After pyrolysis, half the unimpregnated biochar was mixed with Polonite gravel with a Polonite/biochar ratio of 0.3. The chemical composition of the surfaces of the different biochar filters (CBC, FBC, PBC and UBC) was characterised using Fourier Transfer Infrared (FTIR). For this purpose, infrared absorption spectra of the impregnated and unimpregnated biochar were obtained using a Spectrum Two FTIR (Perkin-Elmer) equipped with an ATR Diamond accessory, which allows collection of FTIR spectra directly on a sample without any special preparation. The "pressure arm" of the instrument was used to apply a constant pressure (monitored by software) to the sample positioned on top of the Diamond crystal, to ensure good contact between the sample and the incident infrared beam. All FTIR spectra were collected at a spectrum resolution of 4 cm^{-1}, with 32 scans over the range from 4000 cm^{-1} to 450 cm^{-1}. The spectra obtained by ATR-FTIR were baseline-corrected and normalised.

Impregnated biochar filters – column experiment

The removal of PO_4-P and Tot-P from wastewater was investigated in UBC, CBC, FBC and PBC biochar filters during a one-year experiment. Four 60 cm high acrylic glass columns with 4.5 cm inner diameter were separately filled with the different biochars to form a filtration layer of 55 cm (n=1 for each type of biochar). Below and on the top of the filtration biochar layer, 5 cm coarse untreated biochar (8-15 mm particle size) was added to prevent clogging of the top of the filter and facilitate drainage at the bottom (Figure 1).

All the column filters were fed with real wastewater using a single pass and a down-flow regime at a hydraulic loading rate of 50 L m^{-2} day^{-1} over a period of six months. A rate of 50 L m^{-2} day^{-1} is a common load applied to sand filters used for onsite wastewater treatment. After six months, the operation of the columns was paused and then re-started. This pause and re-start of operation was planned to investigate leaching of the nutrients adsorbed on the biochar and to assess how strongly PO_4-P and Tot-P were bound to impregnated biochar.

The real wastewater with which the filters were fed was brought on a weekly basis from Uppsala wastewater treatment plant (Kungsängsverk) and refrigerated at 2-4 °C. Prior to each feeding, the wastewater was homogenised and then the required dose was pumped from the refrigerated wastewater container to distribution containers placed at room temperature (20 °C). When acclimatised to room temperature for 20 minutes, the wastewater was pumped from the distribution containers to the filters using a peristaltic pump. The filters were fed three times a day, at 24:00, 08:30 and 16:00, with 33% of the daily load on each occasion. Pumping and feeding of the filters were performed automatically and controlled by digital timers.

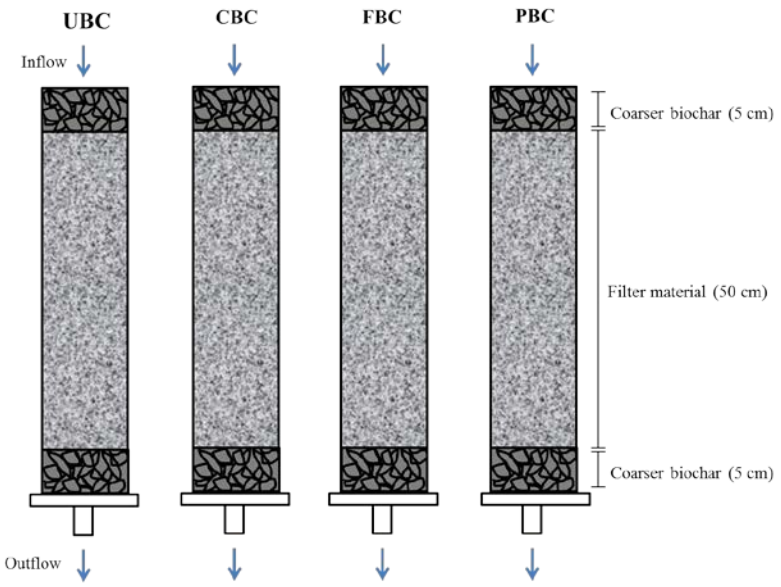

Figure 1. Experimental set-up used for column filters filled with unimpregnated biochar (UBC), biochar impregnated with calcium oxide (CBC), biochar impregnated with ferric chloride (FBC) and biochar mixed with Polonite (PBC). Figure is obtained from Stenström (2017).

Inflow and effluent from the filters were collected weekly and analysed for COD, PO_4-P and Tot-P using Spectroquant® cell kits number 14772-14773 (COD) and 14543 (PO_4-P and Tot-P) (Merck KGaA, Darmstadt, Germany). Prior to Tot-P determination, the samples were digested using Spectroquant® Crack set number 10 14687 **(**Merck KGaA, Darmstadt, Germany). Concentrations were determined colorimetrically using a Nova 60 photometer (Merck KGaA). Removal efficiency was calculated from the difference in concentration between inflow and outflow of the filters:

$$E = 100\ \frac{C_{in}-C_{out}}{C_{in}} \qquad (1)$$

where E is the removal efficiency (%), C_{in} is the concentration in the influent (mg L^{-1}) and C_{out} is the concentration in the effluent (mg L^{-1}).

RESULTS

Surface composition of impregnated biochar

The FTIR spectra of FBC, CBC, PBC and UBC are shown in Figure 2. The peaks observed in the region 1800-650 cm^{-1} in all spectra were attributed to carbonyl stretching vibrations and to -COOH symmetric stretching vibrations. The band observed around 780 cm^{-1} could be attributable to in-plane and out-of-plane C-H bends. The $FeCl_3$-impregnated (FBC) filter (freshly prepared and not yet used in wastewater treatment) caused only a slight change of the spectrum, mainly at around 650 cm^{-1}, where a new peak most probably attributable to iron oxalate was observed. In addition, there was a decrease in the peak at around 1020 cm^{-1}, probably as a result of shifting from COOH to COO^-.

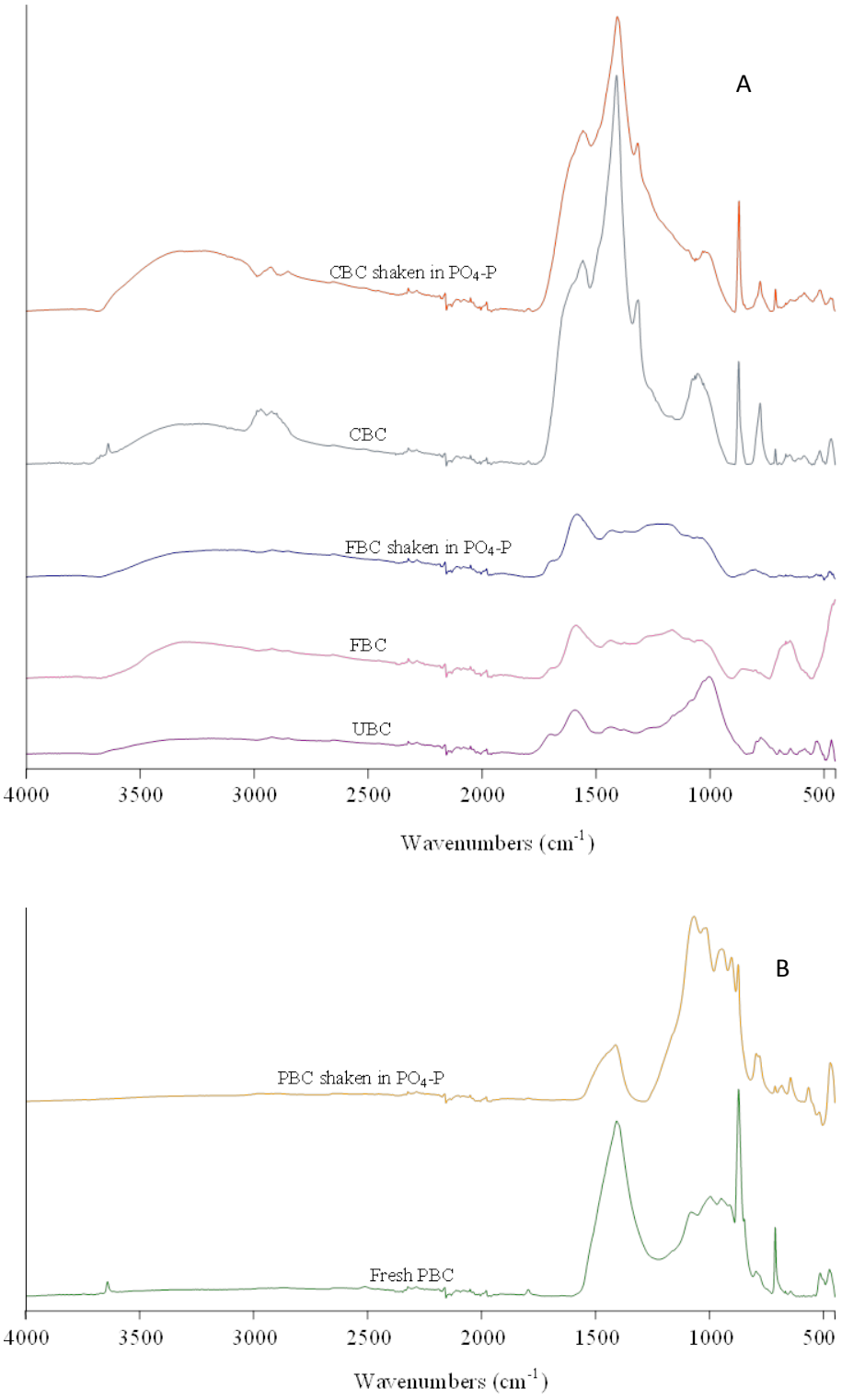

Figure 2. Fourier transform infrared (FTIR spectra) of: A) unimpregnated biochar (UBC, purple), biochar impregnated with calcium oxide (CBC, orange) and biochar impregnated with ferric chloride (FBC, blue); and B) biochar mixed with Polonite (PBC).

After using the FBC filter for Tot-P and PO_4-P adsorption, no significant changes were observed except disappearance of the peak at around 650 cm^{-1} characteristic of iron oxalate. The FTIR spectrum of the fresh CCB filter showed emergence of new characteristic absorption bands around 1410, 1315, 874, and 780 cm^{-1} which could be attributed to C-O and Ca-O bonds. After use of CBC for Tot-P and PO_4-P adsorption, a slight decrease in the intensity of the C-O and Ca-O peaks was observed. The FTIR spectrum of the fresh Polonite filter (PCB) exhibited strong absorption bands at 1410 and 870 cm^{-1}, which are the characteristic bands of carbonate anions, and other peaks around 1080, 980, 944 and 712 cm^{-1}, which were attributed to the vibrations of wollastonite (β-CaSiO3). A small peak observed at 3640 cm^{-1} was attributed to water vibrations (Figure 2). After the PCB was used for Tot-P and PO_4-P adsorption, a significant decrease in the carbonate anion band intensity was observed, suggesting formation of phosphate complexes. For untreated biochar (UBC), there were no significant peaks in the FTIR spectra.

Filter performance in organic matter removal

The pH in the influent wastewater was neutral, with pH 7.1 ± 0.4. The UBC filter effluent was just below neutral (pH 6.7 ± 0.5), while effluents from CBC and PBC had higher pH (7.8 ± 0.4). The largest pH change was observed for FBC, which had an average effluent pH of 2.6 ± 0.9 over the experimental period (Table 1).

The average COD concentration in the influent wastewater was 580 ± 360 mg L^{-1}. Initially, the effluents of FBC and CBC were turbid, but the turbidity disappeared after wash-out of the particles remaining after pyrolysis. However, CBC effluent had a yellow colour which persisted over the whole period of the experiment. After stabilisation of the filters, the average COD concentration in the effluent of UBC, FBC and PBC had effluent concentrations of 31 ± 25, 34 ± 14 and 32 ± 24 mg L^{-1}, respectively. The analysis showed that FBC removed on average 94 ± 6 % of the COD, UBC removed 93 ± 5 % and PBC removed 93 ± 4 % (Figure 3). The efficiency of COD removal in the CBC filter could not be determined due to the presence of the yellow colour, which resulted in high COD values (430 ± 230 mg L^{-1}) being measured in the CBC effluent.

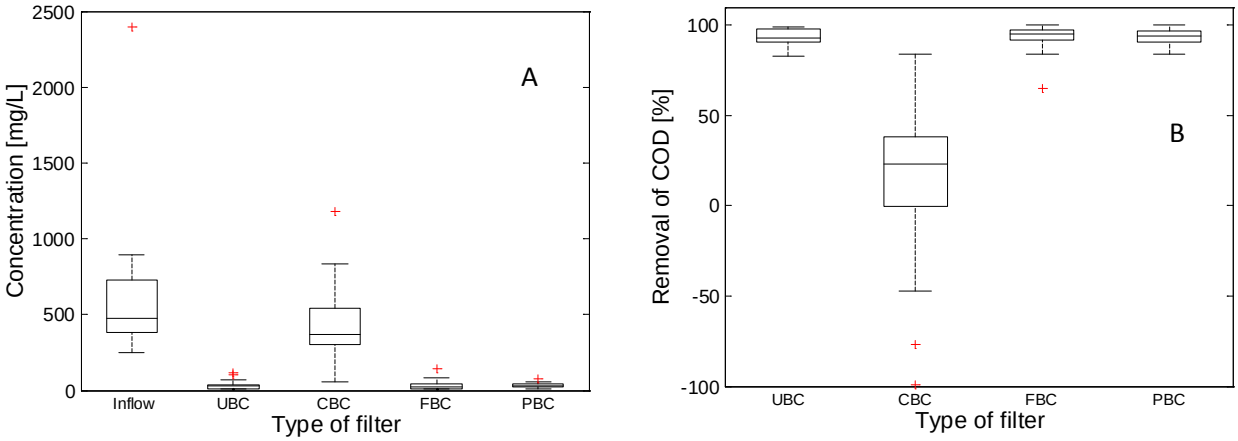

Figure 3. Boxplots of A) chemical oxygen demand (COD) concentration and B) COD removal in the unimpregnated biochar filter (UBC), CaO-impregnated biochar filter (CBC), $FeCl_3$-impregnated biochar filter (FBC) and biochar filter mixed with Polonite (PBC). The box is the quartiles of the data set and the medians are shown as a straight line in the box. Max and min values are the whiskers and outliers are shown as red crosses.

Filter performance in phosphorus and phosphate removal

The concentration of Tot-P in the influent was on average 4.98 mg L^{-1}, with a maximum of 6.25 mg L^{-1} and a minimum of 1.27 mg L^{-1} measured during the experimental period (Figure 4). The Tot-P concentration in effluents of FBC and CBC filters started above 0.5 mg L^{-1}, which was attributed to presence of free (not attached to the biochar surface) Fe and Ca particles binding Tot-P. Continued operation of the filter resulted in wash-out of free particles from the FBC and CBC filters. Consequently, the Tot-P concentrations in FBC and CBC filters decreased significantly after 4 weeks, to reach <0.27 mg L^{-1} during the entire project duration. The average concentration of Tot-P in the effluent from the FBC and CBC filters was 0.25 ± 0.04 mg L^{-1} and 0.27 ± 0.05 mg L^{-1}, respectively, leading to average removal of 92 ± 0.07 % in FBC and 91 ± 0.08 % in CBC (Figure 4).

Table 1. Influent and effluent average concentration (Conc) ± standard deviation and corresponding removal efficiency (E) of total phosphorus (Tot-P), phosphate-P (PO_4-P), chemical oxygen demand (COD) and pH for the unimpregnated biochar filter (UBC), CaO-impregnated biochar filter (CBC), $FeCl_3$-impregnated biochar filter (FBC) and the biochar filter mixed with Polonite (PBC)

Type of	**Tot-P**		**PO_4-P**		**COD**		**Ph**
water	**Conc** (mg L^{-}	**E**	**Conc** (mg L^{-}	**E**	**Conc** (mg L^{-}	**E**	
Influent	3.57±1.33	-	3.25±1.36	-	580±360	-	7.1±0.4
FBC	0.25±0.17	92±6	0.16±0.10	97±2	31±25	94±6	2.6±0.9
CBC	0.27±0.27	91±8	0.24±0.21	96±5	430±230	18±41	7.8±0.4
PBC	1.82±1.18	42±49	1.29±0.49	49±42	34±14	93±4	7.8±0.7
UBC	2.71±1.26	15±64	2.09±0.83	17±59	34±24	93±5	6.7±0.5

The concentration of Tot-P in the effluent from PBC and UBC gradually increased from about 1 mg L^{-1} during the first five weeks to reach a maximum of 5.44 and 5.34 mg L^{-1}, respectively, in week 18. During weeks 18 and 22, breakthrough in the UBC and PBC filters was observed

and the concentration of Tot-P (also PO_4-P) in the effluent of these two filters (5.44 and 5.34 mg L^{-1}, respectively) exceeded that measured in the influent (1.89-3.07 mg Tot-P L^{-1}). The PFU and UBC filters recovered from the breakthrough after week 23. However, the average removal efficiency of PBC and UBC was low (42 ± 49 % and 15 ± 64 % respectively) (Table 1, Figure 4). During the whole experiment, the PBC and UBC filters had higher mean Tot-P effluent concentration (1.82 ± 1.18 and 2.71 ± 1.26 mg L^{-1}, respectively) and lower removal efficiency (15 ± 64 and 42 ± 49 %, respectively) than the FBC and CBC filters (Table 1). In contrast, CBC and FBC had low outflow concentration of Tot-P (0.25 ± 0.17 and 0.27 ± 0.27 mg L^{-1}, respectively) and displayed high removal efficiency (92 ± 6 and 91 ± 8 %, respectively) (Table 1).

The PO_4-P concentrations were similar to the Tot-P concentrations and varied between 1.07 and 5.70 mg L^{-1} in the influent throughout the experiment, with a mean value of 3.25 ± 1.36 mg L^{-1}. The concentration and removal efficiency of PO_4-P showed similar trends to those observed for Tot-P (Figure 4A and 4B). However, all four biochar filters tested displayed 2-5% higher removal of PO_4-P than Tot-P (Figure 5).

The influent Tot-P and PO_4-P concentrations displayed a relatively wide variation, which was also the case for the effluent from the untreated biochar filter (UBC), for PO_4-P in particular (Figure 6). In contrast, FCBC and CBC effluent concentrations were low and did not vary much during the experiment. Concentrations in the effluent of the PBC filter also fluctuated, but varied less than in the UBC effluent.

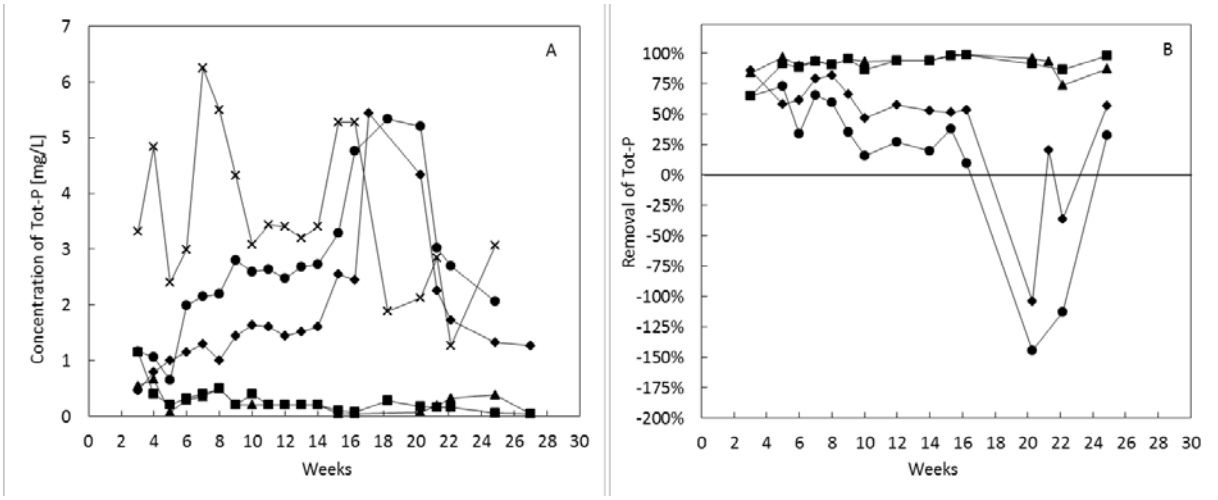

Figure 4. A) Total phosphorus (Tot-P) concentration in the influent (x) and in the effluent from the untreated biochar filter (UBC; ●), CaO-impregnated biochar filter (CBC; ■), $FeCl_3$-impregnated biochar filter (FBC; ▲) and biochar filter mixed with Polonite (PBC; ◆). B) Removal efficiency of Tot-P in UBC (●), CBC (■), FBC (▲) and PBC (◆).

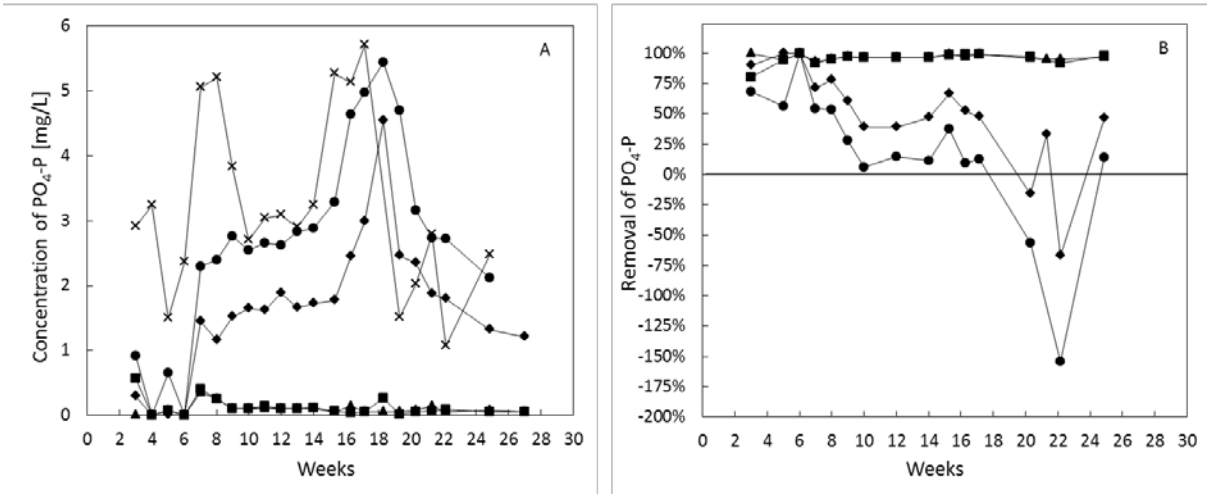

Figure 5. A) Phosphate phosphorus (PO_4-P) concentration in the influent (x) and effluent from the $FeCl_3$-impregnated biochar filter (FBC; ▲), CaO-impregnated biochar filter (CBC; ■), biochar filter mixed with Polonite (PBC; ◆) and unimpregnated biochar filter (UBC; ●). B) Removal efficiency of Tot-P in FBC (▲), CBC (■) , PBC (◆) and UBC (●) filters.

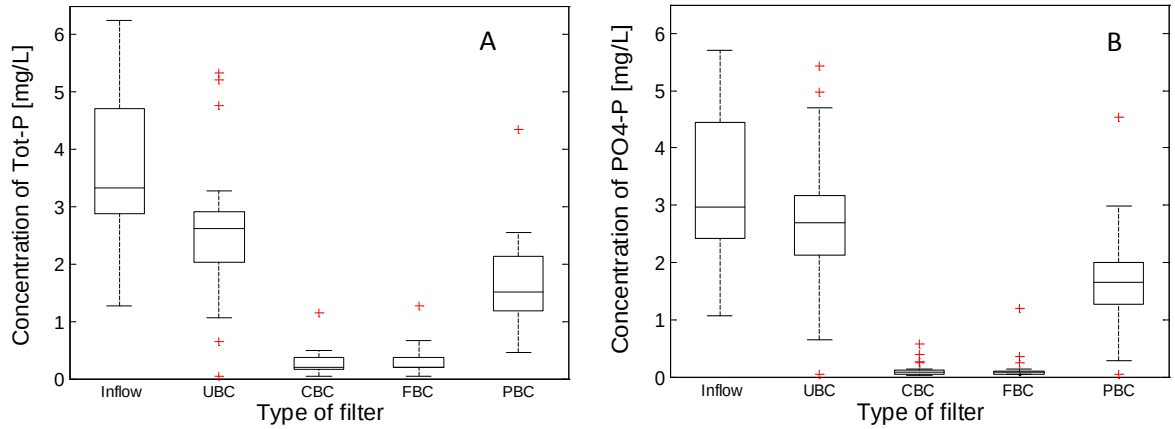

Figure 6. Boxplots of A) Total phosphorus (Tot-P) concentration and B) phosphate-P (PO_4-P) concentration in the unimpregnated biochar filter (UBC), CaO-impregnated biochar filter (CBC), $FeCl_3$-impregnated biochar filter (FBC) and biochar filter mixed with Polonite (PBC). The box is the quartiles of the data set and the medians are shown as a straight line in the box. Max and min values are the whiskers and outliers are shown as red crosses.

Leaching of phosphorus and phosphate after a pause in operations

Following the re-start of the impregnated biochar filters after the pause in operation, the FBC filter showed occasional releases of Tot-P and PO_4-P in its effluent, i.e. 3.6 mg PO_4-P L^{-1} after one day and 2.8 mg PO_4-P L^{-1} seven days after the re-start (Figure 7). During the time between these releases, the concentration of PO_4-P in the effluent of FBC was <0.3 mg L^{-1}. The effluent from the CBC filter showed a PO_4-P concentration of <0.5 after the re-start. One week after the re-start, the CBC filter recovered its high removal efficiency (<0.2 mg PO_4-P L^{-1} and <0.7 mg Tot-P L^{-1}). During the re-start period, the effluent of PBC and UBC showed higher PO_4-P and Tot-P concentrations than the influent wastewater.

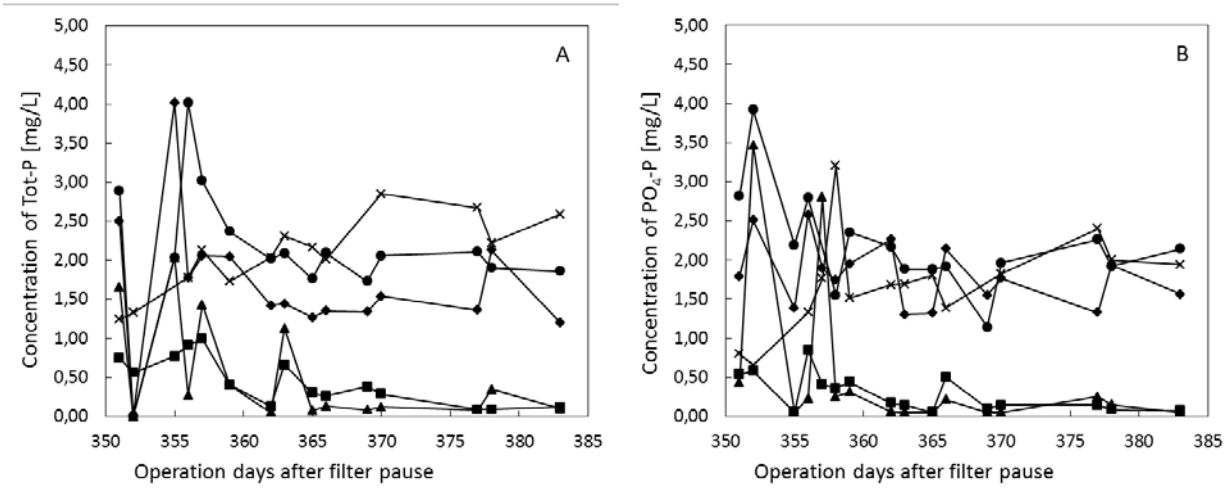

Figure 7. A) Total phosphorus (Tot-P) concentration and B) PO_4-P concentration in the influent (x) and filter effluent after re-start of the filters following a pause in operation: unimpregnated biochar (UBC; ●), CaO-impregnated biochar (CBC; ■), $FeCl_3$-impregnated biochar (FBC; ▲) and biochar mixed with Polonite (PBC; ◆)

DISCUSSION AND CONCLUDING REMARKS

In filters designed for phosphorus removal, the capacity of the filter medium to bind PO_4-P and consequently Tot-P depends on pH and on the availability of surfaces rich in aluminium (Al), iron (Fe) and calcium (Ca) (Arias et al., 2001; Hylander et al., 2006). Thus biochar filters impregnated with Ca (CBC filter) and Fe (FBC filter) showed efficient removal of Tot-P and PO_4-P compared with unimpregnated biochar. The PO_4–P adsorbed to the FBC and CBC filters reacted with Fe and Ca minerals to form strong precipitates or surface complexes. That is why little PO_4–P was released after re-start of the filters following the pause in operation. The pH in the media affected the reaction. The Fe-impregnated biochar (FBC) had a low pH (2-4 SU). At this pH level, the reaction of phosphate with Fe can be expected to result in formation of $FePO_4 \cdot H_2O$. In contrast, in the CBC filter with its higher pH, the PO_4^{3+} ion formed complexes with Ca ions, and $CaHPO_4 \cdot 2H_2O$ and $Ca_4H(PO_4) \cdot 3H2O$ could have been formed (US EPA, 2002). The surface of the biochar was not examined using X-ray diffraction (XRD) to identify its chemical composition.

The concentrations of PO_4-P and Tot-P in the influent to the column filters varied considerably, as can be seen in Figures 4 and 5 and also in the box plot in Figure 6. The variation probably reflected the daily and weekly variations in the quality of the wastewater from the wastewater treatment plant.

The unimpregnated biochar (UBC) filter showed the lowest removal efficiency for Tot-P (15 ± 64 %) of all filters (Table 1). This is even lower than could be expected from a fully functioning vertical soil filter, which has an estimated Tot-P removal of 70 ± 20 % (Olshammar et al., 2015). The Tot-P reduction in UBC was also in the lower end of the range (32-89%) reported for other biochar filters of organic origin (pine-spruce wood or willow wood) (Dalahmeh, 2016). PBC had an average removal of 42 ± 49 % of Tot-P, which is what

could be expected from vertical soil filters and also of previously studied biochar. The Tot-P concentration in the effluent of the UBC and PBC filters continued to increase during the experiment and their ultimate capacity in binding PO_4-P seemed to be soon reached. The Tot-P removal in CBC and FBC was 91 ± 8 % and 92 ± 6 %, respectively, and they had an even higher removal rate of PO_4-P than Tot-P. This is similar to findings in previous batch adsorption studies, where Fe-modified biochar has been shown to remove up to 99% of phosphorus (Chen et al., 2011; Liu et al., 2015).

The FBC filter lowered the pH more than the other filters, by on average 2.6 ± 0.9 (Table 1), which can be expected in Fe-rich environments. The yellow colour of the CBC effluent originated from the parent material, pine bark. Water filtered through bark gains a red-brown colour due to the release of organic acids (Dalahmeh, 2012). Since the colour comes from the bark, this indicates that pyrolysis of the biochar was probably not fully complete, as pure biochar would not release any colour. Organic acids present in bark would lower the pH, meaning that the pH might have been even higher in the CBC effluent if the pyrolysis had been complete. Initially, Ca and Fe ions were most likely released from the CBC and FBC filter materials to the effluents, which could possibly explain the high COD and higher turbidity of these effluents at the beginning of the experiment. The COD removal in PBC and UBC filters was high, as also shown in most previous biochar studies (e.g. 90% was reported by Dalahmeh (2016).

According to the Swedish Agency for Marine and Water Management, the effluent from OWTSs should achieve a minimum reduction of 90% for phosphorus in areas classified as sensitive to wastewater discharge and 70% at general sites (Havs- och vattenmyndighetens, 2016). Furthermore, the reduction in organic material should be at least 90% for all sites. Among the biochars tested, only the CBC (calcium-impregnated biochar) and FBC (iron-impregnated biochar) filters complied with the effluent quality regulation set by the Swedish Agency for Marine and Water Management for general sites and sites in sensitive areas (Table 1). This compliance with the regulations was found during the full 510 days of filter operation (only 365 days are reported here). The long-term performance of the impregnated biochar filters, their service life and their response to variations in hydraulic loading are still under investigation.

In summary, biochar filters impregnated with CaO and iron, i.e. CBC and FBC, significantly enhanced removal of Tot-P and PO_4-P in small-scale onsite systems. Although the FBC and CBC filters had not reached saturation level by the time of this report, their capacity to bind P is expected to be finite and they could become saturated with other pollutants (e.g. organic matter, ammonia, pharmaceuticals and trace metals). Thus, it is recommended that these types of impregnated biochar be used in a separate phosphorus filter module, where solids and organic matter are removed from the wastewater prior to treatment in the iron/calcium impregnated biochar.

COMBINED VERTICAL-HORIZONTAL FLOW BIOCHAR FILTER FOR ENHANCED NITROGEN REMOVAL

BACKGROUND

Total nitrogen is removed in vertical soil filters partly by adsorption of ammonium (NH_4^+), but the main removal is achieved by nitrification-denitrification in biofilm (Pell, 1991) or ANAMOX (Erler et al., 2008). Nitrification is achieved by nitrifying bacteria (aerobic and autotrophic bacteria), which derive their energy from oxidation of NH_4^+ to nitrite (NO_2) in a first step and then further to nitrate (NO_3) (Bassin et al., 2012). In a second stage, facultative bacteria called denitrifying bacteria reduce NO_3 or other nitrogen oxides to form nitrous oxide (N_2O) and nitrogen gas (N_2) under anaerobic conditions, in a process called denitrification (Brar et al., 1978). During denitrification, NO_3 is used as source of oxygen for respiration. Denitrifying bacteria are heterotrophic, which means that they need an organic carbon source to remove nitrogen.

The combined vertical-horizontal flow biochar filter system tested in this project was designed to include two sections (Figure 8). The first of these was a vertical flow (aerobic) section in which the wastewater percolated through the biochar media in unsaturated mode. The unsaturation mode means that the pores in the media had access to air, which would encourage nitrification of NH_4-N. The second section in the system was a horizontal flow section, in which biochar was saturated with water and had little access to air. This enabled anaerobic conditions to enhance the denitrification process, and hence nitrogen removal from the wastewater. In this section of the report, we present the performance of the combined laboratory-scale vertical-horizontal flow biochar filter system in terms of organic matter and nitrogen removal during an operating period of one year.

MATERIALS AND METHODS

A biochar filter with an aerobic vertical flow section combined with an anaerobic horizontal flow section was installed and filled with biochar originating from mixture of pine and spruce wood biomass obtained from Vildelkol AB (Vindelkol, 2017). The horizontal and vertical flow sections were installed using two 70-L boxes, each measuring 74 cm × 29 cm × 40 cm (height x width x depth) cm placed on top of each other. In the vertical flow filter, a 3-cm drainage layer which had a slope of 1.5: 60 (i.e. 2.5%) was prepared with coarse biochar (8-16 mm in diameter) at the bottom. The section was then filled up to 30 cm with biochar with particle size varying between 2.5 and 5 mm. A second 3-cm layer of coarse biochar was placed on top of the main filter to prevent clogging on the surface.

The horizontal flow biochar section was prepared by filling the box with coarse biochar (25-40 mm in diameter) in two 10 cm layers at the inlet and outlet sides. The remaining 54 cm of the section was then filled with biochar (1.6-2.5 mm diameter). The depth of the biochar in the horizontal flow section was 30 cm. The outlet of the horizontal flow section was located 4 cm lower than the inlet. Before the start of the experiment, the filter was gently washed with distilled water. During the experiment, pumps fed the filter with 1.5 L three times a day, at 9:00, 16:00 and 01:00 (total of 4.5 L day^{-1}), which gave a flow of around 21 L m^{-1} day^{-1} (64 L m^{-3} day^{-1}).

The filters were fed with real wastewater brought from Uppsala WWTP (Kungsängsverket).

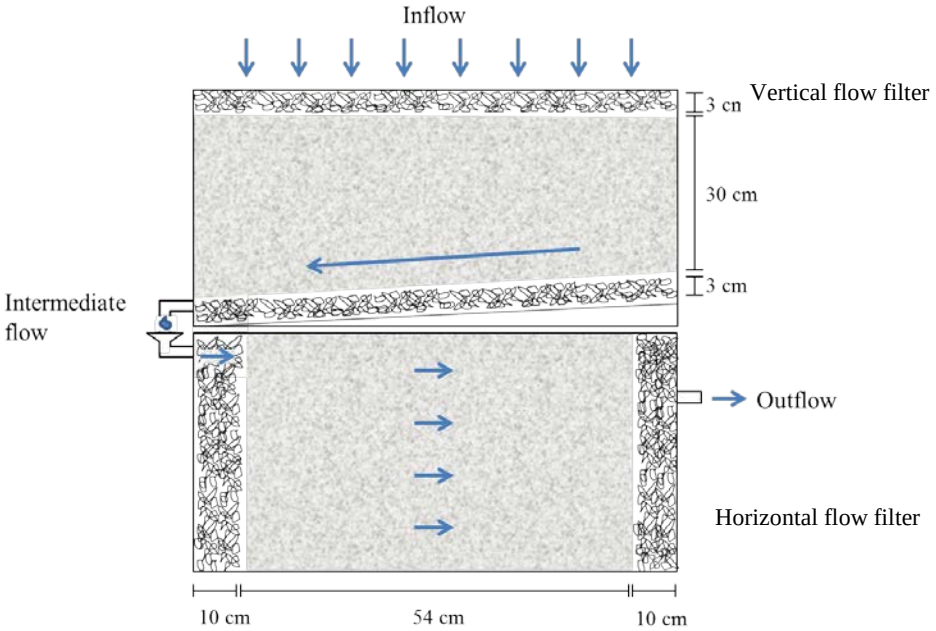

Figure 8. Combined aerobic vertical flow and anaerobic horizontal flow biochar filter unit for wastewater nitrogen removal. The material in the filter was biochar made from hardwood biomass. The figure is obtained from Stenström (2017).

Samples from the inflow, effluent from the vertical flow filter and effluent from the horizontal flow filter were collected once a week and the pH and Tot-N, NH_4-N, NO_3-N and COD concentrations were analysed. Removal efficiency was calculated according to Equation 1 (see page 12 of this report). Nitrification and denitrification efficiency was calculated according to equation 2 and 3, respectively:

$$Nitrification\ efficiency = 100\ \frac{[NO_3{-}N]_{VFF} - [NO_3{-}N]_{inflow}}{[Tot{-}N]_{inflow}} \qquad (2)$$

where $[NO_3 - N]_{VFF}$ is the concentration of nitrate-nitrogen in the effluent from the vertical flow filter, $[NO_3 - N]_{inflow}$ is the concentration of nitrate-nitrogen in the influent wastewater and $[Tot - N]_{inflow}$ is the total nitrogen in the influent.

$$Denitrification\ efficiency = 100\ \frac{[NO_3{-}N]_{VFF} - [NO_3{-}N]_{HFF}}{[Tot{-}N]_{inflow}} \qquad (3)$$

where $[NO_3 - N]_{HFF}$ is the concentration of nitrate-nitrogen in the effluent from the horizontal flow filter.

RESULTS

Removal of organic matter

The average concentration of organic matter COD in the influent was 480 ± 180 mg L^{-1}. After the start-up period of the filters, the COD concentration in the effluent from the vertical and horizontal flow filters decreased significantly, from about 400 mg L^{-1} to reach <80 mg L^{-1} in the effluent of the vertical flow and <20 mg L^{-1} in the effluent of the horizontal flow filter. This gave an overall reduction in COD of 92%. Over the whole experimental period, the average concentration of COD was 87 ± 80 mg L^{-1} in the vertical flow filter and 35 ± 22 mg L^{-1} in the horizontal flow filter (Figure 9). On average, the removal of COD in the combined vertical-horizontal flow filter system was high (92 ± 6 %; Figure 9), with most of the removal (72 ± 25 %) occurring in the vertical flow filter.

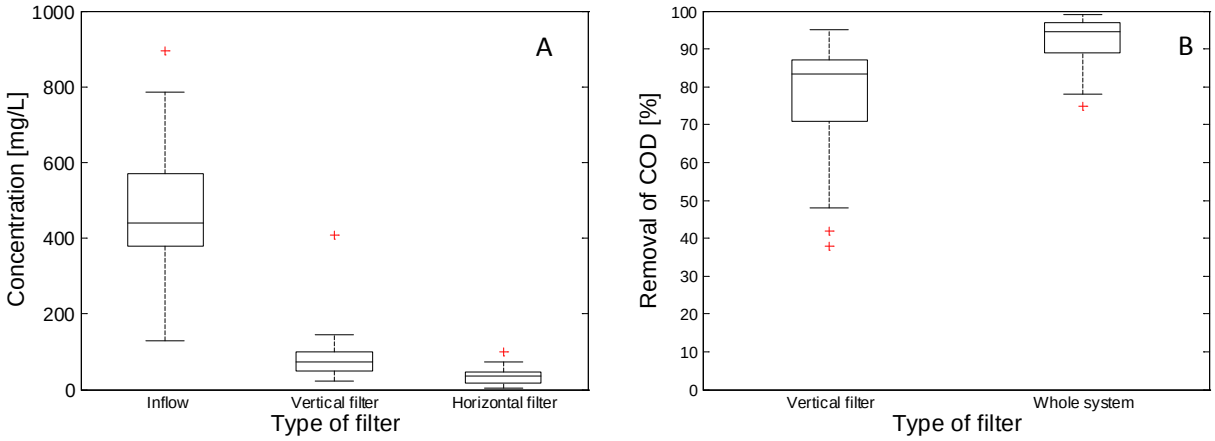

Figure 9. Box plots of A) the concentration of chemical oxygen demand (COD) and B) the percentage removal of COD in the vertical flow filter and in the whole system (vertical + horizontal filters). The box is the quartiles, with the horizontal line in the box showing the median. Max and min values are the whiskers and outliers are presented as red crosses

Removal of nitrogen

The concentration of Tot-N in the influent to the vertical aerobic filter varied between 25 and 63 mg L^{-1}, with a mean of 39.54 ± 8.26 mg L^{-1} (Table 2; Figure 10A). Treatment in the vertical section resulted in the Tot-N concentration decreasing to 21.23 ± 3.36 mg L^{-1}. During the start-up period (initial 9 weeks), removal of Tot-N in the combined vertical-horizontal flow filter was low (19-36%). As the biofilm and population of nitrifying and denitrifying bacteria developed over time, the rate of removal of Tot-N in the system increased, to reach 74% after 14 weeks. The performance of the system improved even more after a pause in operation. After the pause, overall removal of Tot-N reached 88% after one year of operation (Figure 10B). Overall removal of Tot-N during the whole period reported here (57 weeks

excluding the start-up period) was 75 ± 12 %, with an average concentration of 9 ± 4 mg L^{-1} in the effluent from the horizontal flow filter (Table 2).

Table 2. Mean removal efficiency and concentration ± standard deviation for all pollutants measured in a flow filter system with an aerobic vertical flow section followed by an anaerobic horizontal flow section

	Concentration (mg L^{-1})			Removal efficiency (%)		
Pollutant	**Inflow**	**Vertical section effluent**	**Horizontal section effluent**	**Vertical section**	**Horizontal flow section**	**Total Removal efficiency**
pH	7.1±0.1	6.7±0.4	6.8±0.4			
COD	480±180	87±80	35±22	72±25	36±39	92±6
Tot-N	41±10	31±7	9±4	22±20	52±41	75±12
NH_4-N	39±10	10±7	5±4	74±17	11±15	84±10
NO_3-N	1.5±0.7	17±8	3.3±1.6	40±26*	37±26**	

*Nitrification efficiency (estimated according to equation 2).
**Denitrification efficiency (estimated according to equation 3).

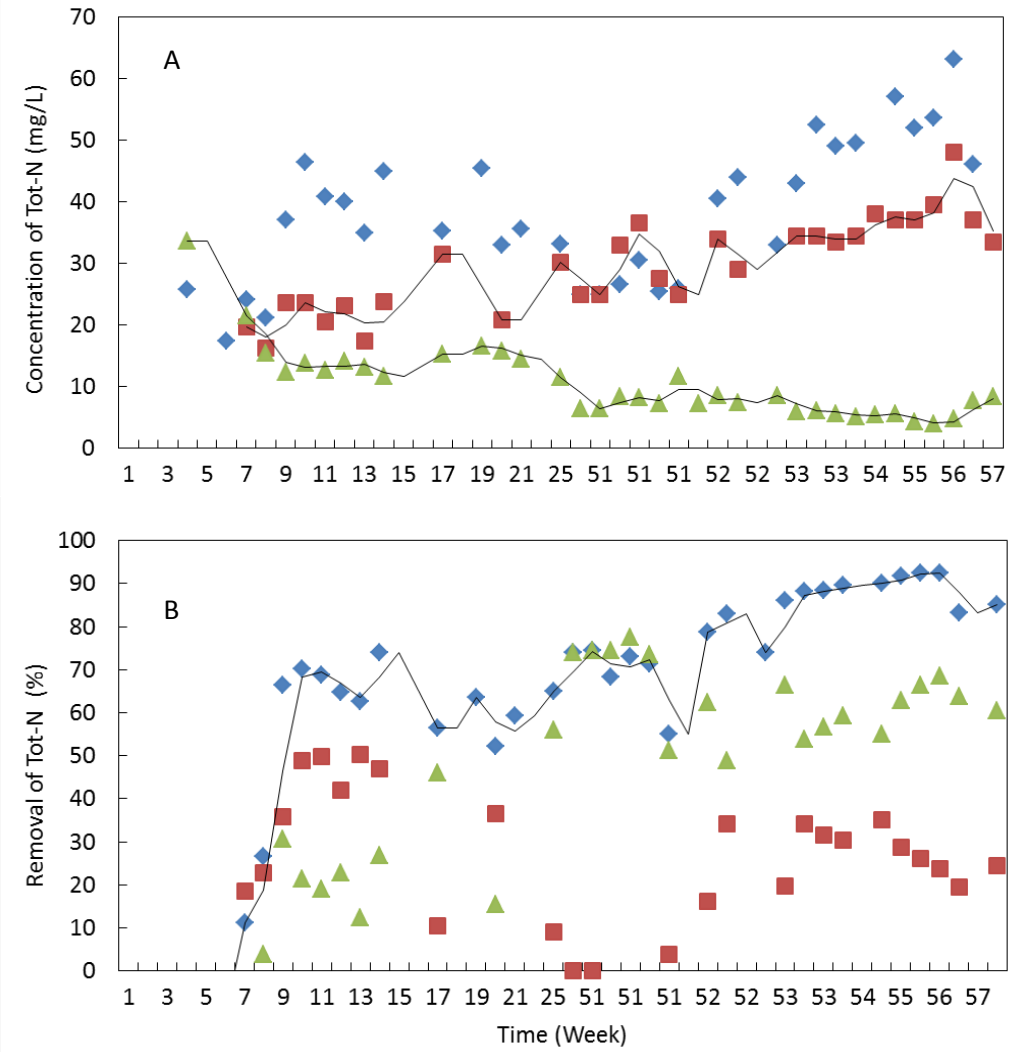

Figure 10. Weekly variation in A) total nitrogen (Tot-N) concentration and B) Tot-N removal in the vertical flow filter section (■), the horizontal flow filter section (▲) and the combined filter system (◆) during 57 weeks of operation.

The influent wastewater contained 39 ± 10 mg L^{-1} of ammonium-nitrogen (NH_4-N), which comprised more than 94% of Tot-N in the influent. In the vertical flow filter, a fraction of the NH_4-N in the wastewater was adsorbed to the biochar filter media and another fraction was converted into nitrate (NO_3-N) via nitrification in this section (Figures 11 and 12). It was difficult to determine the fraction of NH_4-N removed by nitrification and that removed by adsorption. The average concentration of NH_4-N was 9 ± 4 mg L^{-1} in the effluent from the vertical flow filter and 5 ± 4 mg L^{-1} in the effluent from the horizontal flow filter (Table 2).

During the first 9 weeks of filter operation, the nitrification efficiency in the vertical flow section was low (<20%) and hence the NO_3-N concentration in the effluent from the vertical flow filter was low (<10 mg L^{-1}) (Figure 12). As the nitrification bacteria grew and became more active, the efficiency of the nitrification increased over time and the steady state nitrification efficiency varied within 40-50% with NO_3-N concentration (22-29 mg L^{-1}) in the effluent from the vertical flow section (Figure 12). The efficiency of nitrification in the vertical flow filter was on average 40 ± 26 % during the 57-week period excluding the start-up period (Table 2).

While nitrification was occurring in the vertical flow filter, denitrification (conversion of NO_3-N to nitrogen gas or NO_2) was occurring in the horizontal flow filter working under anaerobic conditions. The denitrification efficiency followed a similar trend to that of nitrification, i.e. denitrification was low (<10%) at the beginning of filter operation but increased over time to stabilise at around 40% (Figure 12). The concentration of NO_3-N in the effluent from the horizontal flow filter reached <5 mg L^{-1} after 52 weeks and continued at this level until the end of the reported operating period (Figure 12).

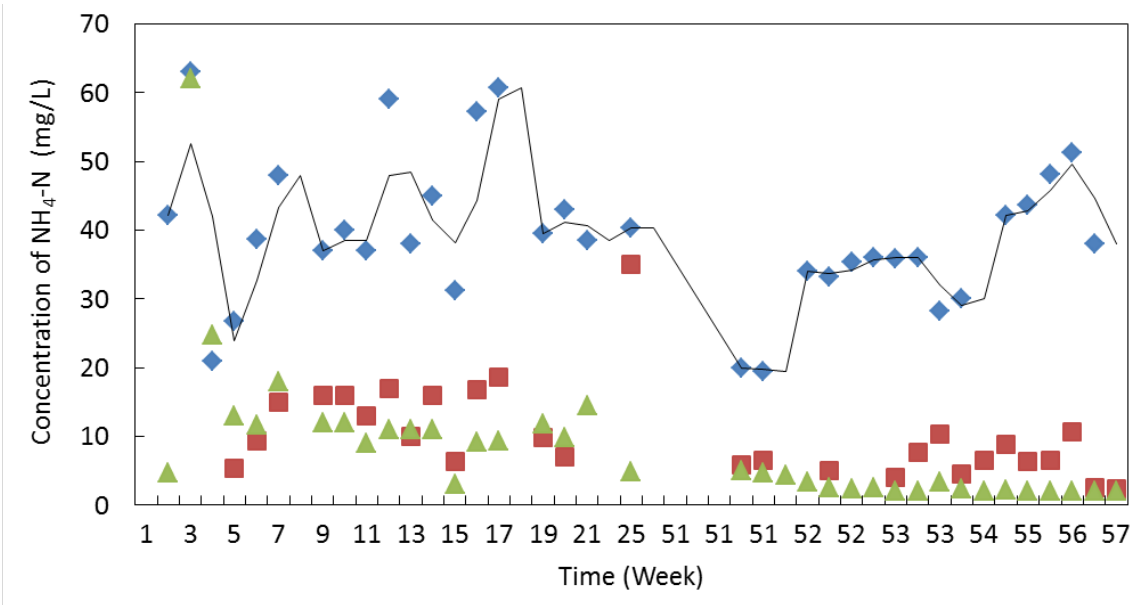

Figure 11. Weekly variation in ammonium-nitrogen **(**NH_4-N) concentration in the vertical flow filter section (■), the horizontal flow filter section (▲) and the combined filter system (◆) during 57 weeks of operation.

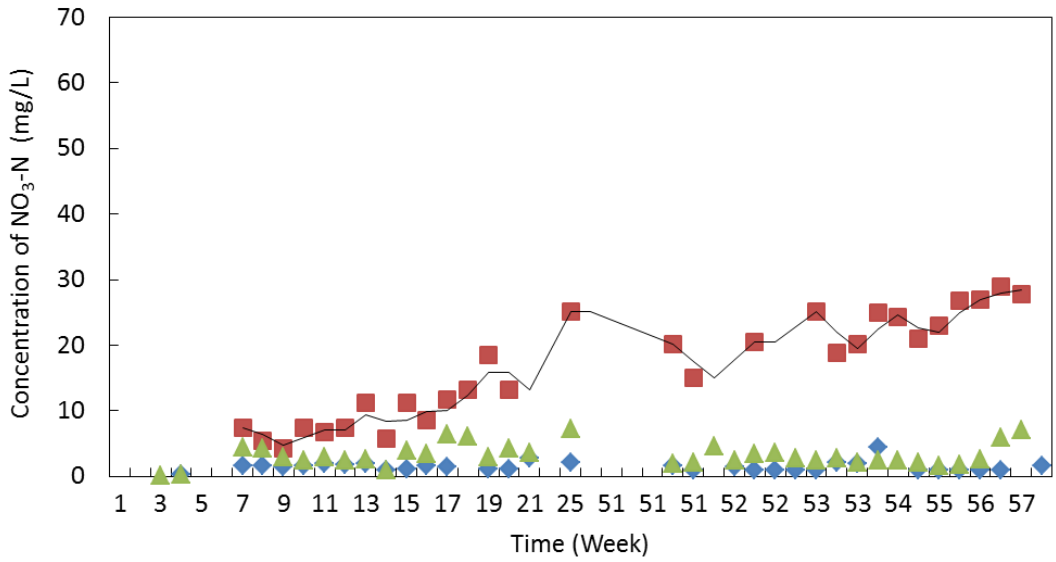

Figure 12. Weekly variation in nitrate-nitrogen (NO_3-N) concentration in the vertical flow filter section (■), the horizontal flow filter section (▲) and the combined filter system (◆) during 57 weeks of operation.

DISCUSSION AND CONCLUDING REMARKS

Overall removal of Tot-N in the combined vertical-horizontal flow filter was 75 ± 12 % (Figure 10; Table 2). This was higher than in conventional vertical soil filers, where removal rates of 30 ± 10 % can be expected (Olshammar et al., 2015) and also higher than in alternative onsite wastewater treatment plant solutions in general.

Combining the anaerobic vertical flow filter with an anaerobic stage (horizontal flow filter) provided a suitable environment for nitrification-denitrification and hence enhanced nitrogen removal significantly. In fact, after stabilisation of the combined horizontal-vertical flow filter, most of the nitrogen removal (52 ± 41 %) occurred in the saturated section (horizontal flow filter) and only small proportion of Tot-N removal (22 ± 20 %) occurred in the vertical flow filter. This indicates that addition of a saturated flow filter, e.g. horizontal flow, is crucial to achieving acceptable levels for nitrogen removal from wastewater. However, the vertical flow filter was still a critical component of the system, since without it nitrification would have not been achieved. The increase in NO_3-N concentration in the vertical flow filter and its decrease in the horizontal flow section showed that nitrification and denitrification took place in the system. However, the nitrification started slowly and gradually improved with time, due to slow growth rate of nitrifying bacteria. Nitrifying bacteria are autotrophic and use inorganic carbon sources, e.g. CO_2, to obtain energy. This causes the slow growth rate of nitrifying bacteria (Bassin et al., 2012). In addition, other factors such as temperature,

pH, concentration of organic matter and concentration of NH_4 affect the growth of nitrifying bacteria (Bassin et al., 2012).

According to the Swedish Agency for Marine and Water Management, effluent from onsite wastewater treatment systems should show a minimum reduction of 50% for nitrogen if the surrounding area is classified as sensitive (Havs- och vattenmyndighetens, 2016). The combined vertical-horizontal flow filter system achieved on average 75 ± 12 % removal of Tot- N, which is in good compliance with the regulations.

In summary, the combined vertical-horizontal flow biochar filter system proved to be efficient in organic matter removal from wastewater and showed promising performance regarding nitrogen removal in onsite wastewater treatment. This report describes the performance of the horizontal-vertical flow filter system only under fixed loading conditions. The performance of the filters under different loading conditions is under investigation and will be reported in an upcoming report in this series. In addition, the design of the filter system with respect to depth:width:length ratio of the horizontal flow system needs investigation and the recommended loading conditions still need to be tested and optimised.

MODELLING POLLUTANT FLOW DYNAMICS AND POLLUTANT REMOVAL IN BIOCHAR FILTERS

BACKGROUND

Modelling flow and pollutant transport in filter systems is a valuable tool in understanding the flow dynamics within the filters and how it influences the transport of pollutants. Commonly used models for such purposes include PHREEQC (Parkhurst, 2013), PHWAT (Mao et al., 2006), FITOVERT (Giraldi et al., 2010) and the HYDRUS wetland module. The HYDRUS model considers transport and reactions of the typical pollutants in wastewater: organic matter, nitrogen and phosphorus present in soil (Langergraber & Šimůnek, 2012). It has been used for simulating flow and pollutant transport through sand-type media in vertical flow filters (Karlsson et al., 2015) and through constructed wetlands (Langergraber & Šimůnek, 2012).

In this section of the report, we present the results obtained in computer simulations of water flow and organic matter removal in vertical flow unsaturated biochar filters used for wastewater treatment. The simulations were carried out using the HYDRUS wetland module. The work presented in this section has also been published by Karlsson et al. (2015) and Ciuk Karlsson (2015).

MATERIAL AND METHODS

Simulated data

The simulated data covered the concentrations of COD, biological oxygen demand (BOD_5); Tot-N, NO_3-N, NH_4-N, Tot-P and PO_4-P in influent and effluent from activated biochar filters (also referred to as charcoal filters) (Table 3). In addition, the input data to the model included microbial activity and substrate-induced potential respiration rate (mg O_2 L^{-1} day^{-1}), which were analysed during the 112-day initial experiment (Run 0), as described by Dalahmeh et al. (2014a), for modelling organic matter degradation (Table 4). The data were obtained from previous studies in which we investigated the performance of vertical flow biochar filters used for wastewater (excluding toilet fractions) treatment in laboratory experiments (Dalahmeh, 2013; Dalahmeh et al., 2014a; Dalahmeh et al., 2014b); (Karlsson et al., 2015) (Table 5). Laboratory-scale biochar filters (diameter 20 cm x height 60 cm) were used for wastewater treatment under fixed and variable hydraulic and organic loading rates in those studies (Table 3).

Table 3. Wastewater characteristics (COD, BOD_5; Tot-N, NO_3-N, NH_4-N, Tot-P and PO_4-P) and hydraulic and organic loads used in simulations of biochar filter operation (Ciuk Karlsson, 2015; Dalahmeh et al., 2014a; Karlsson et al., 2015)

Parameter	**Run 0**	**No. of samples for Run 0**	**Run 1**	**Run 2**	**Run 5**	**No. of samples for Run 1, 2 and 5**
Hydraulic loading rate ($l\ m^{-2}\ day^{-1}$)	32	-	32	64	32	-
Organic loading (g $BOD_5\ m^{-2}\ day^{-1}$)	14	-	13-16	13-16	28	-
Readily available COD ($mg\ L^{-1}$)*	574	-	913	494	1879	-
Slowly available COD ($mg\ L^{-1}$)*	287	-	457	247	940	-
Inert COD ($mg\ L^{-1}$)*	24	-	80	39	241	-
COD ($mg\ L^{-1}$)	885 ± 127	2	1450 ± 470	780 ± 110	3060 ± 830	4
BOD_5 ($mg\ L^{-1}$)	425 ± 152	12	490 ± 49	198 ± 13	875 ± 452	4
Tot-N ($mg\ L^{-1}$)	75.5 ± 9.9	2	72 ± 24	41 ± 5	131 ± 1	4
NH_4-N ($mg\ L^{-1}$)	0.5 ± 0.2	10	19.9 ± 8.9	8.1 ± 2.9	26.1 ± 13.7	4
NO_3-N ($mg\ L^{-1}$)	1 ± 0.6	10	2.2 ± 2.2	1.6 ± 0.3	4.5 ± 1.6	4
Tot-P ($mg\ L^{-1}$)	4.2 ± 2.3	2	2.5 ± 1.4	1.7 ± 0.1	6.3 ± 0.4	4
PO_4-P ($mg\ L^{-1}$)	2.1 ± 0.4	5	1.4 ± 0	1.6 ± 0	5.9 ± 0	4

Table 4. Mean (± standard deviation) measured respiration rate over time at different depths of bark filter and charcoal filter columns during the initial experiment (Run 0) (Ciuk Karlsson, 2015; Dalahmeh et al., 2014a; Karlsson et al., 2015)

Filter depth (cm)	Number of samples (n)	Measured respiration rate (mg O_2 L^{-1} day^{-1})	Estimated biomass concentration (µg biomass COD g^{-1} material)
0-2	10	237 ± 80	478 ± 161
20	10	166 ± 70	335 ± 142
40	10	104 ± 80	210 ± 161
60	10	70 ± 69	141 ± 139

Table 5. Characteristics of the filter materials used in the experiment set-up (Dalahmeh et al., 2012)

Parameter	*Charcoal*
pH (SU)	10.4
Loss on ignition (%)	90
Effective size (mm)	1.4
Uniformity coefficient (-)	2.3
Bulk density (kg m^{-3})	283
Particle density (kg m^{-3})	1900
Porosity (%)	85
Surface area (m^2 g^{-1})	>1000
Hydraulic conductivity (cm h^{-1})	500

Modelling and numerical set-up for flow dynamics

HYDRUS-2D version 2.01.1240 (Šimůnek et al., 2012) was used for simulating the dynamics of wastewater flow through the biochar filters. The domain of fluid transport for each filter model was set to a "2D - Simple" geometry, with diameter 20 cm and height 60 cm, to match the experimental set-up with the filter used in the experiments. This set-up resulted in a two-dimensional finite element mesh with 231 nodes (60 of these are boundary nodes) and 400 triangular finite elements.

To model the hydraulic properties of the biochar filters, parameters for flow dynamics were estimated using the built-in inverse simulation function in HYDRUS-2D, which estimates optimal values of the selected fitting parameters through a Marquardt-Levenberg type parameter estimation technique (Šimůnek et al., 2012). The inverse simulation function requires specification of initial values for the simulated parameters. Different initial values for each estimated parameter were tested and the best result was selected using the "Inverse Solution Results" tab in HYDRUS, which gives the correlation matrix and various goodness-of-fit measures such as the root mean square error (RMSE).

The estimated flow model parameters were validated by using them to simulate a different water flow (approx. 1650 mL day^{-1}) to that used for the calibration (approx. 850 mL day^{-1}).

The simulation with approx. 1650 mL day^{-1} was then compared with observations from that flow (using 1-day mean time series) and goodness-of-fit measures were calculated.

Modelling of biological degradation of organic matter and removal of nitrogen and phosphorus

The HYDRUS wetland module with the CW2D setting was used to simulate degradation of organic matter in active biochar filters. The initial level of heterotrophic biomass (XH) in the filter model was set as the estimated mean value of filter biomass at each depth (0-2, 20, 40 and 60 cm). A linear relationship was used to interpolate the values between the measured depths. Model parameters limiting microbial growth, the heterotrophic lysis rate constant (b_H, day^{-1}) and maximum aerobic growth rate (μ_H, day^{-1}), were adjusted to values which ensured that simulated biomass (XH) matched observations and did not grow excessively during the simulation period (1-5 days).

The maximum aerobic growth rate (μH) was based on the substrate-induced potential respiration rate measured at 0-2, 20, 40 and 60 cm depth. The HYDRUS wetland module describes microbial activity as μg biomass COD per g filter material. Thus, the measured mean potential respiration rates (mg O_2 L^{-1} day^{-1}) were converted to μg biomass COD per g material using equation 4 (Anderson & Domsch, 1978) and equation 5 (Langergraber, 2007):

μg biomass-C g $material^{-1}$ = (μg CO_2 g^{-1} material $hour^{-1}$) x 40.04 + 0.37 (4)

μg biomass COD g $material^{-1}$ = 2.667 (μg O_2) x (μg biomass C g^{-1} material) (5)

Nitrogen and phosphorus compounds were not targeted within the modelling task, but they were specified in input data to allow running of the model (Table 3). Available observed data for nitrogen and phosphorus concentrations were used. Nitrogen was simulated in the HYDRUS wetland module as nitrate (NO_3N), nitrite (NO_2N) and ammonium (NH_4N) and total nitrogen was calculated as NO_3N + NO_2N + NH_4N + nitrogen content of outgoing COD. Phosphorus was simulated as inorganic phosphorus (IP), and total phosphorus (Tot-P) was calculated as IP + phosphorus content of outgoing COD. The filter models were not calibrated to perform with great accuracy for nitrogen and phosphorus simulation.

Model performance and Goodness-of-fit measures

The quality of fitted models was assessed using coefficient of efficiency (E) as a goodness-of-fit measure. The value of E ranges from -∞ to 1, with E = 1 indicating a perfect fit. E was calculated as:

$$E_j = 1 - \frac{\sum_{i=1}^{N} |O_i - P_i|^j}{\sum_{i=1}^{N} |O_i - \bar{O}|^j} \quad (6)$$

where O is the observation, P the model prediction and N the number of observations.

In addition, the index of agreement (d) was used as a goodness-of-fit measure. It was calculated as:

$$d_j = 1 - \frac{\sum_{i=1}^{N}|P_i - O_i|^j}{\sum_{i=1}^{N}(|P_i - \bar{O}| + |O_i - \bar{O}|)^j} \qquad (7)$$

Moreover, the root mean square error (RMSE) between observed and modelled data was calculated. To facilitate comparison between the RMSE of simulations at different hydraulic loading rates, the normalized root mean square error (NRMSE) was also calculated, i.e. as RMSE divided by the range of observed values.

Sensitivity analyses

To check the sensitivity of the filter models, a nominal range sensitivity analysis (NRSA) (Cullen and Frey, 1999) was performed. The base scenario was set as the chosen parameters for filter model and each parameter was then varied as 0.5, 0.8, 0.9, 1.1, 1.2 and 1.5 times the base scenario value. In total, 51 parameters were varied in 306 simulations. If the mean of the absolute value percentage change for a particular parameter was larger than 3%, the model was considered sensitive to that particular parameter.

RESULTS

Simulation of flow dynamics

The simulated flow parameters used for the biochar filter model (Table 6) resulted in a good fit between simulated and measured cumulative effluent at a hydraulic loading rate of 32 L m^{-2} day^{-1} (Figure 13). Analysis of the goodness-of-fit of filter models demonstrated E = 0.92 and d = 0.96. The normalised RMSE ranged from 0.7 to 3.7 % in all simulations, as reported by Ciuk Karlsson (2015).

Table 6. HYDRUS wetland module water flow parameters for the biochar filter model. These results are reported by Ciuk Karlsson (2015)

Parameter (unit)	Value
Residual soil water content (Q_r,-)	0.045*
Porosity (Q_s, -)	0.1357 (0.85**)
Inverse air entry value, α (cm^{-1})	0.3746
Pore connectivity parameter, n (-)	1.596
Saturated hydraulic conductivity, K_s (cm $hour^{-1}$)	500*

*Default value; **Measured value

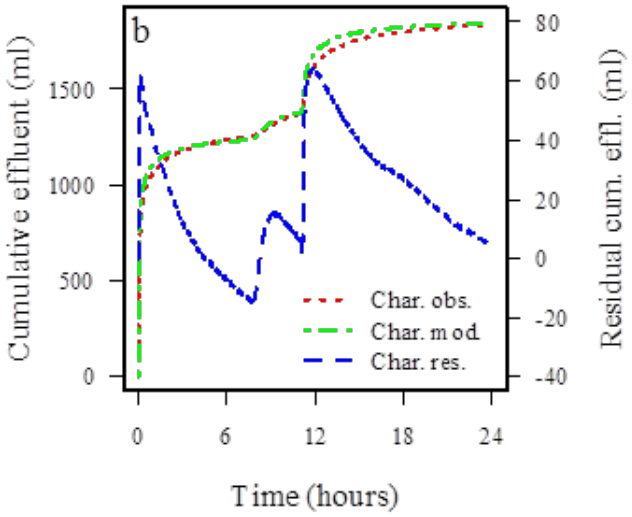

Figure 13. Observed (▪▪▪▪▪) and simulated (▪▪▪▪▪) cumulative effluent (mL) in the biochar filter as a function of time (hours) and calculated residual cumulative effluent (▪▪▪▪▪) of simulated flow, compared with observed cumulative effluent data at a hydraulic loading rate of 32 L m^{-2} day^{-1}. These results are reported by Ciuk Karlsson (2015).

Simulation of biomass growth and organic matter degradation

At a hydraulic loading rate of 32 L m^{-2} day^{-1} and an organic loading rate of 14 g BOD_5 m^{-2} day^{-1}, the simulated biomass growth agreed well with the measured biomass and showed that the most growth (500 g COD g^{-1} biochar) occurred at top surface of the biochar filter (where the wastewater is fed) and the least growth (<200 g COD g^{-1} biochar) occurred at the lowest layer of the filter (effluent level) (Figure 14). The biomass growth declined linearly through the filter layers. On increasing the organic loading rate from 14-15 to 28 g BOD_5 m^{-2} day^{-1}, the simulations showed an increase in biomass in filter layers 0-2, 20 and 40 cm, but not in the deepest layer in the filter (60 cm) (Figure 15).

Simulation of organic matter and other pollutant removal

The simulated biomass growth resulted in good agreement between the measured and simulated COD removal at an organic loading rate of 14 g BOD_5 m^{-2} day^{-1} (Runs 0 and 1) (Figure 16). The simulations of COD degradation in the biochar filters showed that simulated effluent concentration of COD (57 mg L^{-1}) was consistent with the measured concentration (48 ± 11 mg L^{-1}) at a hydraulic loading rate of 32 L m^{-2} day^{-1} and an organic loading rate of 14 g BOD_5 m^{-2} day^{-1} (Figure 16). However, at a hydraulic loading of 64 L m^{-2} day^{-1} (Run 2) and an organic loading rate of 28 g BOD_5 m^{-2} day^{-1}, the simulated COD removal was 15-20% lower than the measured COD removal (Figure 16).

The simulated removal of nitrogen agreed well with the observed removal in biochar filters for organic loading rates of 15 and 28 g BOD_5 m^{-2} day^{-1} (Runs 1 and 2), but the model failed to simulate removal of nitrogen under other loading conditions. Moreover, the model failed to simulate removal of phosphorus in the biochar filters under all loading conditions (Figure 17).

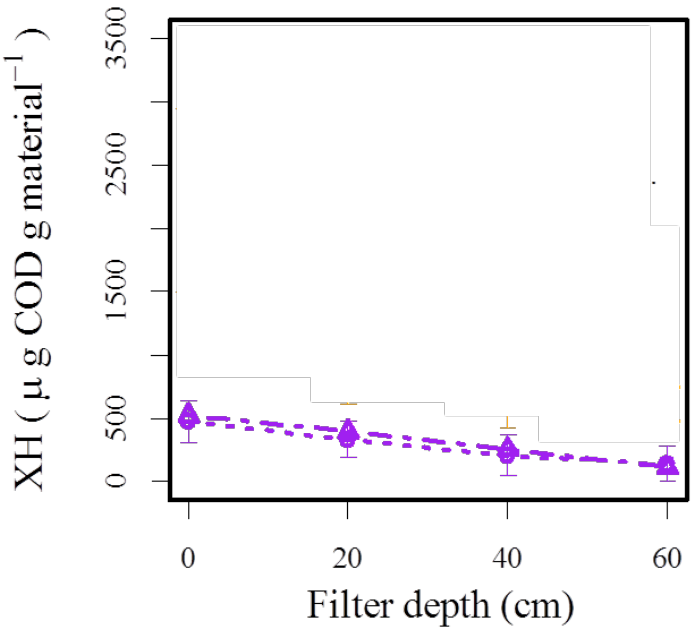

Figure 14. Observed (●) and simulated (▲) concentration of heterotrophic microorganisms (XH) in biochar filters at depths 0, 20, 40 and 60 cm, with hydraulic loading rate 32 L m^{-2} day^{-1} and organic loading rate 14 g BOD_5 m^{-2} day^{-1}. The points show mean values determined during a 5-day simulation and the error bars display standard deviation.

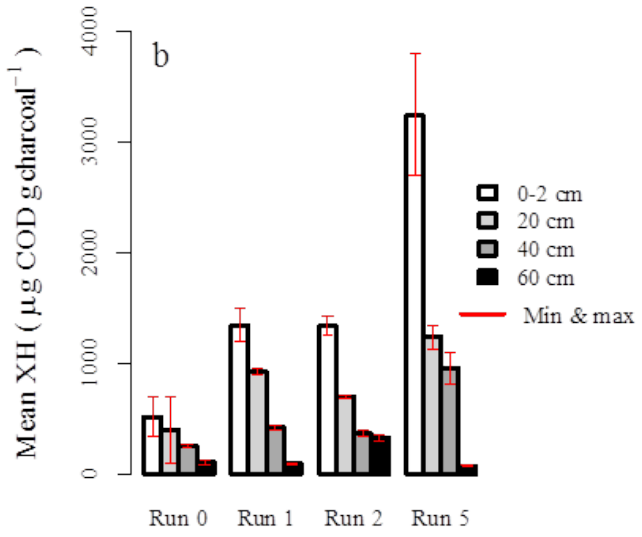

Figure 15. Simulated concentration of heterotrophic microorganisms (XH) in biochar filters at depths 0-2, 20, 40 and 60 cm for different hydraulic (HLR) and organic (OLR) loading rates. Run 0: HLR = 32 L m^{-2} day^{-1} and OLR = 14 g BOD_5 m^{-2} day; Run 1: HLR = 32 L m^{-2} day^{-1} and OLR = 13 ; Run 2: HLR = 64 L m^{-2} day^{-1} and OLR = 13 g BOD_5 m^{-2} day^{-1}, Run 5: HLR = 32 L m^{-2} day^{-1} and OLR = 28 g BOD_5 m^{-2} day^{-1}. These results are reported by Ciuk Karlsson (2015).

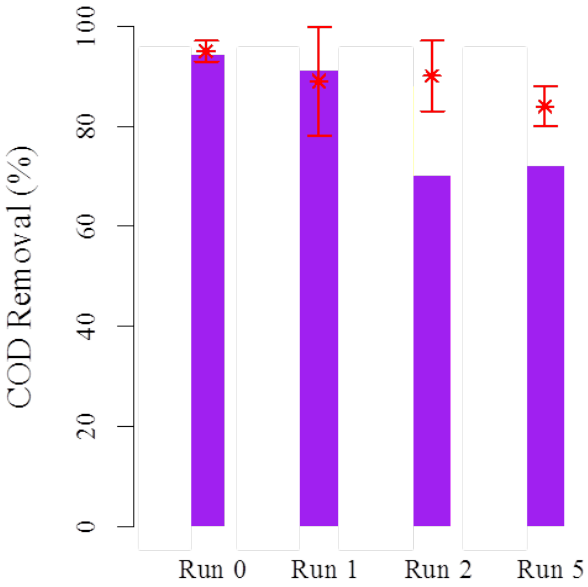

Figure 16. Simulated (columns) and observed (red bars, * with standard deviation, n = 4) removal (%) of chemical oxygen demand (COD) in biochar filters at four different loading regimes. Run 0: HLR = 32 L m^{-2} day^{-1} and OLR = 14 g BOD_5 m^{-2} day; Run 1: HLR = 32 L m^{-2} day^{-1} and OLR = 13 ; Run 2: HLR = 64 L m^{-2} day^{-1} and OLR = 13 g BOD_5 m^{-2} day^{-1}, Run 5: HLR = 32 L m^{-2} day^{-1} and OLR = 28 g BOD_5 m^{-2} day^{-1}. These results are reported by Ciuk Karlsson (2015).

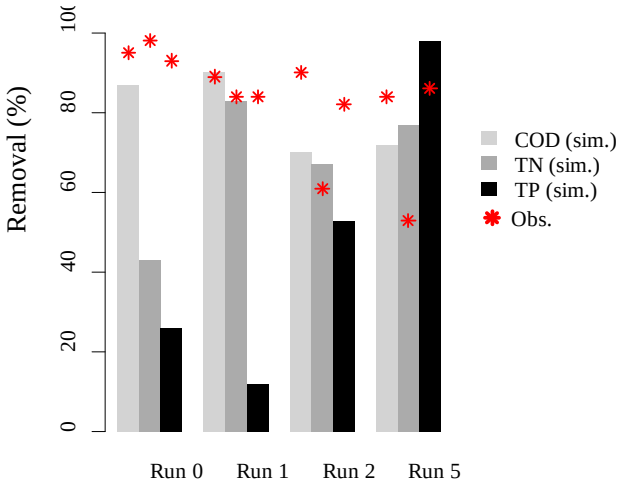

Figure 17. Simulated (sim.) and observed (Obs. *) removal (%) of chemical oxygen demand (COD), total nitrogen (TN) and total phosphorus (TP) at different loading regimes. Run 0: HLR = 32 L m^{-2} day^{-1} and OLR = 14 g BOD_5 m^{-2} day; Run 1: HLR = 32 L m^{-2} day^{-1} and OLR = 13 ; Run 2: HLR = 64 L m^{-2} day^{-1} and OLR = 13 g BOD_5 m^{-2} day^{-1}, Run 5: HLR = 32 L m^{-2} day^{-1} and OLR = 28 g BOD_5 m^{-2} day^{-1}. These results are reported by Ciuk Karlsson (2015).

Sensitivity analyses

The biochar filter model demonstrated inordinate sensitivity to the heterotrophic bacteria rate constant for lysis (bHet_20). The material parameter determining porosity (ths) also showed excessive sensitivity when decreased by 50% (Figure 18). In addition, the model

demonstrated sensitivity to the saturation/inhibition coefficient for hydrolysis (kh_20) (Figure 18).

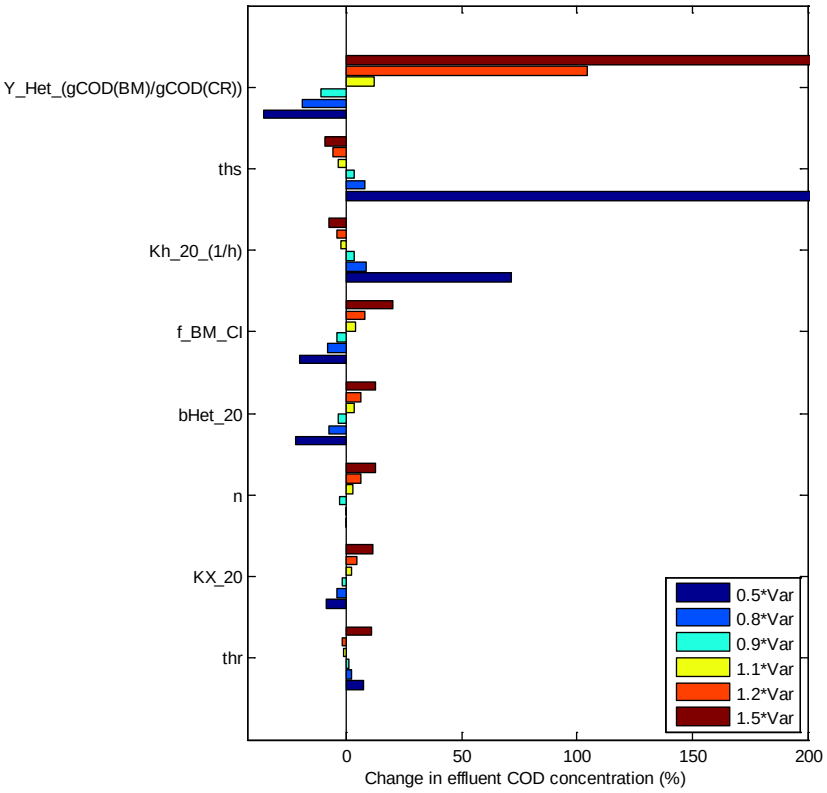

Figure 18. Sensitivity of the simulated chemical oxygen demand (COD) concentration in biochar filters in response to changes in model input parameters in the model. Data reported by Ciuk Karlsson (2015).

DISCUSSION

Hydrus software showed good agreement between observed and simulated parameters describing the flow dynamics of wastewater in biochar filters (Figure 13) and the CWD software showed promise in simulation of organic matter biodegradation in biochar filters, especially under high organic loading conditions. However, simulations using the CWD module failed to model nitrogen and phosphorus removal in biochar filters. The CWD module uses heterotrophic bacterial growth developed for suspended growth of bacteria in an active sludge model to estimate degradation of organic matter and nitrification. In filters, e.g. consisting of biochar, degradation of organic matter and nitrification usually occur in bacteria growing in biofilm (attached growth mode). The active sludge model parameters were not appropriate to estimate the growth of heterotrophic bacteria in biochar and hence biodegradation of organic matter was underestimated (especially for the organic loading conditions. Besides biodegradation, removal of organic matter in biochar is also achieved by adsorption of organic matter to the surface of the medium, as in removal of ammonium and phosphate. In this project, adsorption was not included in the simulation, as the intention was to optimise the model parameters for biodegradation and at later stages include the adsorption. Thus at this stage of the work, we only present the results for pollutant removal

by biodegradation. At a later stage, adsorption models will be added to the simulations to provide the whole picture of pollutant transfer in the biochar filters. The Hydrus wetland and CW3D modules demonstrated promising results with regard to simulation of flow dynamics with biochar filters and COD reduction. However, the CW3D module underestimated the nitrogen and phosphorus reduction achieved using the biochar material. Therefore, more work is needed to include the adsorption process in the model. This will improve simulations and enable use of the model to optimise filter depth, identify optimal loading conditions and evaluate clogging after long-term use of the filters, and hence determine biochar service life.

LONG-TERM PERFORMANCE OF BIOCHAR FILTERS SERVING SINGLE-FAMILY HOUSEHOLDS

BACKGROUND

A biochar filter system was constructed to treat the wastewater generated by a single family household. The biochar treatment system consisted of a septic tank followed by an aerobic biochar filter (Figure 19).

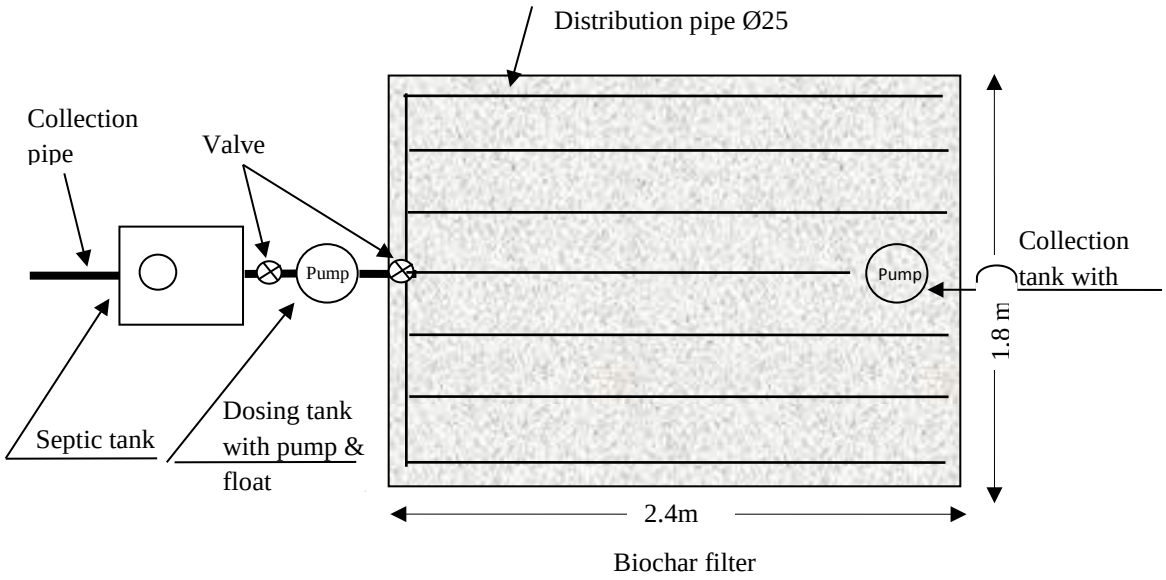

Figure 19. Schematic diagram of the biochar treatment system used for a single-family household, including septic tank, dosing equipment and biochar filter (Dalahmeh, 2016).

The wastewater was conveyed from the household to a single compartment septic tank by gravity. The septic tank effluent flowed to a submersible pump tank, from which effluent was pumped to a single-passage 60 cm biochar filter installed in a 1.8 m x 2.4 m x 0.6 m (width x length x depth) pit. Before adding the biochar, the pit was lined with a 2 mm thick plastic liner and a 15 cm gravel layer to function as a drainage layer. The biochar in the filter had particle size 1-5 mm and comprised waste fines from a biochar factory processing wood from different types of trees. The wet surface area of the biochar filter was 4.3 m^2 with a minimum and maximum surface loading rate of 46 and 186 L m^{-2} day^{-1}, respectively (mean 114 L m^{-2} day^{-1}). The average organic loading rate was 40 g BOD_5 m^{-2} day^{-1}. The treatment system was brought into operation on 26 March 2013.

Long-term (up to three years) performance of the biochar filter serving a single-family household was planned to be monitored during the period 2016-2018. The project team was

informed at the start that the family had moved from the house but used it occasionally, mainly during vacations in spring and summer. However, the project team collected water samples during the two periods when the family was staying in the house. The quality of the influent and effluent from the biochar filter was analysed and is summarised in Table 7. The results for wastewater samples collected on 29 March 2018 showed that the biochar filter was functioning effectively for organic matter removal (BOD_5; 91% removal) and Tot-N (75%) removal, but was not efficient in Tot-P removal (25%), despite a long-term pause in operation (more than 1.5 years). Unfortunately, no conclusion can be drawn regarding the long-term operation of the filter.

Table 7. Quality of wastewater in influent and effluent from household biochar filter serving single family. The samples were collected at different occasion during the period 2013-2018

Date	**19/05/2013**			**03/04/2014**			**31/08/2016**			**29/03/2018**		
Parameter	**Influent**	**Effluent**	**Percentage removal**	**Influent**	**Effluent**	**Percentage removal**	**Influent**	**Effluent ****	**Percentage removal**	**Influent**	**Effluent**	**Percentage removal**
BOD_5 (mg L^{-1})	474	7.2	99	464	14	97	563	11	98	273	24	91
TSS	164	9	95	124	21	83	162	20	88	–	–	–
Tot-N (mg L^{-1})	–	–	–	–	–	–	16	12	25	12.7	3.14	75
Kjeldahl-N (mg L^{-1})	–	–	–	–	–	–	15.4	11.7	24	–	–	–
NO_3-N	–	–	–	–	–	–	<0.3	<0.3	–	–	–	–
NH_4-N	72	<4.5	94	74	5	93	–	–	–	–	–	–
Tot-P (mg L^{-1})	8.57	2.5	71	2.9	4	–	1.75	0.81	54	1.69	1.24	27
E-coli (MPN 100mL^{-1})	$2.4x10^3$	$8x10^2$	0.5 log reduction	$1.3x10^3$	$1.3x10^4$	–	$1.4x10^7$	$1.7x10^2$	4.9 log reduction	$3.5x10^6$	$2.4x10^4$	2.2 log reduction

– Not measured

** The effluent sample collected on this date is not representative of effluent from the biochar filter.

FUTURE RESEARCH

Impregnation of biochar filters with calcium oxide and iron chloride enhanced removal of total phosphorus and phosphate-phosphorus in small-scale onsite wastewater treatment systems. The iron- and calcium-impregnated biochar filters did not reach saturation level during the experimental period, but their capacity to bind phosphorus can be expected to be finite and they may also become saturated with other pollutants (e.g. organic matter, ammonia and trace metals). Thus, it is recommended that these types of impregnated biochar be used in a separate phosphorus filter module, where solids and organic matter are removed from the wastewater before it is treated in the impregnated biochar. Research is ongoing to monitor the performance of these types of impregnated biochar over extended periods and weekly samples are being collected and analysed. The results will be presented in a coming report from the project. We also plan to produce a manual containing dimensioning criteria and design and construction guidelines that can be used by interested householders or contractors interested in implementing such systems.

A combined vertical-horizontal flow biochar filter system was efficient in organic matter removal from wastewater and showed promising performance regarding nitrogen removal in onsite wastewater treatment. However, the performance of the combined filter under different loading conditions is under investigation and will be reported in an upcoming report in this series. In addition, the design of the filter system with respect to depth:width:length of the horizontal flow system has not yet been investigated and the recommended loading conditions still need to be tested and optimised.

The Hydrus module combined with the CW3D module demonstrated promising results with regard to simulation of flow dynamics with biochar filter and chemical oxygen demand reduction. However, the CW3D module underestimated the nitrogen and phosphorus reduction achieved using the biochar material. Therefore, more work is needed to include the adsorption process in the model. This will improve simulations and enable use of the model to optimise filter depth, identify optimal loading conditions and evaluate clogging after long-term use of the filters, and thus determine the service life of biochar filters.

Data on long-term performance of the biochar filters in the field are lacking and results from trials with only one field biochar filter are not sufficient to enable conclusions to be drawn on the performance of biochar filters. However, the biochar filter concept has now been adopted by private contractor Jim Johannson (Skånske Hustech) and another biochar filter is currently under construction in Perstorp kommun. Once construction is completed, the system will be monitored and the quality of the treated wastewater will be assessed.

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