Sveriges lantbruksuniversitet
Swedish University of Agricultural Sciences

Havs och Vatten myndigheten

Institutionen för energi och teknik

Capacity of biochar filters for wastewater treatment in onsite systems – Technical Report

Biokolfilters kapacitet för små avloppsvattenrening – 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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SAMMANFATTNING

I områden utanför tätorter där fastigheterna inte är anslutna till kommunala reningsverk sker rening i enskilda och småskaliga avloppssystem. . De flesta reningssystem består i huvudsak av en slamavskiljare och markbädd eller infiltration, medan ett reningssteg för fosfor saknas i de flesta anläggningarna. Ett stort antal av systemen har dåligt funktion p.g.a ålder och/eller otillräckligt skötsel. Små avloppsanläggningar (upp tom 200 pe) bidrar i hög grad till fosfor- och kväveutsläpp till Östersjön (HELCOM, 2005). Utsläppet av fosfor från små avloppssystem är lika stort som fosforutsläppet från alla kommunala reningsverk i Sverige, trots att endast 15 % av svenska befolkningen är anslutna till små avloppsanläggningar (Havs- och vattenmyndigheten, 2016).

Biokol är pyrolyserat organiskt material som kännetecknas av hög porositet och stor specifik yta med många ytaktiva bindningsplatser. Dessutom har biokol låg vikt och är lätt att transportera. I ett avslutat projekt vid Institutionen för energi och teknik vid Sveriges lantbruksuniversitet har rening med hjälp av biokolbäddar för små avloppsanläggningar under olika belastningsvillkor och kornstorlek utvärderats. Denna rapport sammanställer resultaten och slutsatserna kring kapaciteten hos biokolfilter att rena avloppsvatten från hushåll. Biokolsbehandling av avloppsvatten bygger på fysiska och biologiska processer, d.v.s. adsorption samt biologisk nedbrytning och omvandling av organiska föroreningar och kväve. Biokol har stor specifik yta jämfört med t.ex. sand samt har högre andel av mikro- och makroporer, som möjliggör effektiv avskiljning av partiklar, adsorption av organiska och oorganiska ämnen och påväxt av biofilm för biologisk nedbrytning.

Syftet med detta projekt var att förenkla och kvalitetssäkra användningen av biokol i markbäddar så att biokol på ett säkert och hållbart sätt kan ersätta eller komplettera befintliga markbäddar i små avloppsanläggningar. På lång sikt förväntas projektet bidra med kunskap för att förbättra reningskapaciteten i små avloppssystem och bidra till miljökvalitetsmålen Ingen övergödning och en Giftfri miljö. Projektets mål var att producera en underlagsrapport som jämför reningsfunktionen mellan biokol- och markbäddar. Den tekniska underlagsrapporten förväntas att fungera som ett verktyg som användare och miljöinspektörer kan använda vid val av bäddmaterial i småskaliga avloppsanläggningar, vid rådgivning och tillståndsgivning samt som underlag för entreprenörer. Projektet har genomfört följande aktiviteter:

1. Sammanställning av genomförda analyser på biokols reningsförmåga med avseende på BOD, COD, fosfor, kväve, salmonella och fekala koliformer, samt fysiska, kemiska, geometriska och hydrauliska egenskaper.

2. Litteraturstudier om biokol och dess fysiska och biologiska egenskaper och struktur, samt en litteraturstudie om förväntad långsiktig reningsförmåga hos biokol.

3. Författande av en teknisk underlagsrapport som jämför reningsfunktionen mellan biokol- och markbäddar.

Den tekniska underlagsrapporten sammanställer resultaten från tidigare och pågående forskning vid kretslopptekniksgruppen, SLU, avseende kapaciteten hos biokolfilter för vattenrening i små avloppssystem. Data som presenteras i rapporten erhölls från fem olika forskningsförsök med biokolfiltrering i laboratoriemiljö och ett fullskaligt försök för en anläggning för en familj. Labbförsöken pågick över olika tidsperioder som varierade från 3 månader till 1,8 år. Anläggningen har varit i drift i mer än 2,5 år (rening i filtret pågår sedan våren 2014). I denna studie presenteras resultaten om biokols förmåga att rena avloppsvatten från BOD_5, BOD_7, COD, NH_4 och NO_3, Tot-N, PO_4, Tot-P, E. coli, enterokocker, och bakteriofager (MS2 och PhiX). Dessutom studerades effekter av partikelstorlek (0,7; 1,4; 2,8 och >5 mm) och hydraulisk belastning (32, 40 och >200 200 L / m^2,dag) för biokolets förmåga att minska ovan nämnda ämnen. Biokolfilter jämfördes med markbädd, det vanligaste filtermaterialet i små avloppsanläggningar.

Studien visade att biokol har god kapacitet för att avlägsna organiskt nedbrytbart material (BOD_5; > 90%, COD; > 90%) under olika hydraulisk belastning och kornstorlekar under såväl lång som kort tid. Reningsförmågan hos biokol vad gäller organiskt material (COD) skilde sig inte mellan biokolfilter med olika kornstorlekar (0,7 mm, 1,4 mm, 2,8 och > 5 mm) och inte heller mellan olika hydrauliska belastningar (32-37 och >200 L/ m^2 dag).

Ammoniumadsorption i biokol var effektiv (90-99 %) och minskningen av totalkväve varierade mellan 62 och 88 % beroende på partikelstorlek och hydraulisk belastning. Hög hydraulisk belastning (200 L per m^2 per dag) och stora partiklar (> 5 mm) visade den högsta reduktionen av totalkväve (70-80 %).

Reduktionen av fosfor varierade mellan olika typer av biokol; aktiverat biokol visade 89-90 % reduktion av fosfor, salix-baserat icke-aktiverat biokol hade ca 89 % reduktion av fosfor, medan andra icke-aktiverade träbiokol visade 32-66 % reduktion. Minskning av E.coli och *Enterokocker faecalis* varierade med partikelstorlek och den största minskning var 4.5 log 10-reduktion som uppmättas för 0,7 mm partikelstorlek och en hydraulisk belastning på 32 L per m^2 per dag. Samtidigt mättes den sämsta reduktionen (<1 log 10-reduktion) för >5 mm partikelstorlek och en hydraulisk belastning 200 L/ m^2 dag.

Jämfört med markbädd visade resultaten att biokol har bättre reningsförmåga för BOD_5, COD och kväve än sandfilter som drevs med liknande partikelstorlek och hydraulisk belastning. Fosforreningen i aktiverat biokol är mycket bättre än i sandfilter, medan icke-aktiverat biokol och sand betedde sig likartat vad gäller fosforrening.

Vid behandling av avloppsvatten från svenska hushåll rekommenderar vi att biokolfilter (aktiverat och icke-aktiverat) används för att avlägsna organiskt material (BOD_5 och COD) vid partikelstorlekar mellan 1,4 och 5 mm, och hydraulisk belastning (upp till 50 L per m^2 per dag). För optimal reduktion av bakterier är det lämpligt att använda biokolfilter med liten kornstorlek (0,7-1,4 mm).

Fler studier krävs för att identifiera de kemiska och strukturella egenskaper hos biokol som ligger bakom en effektivare reduktion av fosfor. Dessutom kan denitrifikation i biokol

potentiellt förstärkas/optimeras genom att bygga in ett anaerobt skikt av biokol, eller genom horisontellt flöde i filter och detta bör undersökas i ytterligare forskning.

Vidare studier bör innefatta att optimera och designa biokolfilter för att förbättra kväve- och fosforreduktion från avloppsvatten. Detta kan göras genom laboratorieförsök med anaeroba biokollager eller biokol med horisontellt flöde. Modellering av föroreningstransport och nedbrytning i mjukvaran Hydrus kan komplettera försöken.

1. INTRODUCTION

There is a great need for simple and robust systems for high-quality treatment of wastewater at both local and global scale. In Sweden, about one million people (10% of the population) live in rural areas not connected to a public sewerage system, and instead use around 675,000 onsite wastewater systems (SMED, 2011). A typical onsite wastewater treatment system is composed of a septic tank followed by a sand-filter trench or soil infiltration system (Pell, 1991; WRS, 2009). These systems are often poorly functioning in treating the wastewater. Centralised treatment systems with efficient treatment performance are not economically feasible for these sparsely populated areas. Consequently, more cost-effective decentralised and/or individual household treatment solutions are needed.

Household wastewater is composed of blackwater (faecal matter and urine) and greywater (water from dishwashing, laundry, showering and cleaning). Domestic wastewater contains different types of pollutants such as easily degraded organic matter (BOD_5), nitrogen, phosphorus, faecal bacteria and even pathogens (viruses, bacteria and protozoa) in the event of infections among household inhabitants. Treatment of wastewater in onsite systems is essential to protect water resources from eutrophication and to protect public health by preventing the spread of microbiological contaminants and nitrate to drinking water wells close to onsite treatment systems. The NFS 2006:7 recommendations for levels of pollutant removal from onsite systems require at least 90% reduction of organic substances (BOD_7) and at least 70% reduction of phosphorus for areas with normal protection level. For environmentally sensitive areas, at least 90% reduction of phosphorus and 50% reduction of nitrogen are required (Naturvårdsverket, 2006).

The current quality of the wastewater effluent from onsite systems might not be in full compliance with the environmental recommendation regarding phosphorus. For example, although only 15% of the Swedish population is connected to onsite wastewater treatment systems, the phosphorus discharge from these systems exceeds the gross discharge of phosphorus from all municipal wastewater treatment plants in Sweden (Havs- och vattenmyndigheten, 2016). Contamination of private and individual drinking water wells by wastewater pollutants from onsite systems is another issue. During the period 1975-1991, 40 cases of drinking water contamination were reported in Sweden and at least 12 people were affected in each case (Stenström et al., 1994). The most common cause of groundwater contamination reported for private wells is cross-contamination with wastewater from onsite wastewater treatment systems in the surroundings of the well. A survey carried out in 2007 by the Swedish Board of Health found that about 20% of all water samples from private drinking water wells were not fit for purpose (Socialstyrelsen, 2008). Furthermore, it was noted that microbiological contamination was the most common form of drinking water contamination (Socialstyrelsen, 2008). Compared with effluents from municipal systems, onsite systems can be free of pathogens for most of the year, but can contain very high levels of pathogens in the event of gastrointestinal infection within the household/s connected to the treatment system. Typical diseases that can be spread through onsite wastewater facilities are pathogens causing stomach diseases such as Norwalk virus, rotavirus and salmonella (Ottoson, 2013).

Sand filters, which are within the fine macropore range, are the most commonly applied filters for onsite treatment of greywater (Burnat & Eshtaya, 2010; Friedler & Hadari, 2006; Suleiman et al., 2010) and mixed household wastewater (Pell, 1991; US EPA, 2002). Besides physical filtration through the sand, an active biofilm develops and attaches to the sand particle surfaces and mineralises organic matter from the wastewater (Rodgers et al., 2005). Clogging problems (Spychała & Błazejewski, 2003), scarcity of well-graded sand in some regions and high transportation costs due to the high bulk density are the main obstacles to using sand filters. In addition, the high bulk density means that major efforts are required for transporting virgin sand and recycling or disposing of spent sand.

Biochar is a material of organic origin (forestry or agricultural by-products) charred at elevated temperature at the absence of oxygen. The pyrolysis of organic substances produces gas and organic liquid, but leaves pure carbon (biochar). Activated biochar is a biochar-type substance that has been treated by different possible processes in order to increase the specific surface area. After initial pyrolysis, biochar can usually be activated by gasification with oxidising gases such as CO_2, steam or air, or by addition of zinc salts or phosphoric acids (Downie et al., 2009). Activation of biochar usually results in increasing porosity and specific surface, which enhances the treatment capacity of the materials.

The term ‘biochar’ is applied when the charred material is used for soil amendment as a carbon sink or for filtration of percolating water. The first evidence of biochar use in history goes back to the Amazonian dark earth (*terra preta*) that formed as a result of indigenous settlements in Brazil (Steiner, 2007). Research has long documented positive effects of biochar addition to soil on plant growth, but its use for environmental management on a global scale is quite recent (Amonette & Josheph, 2009). The objectives of biochar application for environmental management are soil improvement, waste management, climate change mitigation and energy production (Igalavithana et al., 2016; Lal, 2016; Miles et al., 2016; Saarnio, 2016; Sizmur et al., 2016). Forestry, agricultural wastes and other by-products can be used as a resource for pyrolysis, resulting in biochar (He et al., 2016).

Pyrolysis with or without activation converts organic wastes into material with beneficial properties which can serve as an adsorbent and biofilm carrier for wastewater treatment. Under the name ‘charcoal’, biochar has been studied and used for the adsorption of different pollutants, such as heavy metals (Babel, 2004) and aromatic hydrocarbons (Mukherjee et al., 2007). Use of biochar for onsite wastewater treatment started more recently, initially in low income countries (Niwagaba et al., 2014).

Wastewater pollutants have difference mechanisms and conditions for their removal. For example, biological oxygen demand (BOD) is removed by biological degradation; phosphate is removed by adsorption and chemical precipitation and ammonia is transformed under aerobic conditions into nitrate, while nitrate is denitrified under anoxic conditions. Thus, successful removal of these different types of pollutants requires special capabilities of the treatment medium, with specific properties that combine adsorption and biological degradation with co-occurrence of aerobic and anaerobic conditions.

The capacity of filters to remove pollutants differs between materials due to different characteristics such as porosity, specific surface area and reactivity, adsorption capacity and ability to promote biofilm development (Rolland et al., 2009). In addition, wastewater production in households can vary on a daily, weekly and seasonal basis, which leads to variability in organic and hydraulic loading rates to the wastewater treatment system of households. Under peak conditions, fluctuating hydraulic and organic loads can lead to a temporary breakdown of the infiltration system, so-called episodic failure (Beal et al., 2008). Thus, it is necessary for the infiltration beds to have the capacity to withstand variations in hydraulic and organic loading and maintain resilient and steady treatment performance. This requires knowledge about the capacity of the particular filter material to buffer high variations in water flow and organic loading. This report summarises knowledge gained at Department of Energy and Technology at the Swedish University of Agricultural Sciences regarding the performance of biochar filters in removing various types of pollutants from wastewater under different loading conditions for different types and properties of biochar.

2. AIMS AND OBJECTIVES

The aim of this report was to describe the performance of biochar filter beds in wastewater treatment with regard to removal of various types of wastewater pollutants (organic matter, solids, phosphorus, nitrogen and pathogen indicators) and to assess the potential of biochar filter beds for use in onsite wastewater treatment instead of, or as a complement to, sand filter beds.

The specific objectives of this report were to:

1- Describe the physical, chemical and hydraulic properties of biochar and assess the effects of these properties on the performance of biochar filters for onsite wastewater treatment, in comparison with sand filters.
2- Demonstrate and assess the performance of biochar filters in wastewater treatment under different organic and hydraulic loading regimes, in comparison with sand filters.
3- Describe the performance of biochar filters of different particle sizes in wastewater treatment.
4- Describe the performance of a household biochar filter in treatment of wastewater from single family household during start-up and in the first two years of operation.

3. MATERIALS AND METHODS

3.1 Experimental setup of the laboratory-scale biochar and sand filters

Laboratory-scale biochar and sand filters were installed and operated under controlled conditions. Three types of biochar and one type of sand were investigated in the studies described in this report and their performance in wastewater treatment in small-scale wastewater treatment was evaluated and compared with that of sand. The filter materials comprised:

1. Willow (*Salix*) biochar which originated from chopped willow, a broadleaf tree with low-density wood. The willow was grown in Germany and charred at 450°C. The material was tested in filters with effective particle size (d_{10}) = 1.4 mm and uniformity coefficient = 2.2.
2. Hardwood biochar of undefined wood origin. The material was tested in various filters with differing effective particle size (d_{10} = 0.7, 1.4 and 2.8 and >5 mm) and uniformity coefficient = 2.2. The specific surface area of this material was 170-200 $m^2\ g^{-1}$.
3. Activated biochar in a mixture of 1.5 mm pelleted activated biochar with diameter 1.5 mm and length 3-5 mm. The specific surface area of both carbon fractions was >1000 $m^2\ g^{-1}$, with effective size = 1.4 mm and uniformity coefficient = 2.2.
4. Sand with particle effective size = 1.4 mm, uniformity coefficient = 2.2 and specific surface = 0.15 $m^2\ g^{-1}$. The sand was obtained from Rambo Jordi (Rambo, Sweden).

All laboratory-scale filters were installed as column filters, with filter depth 55-60 cm (Figure 1). Depending on the study objective, the diameter of the laboratory filters was 200, 75 or 50 mm. All studies used triplicate filters for each material except one study with activated biochar, in which duplicate activated biochar filters and duplicate sand filters were tested.

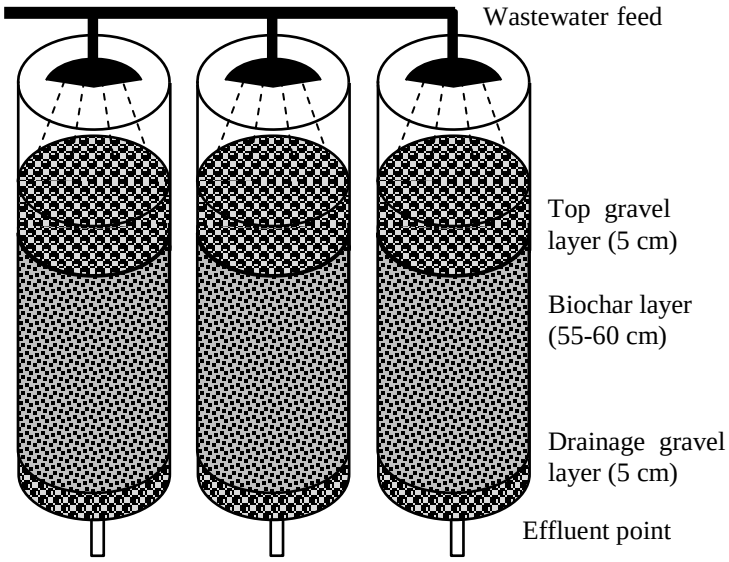

Figure 1. Schematic diagram of the laboratory-scale filters used for testing wastewater treatment effects.

Before using a material as a filter medium, the following properties of the material and the filter were identified: water content, loss on ignition, effective size and uniformity coefficient of the filter, specific surface area, internal structure, surface topography, surface composition and particle density of the material. After preparing the filters using the different biochar and sand materials, the following filter properties were identified: bulk density, total porosity and hydraulic residence time of the filters. Constant head hydraulic conductivity was determined only for the sand and activated biochar filters. Methods used for determination of the properties listed are described in the following sections.

Synthetic wastewater and real wastewater were used to test the performance of the biochar and sand filters in wastewater treatment. In three of the studies included in this report, the

synthetic wastewater used was of similar strength (in terms of concentrations of pollutants) to real wastewater from Kungsäng wastewater treatment facility in Uppsala. In a further two studies, the synthetic wastewater was designed to represent low strength (low polluted) wastewater and high strength (highly polluted) wastewater. The synthetic wastewater was composed by mixing nutrient broth, different types of detergent, real wastewater and bacterial mixtures cultivated in the laboratory. The synthetic wastewater was used in order to control the quality of the test wastewater and to ensure repeatability of the quality, especially since the studies were not all performed at the same time. In addition to studies with synthetic wastewater, one study was carried out using real wastewater from the Kungsäng wastewater treatment plant. In that study, biochar was compared with a sand filter.

The most common loading (feeding) of wastewater into sand filter beds in small-scale (household) wastewater treatment is down-flow, non-saturated and intermittent feeding regime. Thus all the biochar and sand filters used here were fed with wastewater using a down-flow, non-saturated and intermittent loading regime. The performance of biochar filters for small-scale wastewater treatment was tested for different biochar particle sizes (d_{10} = 0.7, 1.4, 2.8 and >5 mm) at three organic loading rates (OLR = 5±2, 20±5 and 70 g BOD_5 m^{-2} day^{-1}) and two hydraulic loading regimes (HLR = 32±5 and 200 L m^{-2} day^{-1}). Table 1 shows the detailed variables tested for each type of biochar (d_{10}, HLR, OLR and type of wastewater), the duration of the experiment and the pollutants analysed.

3.2 Household biochar filter

A single-family 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 2).

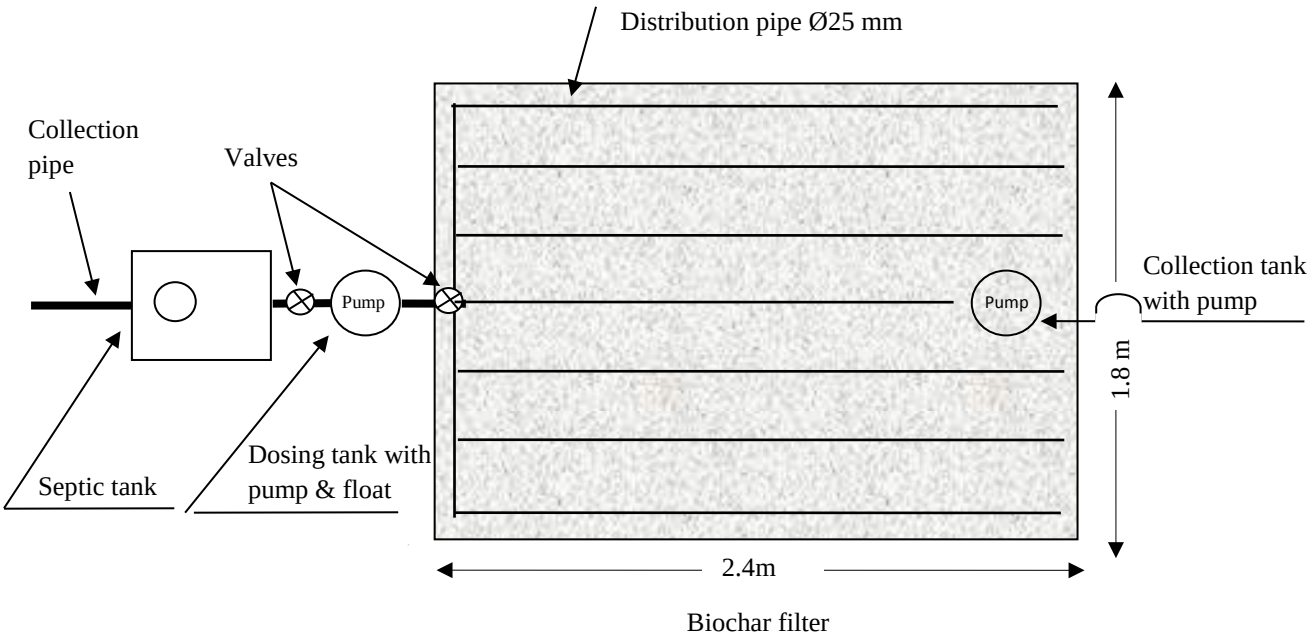

Figure 2. Schematic diagram of the biochar treatment system used for a household, including septic tank, dosing equipment and biochar filter.

Table 1. Variables tested to assess the performance of biochar filters for small-scale wastewater treatment in the six different studies included in this report

	Study 1	Study 2	Study 3	Study 4	Study 5	Study 6
Type of material	Non-activated willow wood biochar Activated biochar	Non-activated hardwood biochar Activated biochar	Non-activated hardwood biochar	Non-activated hardwood biochar	Activated biochar Sand	Non-activated hardwood biochar Sand
Effective size (d_{10}, mm)	1.4	1.4	0.7, 1.4 and 2.8	1.4, 2.8 and >5	1.4	1.4
Hydraulic loading rate (L m^{-2} day^{-1})	34	34	34	200	Fluctuating (32, 64 and 128)	37±7
Organic loading rate (g BOD_5 m^{-2} day^{-1})	15	70	20±5	5±2	Fluctuating (15, 28 and 70)	5±1 (BOD_7 basis)
Type of wastewater	Synthetic wastewater	Synthetic wastewater	Synthetic wastewater	Real	Synthetic wastewater	Real wastewater
Chemical pollutants	COD, NH_4, surfactants (MBAS*), NO_3, Tot-N, PO_4-P and Tot-P	COD, NO_3, Tot-N, PO_4-P and Tot-P,	COD, BOD_7, NO_3, Tot-N, PO_4-P and Tot-P	COD, BOD_5, NH_4, NO_3, Tot-N,	COD, BOD_5, NO_3, Tot-N, PO_4-P and Tot-P	COD, BOD_7, NO_3, Tot-N, PO_4-P and Tot-P
Microbiological pollutants		Salmonella spp., *Enterococci faecalis*, PhiX-174, MS2	*E. faecalis*, *Escherichia coli*, PhiX-174, MS2	E. *faecalis*, *E. coli*, yeast, PhiX-174, MS2		
Filter operation period	20 weeks	20 weeks	6 months	4 months	6 months	6 months

*Methylene blue active substances.

The wastewater was conveyed from the household to a 200 L 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 filling with 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 filter had a particle size of 1-5 mm and comprised waste fines from a biochar factory processing wood from different types of trees. The septic tank effluent was pressurised and distributed over the biochar through perforated pipes (diameter. 25 mm) laid over the biochar. Perforations (diameter. 3 mm) were drilled every 30 cm along the pipes to ensure even distribution of the wastewater over the filter. The distribution pipes were covered with coarse gravel to prevent wastewater splashing and to decrease evaporation. The wet surface area of the biochar filter was 4.3 m^2 and the daily flow rate ranged between 200 and 800 L day^{-1}, with an average flow of 490 L day^{-1}, yielding 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}. Following infiltration, the wastewater was collected in a tank fitted with a pump (see Figure 1). The treatment system was started on 26 March 2013 and is still in operation at the time of writing (May 2016).

3.3 Sieve analysis, uniformity coefficient and effective size

In order to have particle sizes that were comparable to those commonly used in sand filters, non-activated biochar was sieved on a stack of sieves with mesh openings of 7, 5, 3, 2, 1 and 0.8 mm placed on the mechanical shaker (Retsch, Haan, Germany). The uppermost sieve was loaded with four cups (around 170 g) of pre-sieved biochar material. The shaker was run at 30 rpm for 10 minutes and the fractions retained on the sieves were selected to be packed in the cylinders. The different types of biochar were air-dried before being sieved through screens. The activated biochar was obtained in the form of pellets, which were crushed to obtain particle size less than 1.5 mm, and the 1.5 mm, 3-5 mm and crushed fractions were mixed in a 1:3:1 ratio to obtain a uniformity coefficient and effective size similar to that of the sand.

The effective size (d_{10}) was determined as the size of screen opening at which 90% of a biochar sample was retained on the screen and 10% passed through. The uniformity coefficient (Uc), which is a numerical estimate of how the filter material is graded, was calculated by dividing the d_{60} value (the size of screen opening where 60% of a sample passes through and 40% is retained) by d_{10} (effective particle size).

3.4 Determination of physical properties

In the biochar studies, water content (w), loss on ignition, bulk density, particle density, total porosity and specific surface area of the different types of the biochar and of the sand material used for comparisons were determined.

3.4.1 Water content

The water content (w) was determined by drying the biochar material for 24 h in a furnace at 105°C. The mass of water was calculated by subtracting the weight of the oven-dried material from the weight of the air-dried material. Loss on ignition was determined at 550°C for 4 h

(Wright et al., 2008). The water content of the air-dried filter materials was determined on a dry matter basis by applying the formula:

$$w = M_w / M_s \quad \text{(eq. 1)}$$

[w]	g g^{-1}	gravimetric water content
[M_w]	g	mass of water
[M_s]	g	mass of solids

3.4.2 Bulk density and particle density

Bulk density was determined by dividing the dry weight of the filter medium by the volume occupied by the medium. Particle density of non-activated biochar, activated biochar and sand was determined by dividing 25 g sample by the corresponding volume of particles excluding pores. Volume of particles excluding pores was determined using the liquid immersion method, where the volume of deionised water displaced by the particles was measured. Air-filled pores were eliminated by gentle boiling of the mixture. The submerged particles were left for saturation for 24 h.

The particle density of solids (ρ_s) was determined by applying the formula:

$$\rho_s = M_s / V_s \quad \text{(eq. 2)}$$

[ρ_s]	g cm^{-3}	particle density
[M_s]	g	mass of solids
[V_s]	cm^3	volume of solids

3.4.3 Specific surface area

Specific surface area of the different types of biochar and the sand was determined using the Brunauer-Emmett-Teller (BET) method (Brunauer et al., 1938). The BET equation was used to calculate the specific surface area of bark, biochar and sand based on measurements at 99,834 Pa and 20°C (Flowsorb II 2300, 1996), where 1 mL N_2 gas corresponded to 2.86 m^2 (Brunauer et al., 1938). A kaolinite sample with a defined area of 15,900 m^2 kg^{-1} was used as the control (Brunauer et al., 1938).

3.4.4 Internal structure, surface topography and chemistry

The internal structure, surface topography and surface chemistry of the hardwood non-activated biochar, activated biochar and sand were identified using elemental scanning electron microscopy (SEM). The SEM micrographs and energy dispersive X-ray spectrographs (EDS) of the samples were obtained using a HITACHI TM-1000 scanning electron microscope equipped with an Oxford Instruments EDX detector. To obtain reliable statistics in the elemental analysis, the value used for each point was the average of three individual measurements. The scanned surface was mapped by moving over the sample with steps of 10 μm.

3.5 Determination of hydraulic properties

3.5.1 Porosity

Total porosity of the filters was determined using two approaches. The first approach was based on the particle density and bulk density of the filters using the formula:

$$f = 1 - \frac{\rho_b}{\rho_s} \qquad \text{(eq. 3)}$$

[f]	$cm^3\ cm^{-3}$	porosity
[ρ_b]	$g\ cm^{-3}$	bulk density
[ρ_s]	$g\ cm^{-3}$	particle density

The second approach was based on the amount of water required to fill the pores inside the filter.

3.5.2 Hydraulic conductivity and hydraulic residence time

Constant head hydraulic conductivity was determined for all the active biochar filters and the sand filters used in laboratory studies, according to Jacob et al. (2002). For all the biochar filters except the household filter, the residence time was also determined. Initially the filters were fed with distilled water. Thereafter, a suitable pulse of 1% sodium chloride solution was added as a tracer for residence time measurements and the electrical conductivity (EC) of the outflow water from the filters was measured as a function of time water using a Conductivity Pocket Meter (WTW, Germany). Each time the EC in the effluent was measured, the respective outflow volume was recorded. The recorded effluent volume was multiplied by the measured EC and the values added together over time. The total EC of the tracer added to each filter was 776.2 mS mL^{-1} cm^{-1}. The residence time of the tracer within a material was calculated as the mean of the cumulative EC value of the three filters of one filter material, divided by 776.2 mS cm^{-1} and then plotted. The time it took to recover 50% of the tracer was taken as mean residence time.

The shortest hydraulic residence time, defined as the time lapse between the wastewater dosage and the first outflow from the filters, was repeatedly determined throughout the experiment for the laboratory-scale filters. It was used as an indicator of clogging of the internal pores in the filters due to accumulation of biofilm materials and solid deposits.

3.6 Chemical analysis

3.6.1 Laboratory-scale filters

For the studies using laboratory-scale filters, samples of the inflow and outflow were collected for chemical analysis in all studies included in this report. The following chemical parameters were determined with a frequency of twice to once per week: NH_4-N, COD, MBAS, NO_3-N and Tot-N, EC, pH, PO_4-P and Tot-P.

The parameters were determined using chemical kits and according to methods shown in Table 2. The chemical kit methods are in accordance with the standard APHA methods (APHA, 2007). The analytical quality was ensured by using control solutions with known

concentrations of the substance for every measurement series (specified in Table 2). For the EC determination, adjustment due to temperature deviation was needed. Nonlinear temperature compensation was selected on the EC meter and therefore EC values were already temperature-adjusted. The pH meter had an integrated thermometer and pH values were also temperature-adjusted automatically.

3.6.2 Household filter

To evaluate the performance of the household biochar filter during the initial phase, 10 samples each of influent and effluent were collected weekly during the first three months of operation. After one year, four additional samples of influent and effluent were collected on four occasions separated by one-week intervals, to monitor the performance of the filter after it had fully established. The samples were collected as grab samples from the influent, from the septic tank effluent and from the biochar filter effluent. All analyses were performed according to Standard Methods for the Examination of Water and Wastewater (APHA, 1998) using the following protocols: pH (4500-H and B), biochemical oxygen demand (BOD_5; 5210-B), total suspended solids (TSS; 2540-D), total phosphorus (Tot-P; 4500-P) and ammonium (NH_4; 4500-NH_3/B and C).

Filter efficiency in reducing the measured substances was calculated with the formula:

$$E = \frac{C_{in} - C_{out}}{C_{in}} \quad \text{(eq. 4)}$$

[E]	-	efficiency
[C_{in}]	mg L^{-1}	influent concentration
[C_{out}]	mg L^{-1}	effluent concentration

3.7 Microbiological analysis

3.7.1 Laboratory-scale filters

For the purposes of studying the capacity of the biochar for removal of human pathogens from wastewater, the following bacterial indicators were measured in the influent and effluent of the filters: *Escherichia coli* (*E. coli*), *Enteroccocci faecalis* (*E. faecalis*) and *Salmonella* spp. In addition, two model bacterial viruses (phages), MS2 and PhiX-174, were used to assess the removal of human viruses (rotavirus and norovirus) from the wastewater. Removal of the protozoa C*ryptosporidium paravum* by biochar filters was assessed by testing the removal of the eukaryote yeast *Saccharomyces cerevisiae* as a model. The wastewaters used for testing the laboratory-scale filters were continuously spiked with known concentrations of *E. coli, E. faecalis, Salmonella* spp., MS2, Phix-174 and *Saccharomyces cerevisiae.* The reason for spiking was to ensure stable concentrations of the microorganisms, in order to allow proper evaluation of the treatment capacity for microbial contaminants.

Buffered NaCl peptone water with Tween (pH 7) was used for the dilution, which was matched to the expected detection level of the microorganisms in the wastewater and the effluent from different materials.

Slanetz & Bartley agar (SlaBa) plates were used as cultivation subtract for *E. faecalis* (ATCC 29212) and were incubated at 44°C for 48 h. Xylose lysine deoxycholate (XLD) plates were used to grow *Salmonella* spp and were incubated at 37°C for 24 h. Enumeration of the bacteria was performed by counting the colony-forming units (CFU). The soft agar solution was melted in a microwave and then kept at 55°C to avoid solidification.

Blood agar (BAB) plates were used as the cultivation subtrate, the MS2 phage with host bacteria WG 49 (ATTC 700730), and PhiX-174 phage with host bacteria *E. coli* (ATCC 13706). The bacteria were cultivated in nutrient broth for 3-5 h at 37°C before sampling. One mL of the host strain and 1 mL of the virus sample were added to 2 mL of the soft agar in an assay tube placed on the heating block to prevent the soft agar solidifying. The mixture was poured on the BAB plates and incubated at 37°C overnight. Enumeration of bacteriophages was performed by counting the plaque-forming units (PFU) with their respective host bacteria. Duplicate plating was applied for each microbial sample.

Rich yeast extract, peptone, dextrose (YPD) medium was used for growing *Saccharomyces cerevisiae* under non-selective conditions at concentrations of 1% yeast extract, 2% peptone and 2% glucose at 100 mg L^{-1} chloramphenicol. Enumeration of *Saccharomyces cerevisiae* was performed by counting the colony-forming units (CFU) with their respective host bacteria. Triplicate plating was applied for each yeast sample.

3.7.2 Household filter

To evaluate performance of the household biochar filter in removal of pathogen indicators and bacterial viruses, 10 samples each of influent and effluent were collected weekly during the first three months of operation. Four additional samples of influent and effluent were collected after one year of operation on four sampling occasions separated by one-week intervals. In addition, two extra samples were collected in November 2015 to analyse *Salmonella* in the influent and effluent of the household filter. The samples were collected as grab samples from the influent, from the septic tank effluent and from the biochar filter effluent. The microbiological analysis was performed according to standard methods for the examination of water and wastewater (APHA, 1998) using the following protocols: *Escherichia coli* (9221-F), faecal enterococci (9230-B), *Salmonella typhi* (9260-B), male-specific coliphages (MS2) and somatic coliphages (9224-E).

Removal of bacteria, phages and yeast was estimated using the equation:

$$Reduction\ (Log_{10}\ 10) = Log_{10}(C_{in}) - Log_{10}(C_{out}) \text{ (eq. 5)}$$

Table 2. Chemical kits, their ranges and methods; chosen control solutions, apparatus and measurement days used for the analysis

Substance		Kit name	Measurement range	Units	Standard method	Control solution name and value	Apparatus
EC	Electrical conductivity			mS cm^{-1}		Calibration liquid	Conductivity Pocket Meter, Cond340i WTW, Germany
pH						Calibration liquid: pH 7 and pH 9	pH-meter Ino Lab pH Level 1, WTW pH-electrode Blueline 14 pH, Schott instruments
COD	Chemical oxygen demand	Spectroquant COD Cell Test (Hg-free) 1.09772.0001 and 1.09773.0001	10-150 and 100-1500	mg L^{-1}	No standard, but Hg-free	Potassium hydrogen phthalate solution 1.11769.0100, Merck 170 mg L^{-1} and Combi R1, Combicheck 20 1.14675.0001, Merck 750±75 mg L^{-1}	Thermoreactor TR 420, Merck, Germany Spectroquant NOVA 60, Merck, Germany Pipettor, VWR, Poland Analog Vortex Mixer, VWR, USA
MBAS	Anionic surfactants	Spectroquant Surfactants (anionic) Cell Test 1.14697.0001	0.05-2	mg L^{-1}	EPA 425.1, US Standard Methods 5540 C, and EN 903	Dodecane-1 sulphonic acid sodium salt for tenside test 1.12146.0005, Merck 1 mg L^{-1} and deionised water 0 mg L^{-1}	Spectroquant NOVA 60, Merck, Germany Pipettor*, VWR, Poland Analog Vortex Mixer, VWR, USA
NH_4-N	Ammonium	Spectroquant Ammonium Cell Test 1.14544.0001	0.5-16	mg L^{-1}	EPA 350.1, US Standard Methods 4500-NH3 D, and ISO 7150/1	Combi R1, Combicheck 20 1.14675.0001, Merck 12±1 mg L^{-1}	Spectroquant NOVA 60, Merck, Germany Pipettor*, VWR, Poland Analog Vortex Mixer, VWR, USA
NO_3-N	Nitrate	Spectroquant Nitrate Cell Test 1.14764.0001	1-50	mg L^{-1}		Nitrate standard solution 1.19811.0500, Merck 1000 mg L^{-1}	Spectroquant NOVA 60, Merck, Germany Pipettor*, VWR, Poland Analog Vortex Mixer, VWR, US

Table 2 (contd.). Chemical kits, their ranges and methods; chosen control solutions, apparatus and measurement days used for the analysis

Substance		Kit name	Measurement range	Unit	Standard method	Control solution name and value	Apparatus
Tot-N	Total nitrogen	Spectroquant Nitrogen (total) Cell Test 1.147630001 and 1.00613.001	10-150 and 0.5-15	mg L^{-1}	EN ISO 11905-1 (digestion)	Nitrate standard solution 1.19811.0500, Merck 1000 mg L^{-1}	Thermoreactor TR 420, Merck, Germany Spectroquant NOVA 60, Merck, Germany Pipettor*, VWR, PolandAnalog Vortex Mixer, VWR, USA
Tot-P	Total phosphorus	Spectroquant Phosphate Cell Test 1.14543.0001	0.05-5	mg L^{-1}	EPA 365.2+3, APHA 4500-P E, and DIN EN ISO 6878	Phosphate standard solution 1.19898.0500, Merck 1000 mg L^{-1}	Thermoreactor TR 420, Merck, Germany Spectroquant NOVA 60, Merck, Germany Pipettor*, VWR, Poland Analog Vortex Mixer, VWR, USA
PO_4-P	Phosphate	Spectroquant Phosphate Cell Test 1.14543.0001	0.05-5	mg L^{-1}	EPA 365.2+3, APHA 4500-P E, and DIN EN ISO 6878	Phosphate standard solution 1.19898.0500, Merck 1000 mg L^{-1}	Spectroquant NOVA 60, Merck, Germany Pipettor*, VWR, Poland Analog Vortex Mixer, VWR, USA

*Ergonomic high-performance pipette

4. RESULTS AND DISCUSSION

4.1 Physical properties of biochar compared with sand

The physical properties of the different types of biochar are shown in Table 3. Biochar filters had smaller particle density and bulk density than sand filters of the same particle size (d_{10} = 1.4 mm; Table 3). This meant that biochar was lighter than sand, making it easier to transport and carry than sand of similar particle size. The specific surface area of the biochar varied from 170-1000 m^2 g^{-1}, which was much greater than that of the sand filter (0.152 m^2 g^{-1}). The activated biochar showed the highest specific surface area (1000 m^2 g^{-1}), while that of non-activated biochar was at the lower end of the biochar range (170-200 m^2 g^{-1}). The specific surface area is an important parameter for evaluating the suitability of a material for use in wastewater filters, as the specific surface of a filter medium plays a significant role for development of biofilm over the surface of the medium. Within this biofilm, biological degradation of organic matter, nitrification and denitrification occurs. Moreover, the larger the specific area, the higher the adsorption capacity of the material. All biochar filters showed comparable porosity (60-74%), which was found to be significantly larger than that in the sand filters (35%). This means that a filter made up of biochar would have a better capacity to hold water in macropores than a sand filter, as well as better capacity to grow biofilm in the pores, which is important for wastewater treatment.

Table 3. Properties of activated biochar, willow biochar, hardwood biochar and sand filters. The hydraulic properties (porosity and mean residence time) were measured at a hydraulic residence time of 32±7 L m^{-1} day^{-1}

Filter material	Activated biochar	Willow Biochar	Hardwood biochar	Sand
Particle size (mm)	1.5 and 2.8-5	1-1.4 and 2.8-5	1.4-5	1.4-5
Air-dry water content (%)	0.6	6.3		
Specific surface area (m^2/g)	>1000		170-200	0.152
Bulk density (kg m^{-3})	560	270	187	1690
Particle density (kg m^{-3})	1890	740		2570
Total porosity (%)	70.6	63.3	72-74	34
Water-filled porosity (%)			48-53	
Mean residence time (hours)	119	108	87[1]; 85[2]; 66[3]	0.5
Hydraulic conductivity (cm h^{-1})	500			360

[1]Residence time of hardwood biochar filters with d_{10} = 0.7 mm. [2]Residence time of hardwood biochar filters with d_{10} = 1.4 mm. [3]Residence time of hardwood biochar filters with d_{10} = 2.8 mm

The corresponding hydraulic residence time in the biochar filters proved to be much longer than in sand filters (3.5-4.9 days in different types of the biochar compared with 0.5 h in the sand filters; Figure 3). There was no significant difference between residence time in biochars with particle size d_{10} 0.7 and 1.4 mm, which had residence time 85-87 h (Figure 4). However, biochar with d_{10} 2.8 mm showed a significantly shorter residence time than all 0.7 and 1.4 mm biochar filters. The activated biochar filters had the longest hydraulic residence time among the biochar filters (4.9 days; Figure 3), owing to activation of biochar enabling development

of nano-, macro- and micropores. As a result, the activated biochar had better retention of liquid inside its pores than non-activated biochar. Increased contact time of the water with the active biofilm allows for more efficient reduction of pollutants. Consequently, wastewater in biochar makes contact with the medium for a longer time than in sand filters, promoting better biological degradation of organic pollutants and enhancing the chances of adsorption of other pollutants such as NH_4-N and PO_4-P.

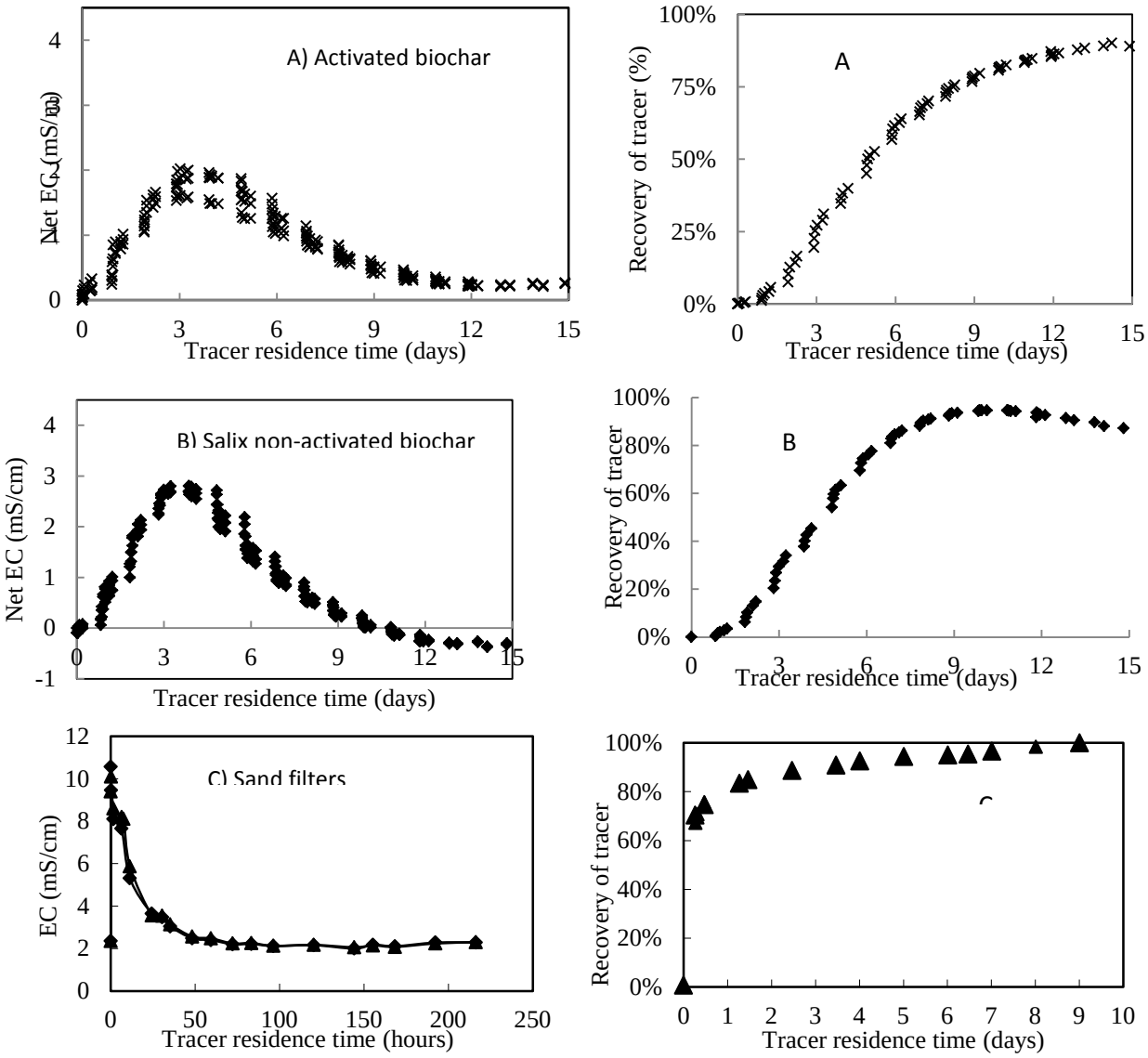

Figure 3. Response curves of the filters to addition of NaCI, measured as electric conductivity (EC) of the effluents in (A, left) activated biochar, (B, left) non-activated willow biochar and (C, left) sand filters. Hydraulic residence time and percentage of recovered tracer as mean values of NaCl tracer after adding a pulse of 10 g L^{-1} to (A, right) activated biochar filters (B, right) non-activated willow biochar filters and (C, right) sand filters. All filters had d_{10} = 1.4-1.5 mm and a hydraulic loading rate of 32 L m^{-2} day^{-1}.

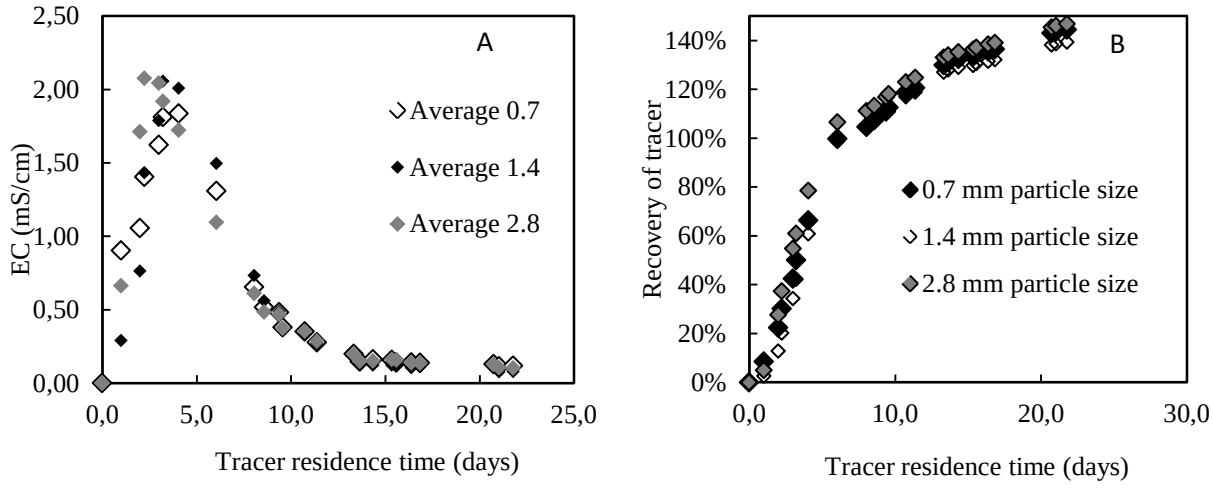

Figure 4. (A) Response curves of hardwood non-activated biochar filters to addition of NaCI, measured as electric conductivity (EC) in the filter effluent, for filters with d_{10} = 0.7, 1.4 and 2.8 mm. (B) Hydraulic residence time and percentage recovery of tracer (mean value) after adding a pulse of NaCl tracer. All filters had a hydraulic loading rate of 32 L m^{-2} day^{-1}.

4.2 Internal structural, surface topography and chemistry

Scanning electron micrographs for the non-activated hardwood biochar, activated biochar and sand particles are shown in Figure 5, in which the surface was magnified 1500 to show features at a scale of 50 µm. The images revealed that the non-activated hardwood biochar retained its original wood structure with high porosity. Its structure is generally characterised by longitudinal hollow tubes, but the image shown in Figure 5 was taken at an orientation which did not show the cross-section of these hollow tubes. However, cross-sectional areas of the hollow tubes in biochar from other resources was imaged using SEM and these showed that the non-activated biochar had high porosity with larger pores than the activated biochar (Figure 6). The SEM image of the activated biochar revealed random structure on the surface of the material and pores that seemed to be more distributed over its surface. The SEM image of the sand particles showed these had a solid structure, with limited occurrence of micropores. In fact, the surface of the sand appeared to have the fewest micropores of the three materials for which SEM images were obtained. However, there appeared to be more minerals on the sand surface than on that of the non-activated hardwood biochar.

Based on the SEM images of the non-activated biochar, activated biochar and sand, the non-activated biochar could be expected to provide better conditions for bacterial attachment and biofilm development on its surface, which usually leads to efficient biological degradation of organic matter and nitrification. The micro- and nanopores on the surface of the activated biochar might be prone to clogging, due to faster biofilm accumulation during wastewater treatment than with the non-activated biochar (which has large pores). This means that non-activated biochar might be more suitable for long-term treatment that activated biochar and sand. In addition, the surface areas surrounding these internal pores of the non-activated biochar will more accessible to wastewater than the surface areas around micro- and nanopores of the activated biochar, which will enhance removal of some pollutants (organic, NH_4-N and PO_4-P) by adsorption.

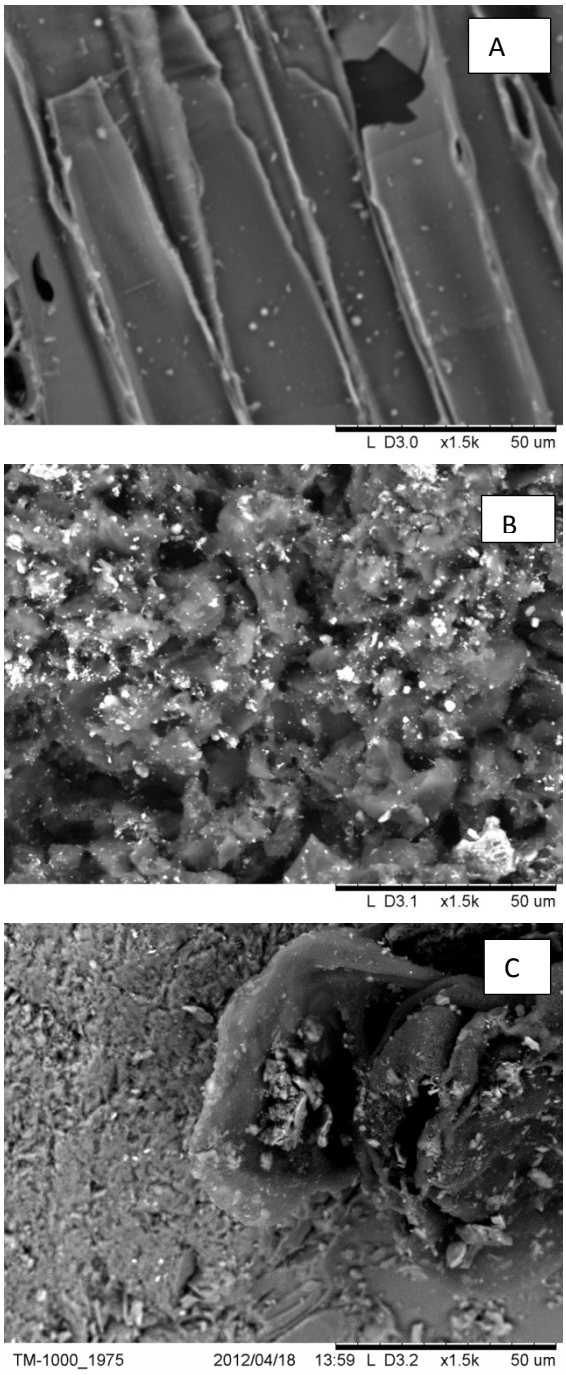

Figure 5. Scanning electron microscope image of the surface of (A) non-activated hardwood biochar, (B) activated biochar and (C) sand. Magnification factor x1500.

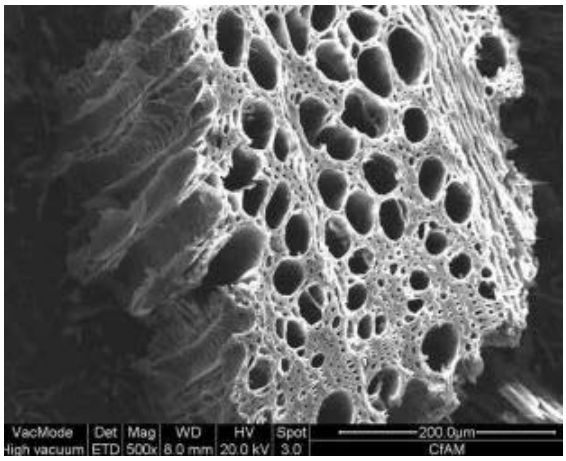

Figure 6. Scanning electron microscope image of the surface of wood biochar charred at 450°C for 48 h. (Photo by Peter Harris).

4.3 Chemical composition of biochar surface compared with sand

The SEM micrographs and the energy dispersive X-ray spectroscopy of the non-activated hardwood biochar did not show a rich mineral content on surfaces (Table 4). The most important minerals for wastewater treatment include those required for precipitation of PO_4-P, such as calcium (Ca), aluminium (Al), iron (Fe) and magnesium (Mg). Of all the minerals found on the surface of the hardwood biochar, Ca was found to correspond to 13% by weight. No Fe or Mg was observed on the surface of the hardwood biochar. Some Al was found, but it was probably not from the biochar surface itself, but from the aluminium holder on which the biochar sample was placed for scanning.

In contrast, among the minerals observed on the surface of the activated biochar, substantial proportions of Fe (41%) and Ca (30%) were present. The surface of sand particles contained more Ca and Fe than the surface of the non-activated hardwood biochar and less than the activated biochar. According to the supplier of the sand used in the studies, this sand has been mixed with 5% lime.

Table 4. Chemical composition of the surface of different types of biochar and sand tested in the studies reported in this report. The values reported in the table are mean ± standard deviation

Element	Hardwood biochar	Activated biochar	Sand
Sodium			2±0
Magnesium		2.0 ±0.3	4±5
Aluminium	39	11 ± 3	13±4
Silicon	13	16 ±5	33±2
Chlorine			8±4
Potassium	34		5±3
Phosphorus		1±0	1±0
Calcium	13	30±8	15±10
Titanium			18±0
Iron		41 ±13	21±14

4.4 Characteristics of the influent wastewater

Wastewater with different pollutant concentrations was used to test the performance of the biochar filters in small-scale wastewater treatment (Table 5). The pollutant concentrations varied from intermediate (e.g. 330 mg COD L^{-1}; 20 mg Tot-N L^{-1} and 4 mg Tot-P L^{-1}) to very high (e.g. 4600 mg COD L^{-1}; >100 mg Tot-N L^{-1} and 19 mg Tot-P L^{-1}). The wastewater quality tested in this project represented different types of household wastewater: (i) conventional household wastewater (which includes wastewater from toilet, kitchen, shower and laundry); (ii) greywater and (iii) very concentrated wastewater or blackwater.

The microbiological contaminants in the wastewater included the most common bacteria used for assessment of wastewater microbial load, including 10^2-10^6 CFU *E. coli* mL^{-1} and 10-10^6 CFU *E. faecalis* mL^{-1}. Onsite systems can be free of pathogens most of the time during the year, but sometimes contain very high levels of pathogens when a gastrointestinal infection occurs in the household connected to the treatment system. Therefore, episodic contamination with salmonella was mimicked by spiking the wastewater with high doses of salmonella for a limited period of 6 weeks, which resulted in 10^5-10^7 CFU *Salmonella* spp. mL^{-1}. The wastewater was also spiked with bacterial viruses (bacteriophages MS2 and PhiX) for periods of 6-8 weeks to mimic outbreaks of human viruses, while *Saccharomyces cerevisiae* was spiked for 6 weeks to mimic an outbreak of *Cryptosporidium paravum*, resulting in 10^6-10^8 PFU MS2 mL^{-1}, 10^5-10^7 PFU Phix-174 mL^{-1} and 10^2-10^3 PFU *Saccharomyces* mL^{-1}.

The bacteriophages MS2 and PhiX-174 (viruses that infect bacterial cells), which have differing surface charge, were selected as model particles for human viruses, e.g. rotavirus and norovirus. MS2 and PhiX-174 are similar in structure, morphology and size to many human enteric viruses (Bradley et al., 2011). They are regularly used in studies to analyse sorption aptitude in aquatic environments (Michen & Graule, 2010). Their small size, survival in harsh conditions and resistance to different treatments make them a good indicator to predict the behaviour of enteric viruses (Ottoson & Stenström, 2003). Furthermore, detection of bacteriophages is simpler and cheaper than other methods for detection of enteric viruses.

Table 5. Characteristics of the wastewater used as feed for the biochar filters tested in this study

Simulated wastewater	medium strength wastewater	High strength wastewater		Low strength Wastewater (greywater)	Blackwater
COD (mg L^{-1})	496±87	1389±100	1229±320	332±103	4600±232
BOD_7 (mg L^{-1})	131±50		629±105 (BOD_5)	26±10 (BOD_5)	
NO_3-N	6±6	1.3±0.2	1.3±2.5	17±8	1.2±0.6
NH4-N		3.7±0.5	11±9	7±3	24±16
T-N (mg L^{-1})	30±4	95±6	78±27	26±8	>100 mg L^{-1}
PO_4-P (mg L^{-1})	1.87±0.94	2.6±0.1	3.2±0.8		
Tot-P (mg L^{-1})		3.6±0.1	3.8±0.7	19±16	
MBAS (mg L^{-1})		82±15			
E. coli (CFU)			10^5-10^6	10^2-10^4	
E. faecalis (CFU mL^{-1})			10^4-10^5	10^1-10^3	10^4-10^6
Salmonella (CFU mL^{-1})					10^5-10^7
MS2 phage (PFU mL^{-1})			10^7-10^8	10^6-10^7	10^6-10^7
Phix-174				10^5-10^6	10^5-10^7
Saccharomyces cerevisiae				10^2-10^3	

4.5 Performance of filters in removal of chemical pollutants from wastewater

4.4.1 Removal of organic matter in biochar filters

The hardwood non-activated biochar filters that were operated at a hydraulic loading rate of 37±7 L m^{-2} day^{-1} and an organic loading rate of 5±1 g BOD_5 m^{-2} day^{-1} showed high efficiency in COD and BOD_7 removal from wastewater (95% and 98%, respectively). This high efficiency led to low concentrations of organic matter in the effluent (Tables 6 and 7). Overall, the removal of COD and BOD_7 in the hardwood non-activated biochar filter was comparable to that achieved by sand filters operated under similar conditions and loading rates. Different results were obtained when activated biochar was compared to sand filters 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}. In this case, the sand removed only 70% of the BOD_5 from the wastewater, while the activated biochar removed 99% (Figure 7).

In another study, activated biochar were compared to sand filters for wastewater treatment under fluctuating hydraulic and loading conditions (hydraulic loading rate varied from 32 to 128 L m^{-2} day^{-1} and organic loading rate from 15 to 76 g BOD_5 m^{-2} day^{-1}). Under the variable loading conditions, the activated biochar filters continued to show efficient removal of BOD_5 (>90%) and COD (>85%) (Table 8). The removal of organic matter from wastewater by activated biochar under fluctuating loading conditions was 10% better than the removal rate by sand filters operated under same conditions (Table 8 and Figure 8).

One significant difference between the behaviour of biochar and sand in terms of organic matter removal was that the biochar achieved high BOD and COD removal immediately after it was taken into service for wastewater filtration. In contrast, the removal of BOD and COD in the sand filters was low at start-up of the filter and increased with time, but did not reach the treatment effect achieved by the biochar (Figure 8).

The capacity of filters to remove pollutants differs between materials due to different characteristics such as porosity, specific surface area and reactivity, adsorption capacity and ability to promote biofilm development for biological breakdown of organics (Rolland et al., 2009). The initial effective reduction of organic matter (BOD_5 and COD) in biochar was attributed to adsorption. Biochar filters are characterised by large specific surface and high porosity, which provides better absorption capacity and thus achieves a greater reduction of organic matter from start-up compared with sand. After the initial period dominated by physical and chemical filtration processes, biological activities gradually take over due to development of a biofilm in biochar. The specific surface of the different types of biochar was much larger than that of the sand (see Table 4), providing even better coverage of biofilm, which is responsible for organic matter degradation. Organic matter degradation by biofilm activity is the dominant removal process in sand and any other biofilter (Pell & Nyberg, 1989) and the rate seemed to be limited by the surface area of the filter material. Pell and Nyberg (1989) reported markedly higher organic matter reduction rates in sand filters with 0.21 mm effective particle size, providing a much larger specific surface area.

Comparisons of non-activated hardwood biochar, willow biochar and activated biochar in terms of removal of COD and BOD from wastewater revealed that all types of biochar tested were very efficient in COD and BOD removal and there were no significant differences between the biochars in removal of COD or BOD (Table 9 and Figure 9). All the different types of biochar tested had comparable properties in terms of surface area, porosity and hydraulic residence time, which led to comparable performance in terms of organic matter removal.

Comparison of the levels of BOD removal in the different biochar filters to the 2006:7 NFS recommendations revealed that the effluent from the biochar filters met the requirement for at least 90% reduction of organic substances (Naturvårdsverket, 2006).

Table 6. Concentrations of pollutants in the wastewater and in effluent from non-activated hardwood biochar filters and sand filters. The filters were fed with real wastewater at loading rate of 37±7 L m^{-2} day^{-1} over a six-month period. The values shown are mean ± standard deviation

Parameter	Wastewater inflow	Hardwood biochar (d_{10} = 1.4 mm)	Sand (d_{10} = 1.4 mm)
COD (mg L^{-1})	496±87	23±14	25±16
BOD_7 (mg L^{-1})	131±50	5±2	14±16
NO_3-N (mg L^{-1})	6±6	14±9	26±10
Tot-N (mg L^{-1})	30±4	15±10	29±7
PO_4-P (mg L^{-1})	1.87±0.94	0.83±0.50	0.55±0.57

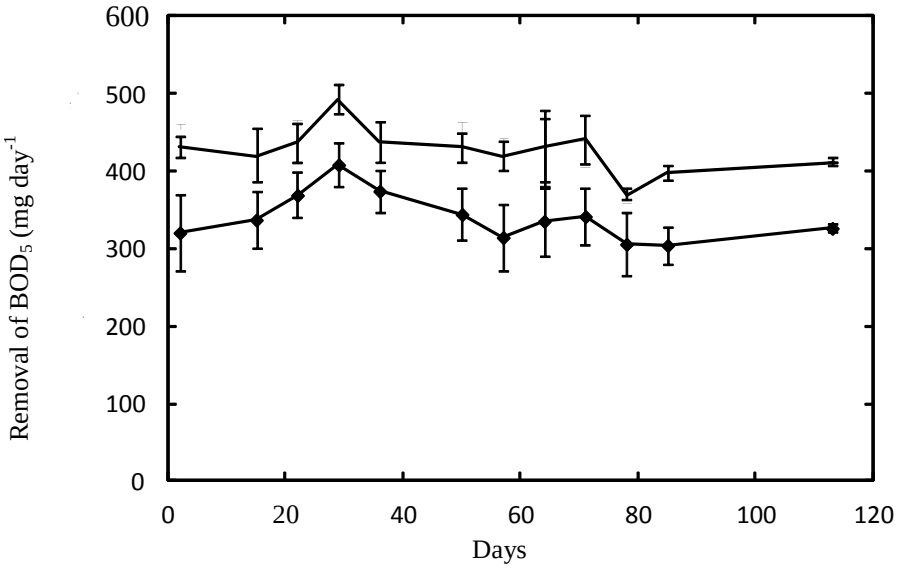

Figure 7. Amount of BOD_5 removed in the activated biochar (—) and sand (♦) filters loaded with wastewater at 14 g BOD_5 m^{-2} day^{-1} during 116 days (mean ± standard deviation).

Table. 7. Percentage removal of wastewater pollutants in biochar and sand filters fed with real wastewater at a hydraulic loading rate of 37±7 L m^2 day^{-1} and an organic loading rate of 5±2 g BOD_7 m^2 day^{-1}. The values shown are mean ± standard deviation

Parameter	Hardwood biochar (d_{10} = 1.4 mm)	Sand (d_{10} = 1.4 mm)
COD	95±3	94±4
BOD_7	98±2	97±5
T-N	52±29	3±16
PO_4-P	62±18	80±13

Table. 8. Percentage removal of wastewater pollutants in activated biochar and sand filters fed with wastewater at fluctuating hydraulic and organic loading regimes over a 6-month period. The values shown are mean ± standard deviation

Parameter	Activated biochar (d_{10} = 1.4 mm)	Sand (d_{10} = 1.4 mm)
COD	85±7	75±9
BOD_5	93±5	83±11
NH_4-N	95±4	83±8
T-N	55±20	3±16
PO_4-P	93±3	70±26
Tot-P	86±4	62±23

Table. 9. Percentage removal of wastewater pollutants in willow biochar and activated biochar filters fed with wastewater at a hydraulic loading rate of 34 L m^{-2} day^{-1} and an organic loading rate of 14 g BOD_5 m^{-2} day^{-1}. The values shown are mean ± standard deviation

	Activated carbon	Willow biochar
MBAS (%)	99±1.3	99±1.4
COD (%)	99±0.3	99±0.3
Tot-P (%)	78±9	89±7
PO_4-P (%)	70±14	86±9
Tot-N (%)	97±4	91±9
NH_4–N (%)	98±3	

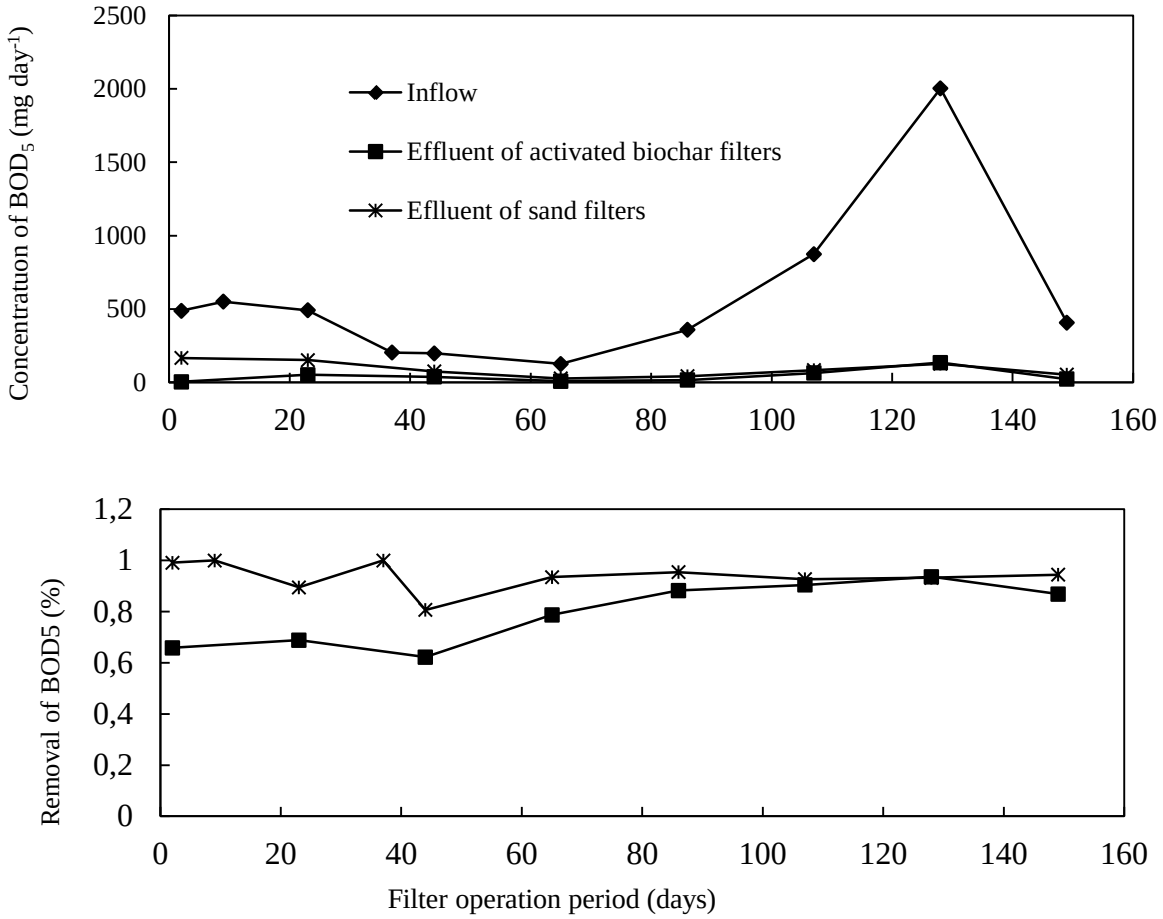

Figure 8. (a) Concentration of BOD_5 in the test wastewater (diamonds) and in effluent from the activated biochar (asterisks) and effluent of the sand filter (squares). (b) Removal efficiency of BOD_5 from the activated biochar filters (asterisks) and sand filters (squares) during 6 months of operation under fluctuating conditions.

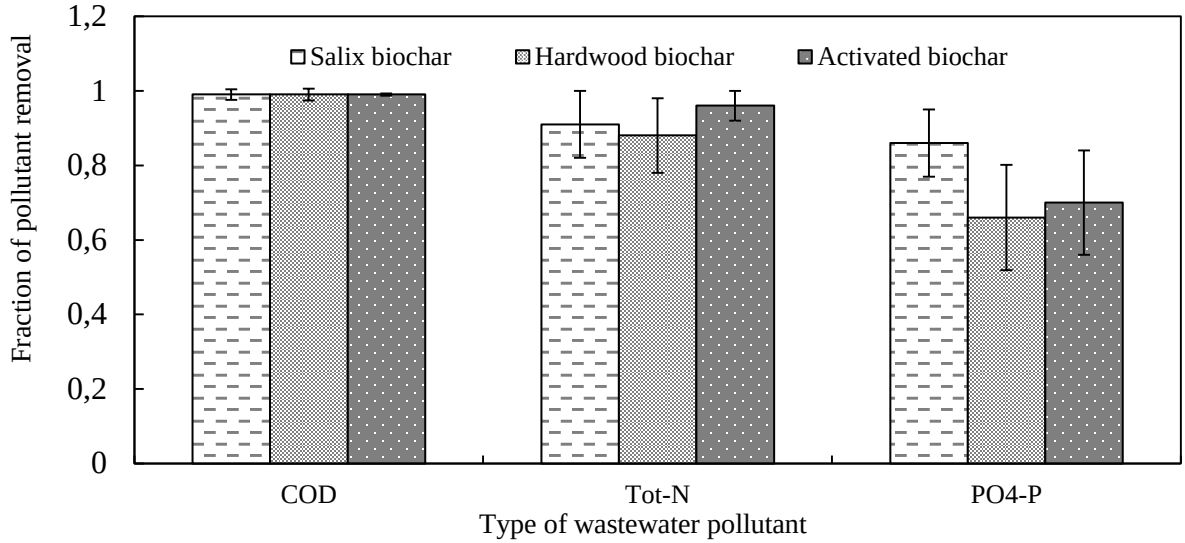

Figure 9. Performance of willow non-activated biochar, hardwood non-activated biochar and activated biochar filter in removal of different types of wastewater pollutants at a hydraulic loading rate of 32±5 L m^{-2} day^{-1}, an organic loading rate of 20±5 g BOD_5 m^{-2} day^{-1} and particle size 1.4 mm.

4.4.2 Removal of nitrogen in biochar filters

The removal of Tot-N in the different types of biochar varied from 52% in non-activated hardwood biochar to 97% in activated biochar operated at a stable hydraulic loading rate of 32-37 L m^{-2} day^{-1}. Very efficient removal of Tot-N (>90%) was observed for all types of biochar during the initial stages of operation of the different biochar filters (up to three months). However, the removal of Tot-N declined gradually with time, mainly in the non-activated and activated biochar filters, to reach a steady state removal of about 50% (Figure 10). The removal of Tot-N in the different biochar filters (50-52%) was much higher than in the sand filters (<5%), despite all filter types being operated at similar loading rates. The performance of biochar in terms of nitrogen removal was 12-fold better than that of sand filters operated under the same conditions. Biochar particle size of 1.4 mm seemed to provide the best performance under hydraulic loading rates <50 L m^{-2} day^{-1}, which are common loading rates for onsite wastewater treatment in Sweden. In biochar filters, the removal of Tot-N is achieved by adsorption of NH_4-N, biological assimilation in biofilm and denitrification. The large surface area of the activated and non-activated biochar (Tables 8-9) enhances the adsorption of ammonium (NH_4-N) from wastewater (Rodrigues et al., 2007). The biofilm developed on the large biochar surface needs nitrogen for its growth, which also enhances the removal of Tot-N. Moreover, biochar is characterised by high porosity and richness in micro- and nanopores (especially activated biochar, see Figure 3). Under extended operation of the biochar filters, occurrence of anaerobic zones in the micro- and nanopores is very likely, due to filling of pores and coverage by the biofilm layer. Occurrence of anaerobic zones promotes growth of denitrifying bacteria and results in enhanced nitrogen removal in biochar. In contrast, sand filters have low porosity and small specific surface, which does not provide good conditions for adsorption. Moreover, as seen in the SEM images of the sand, the surface of sand particles is solid, with few micro- or nanopores which does not provide the right conditions for nitrogen removal by nitrification.

The different types of biochar filters did not show statistically significant differences in terms of nitrogen removal under a hydraulic loading rate of 32±7 L m^{-2} day^{-1}, an organic loading rate of 20±5 g BOD_5 m^{-2} day^{-1} and particle size 1.4 mm. However, the activated biochar still showed a tendency to remove nitrogen from wastewater more effectively than the non-activated willow and hardwood biochar (Figure 9).

Despite the low removal (50%) of Tot-N in the biochar filters, they showed high potential for nitrogen removal by adsorption and denitrification. The biochar filters tested were not designed to remove nitrogen, but rather were fully operated under aerobic conditions to achieve high removal of organic matter (COD and BOD). However, once the existing design of the biochar filters is optimised to involve proper denitrification, the total nitrogen removal can be expected to increase significantly. Good performance of biochar in terms of nitrogen removal would make it a potential alternative medium to be used in onsite wastewater treatment systems in environmentally sensitive areas, where at least 50% reduction of nitrogen is required.

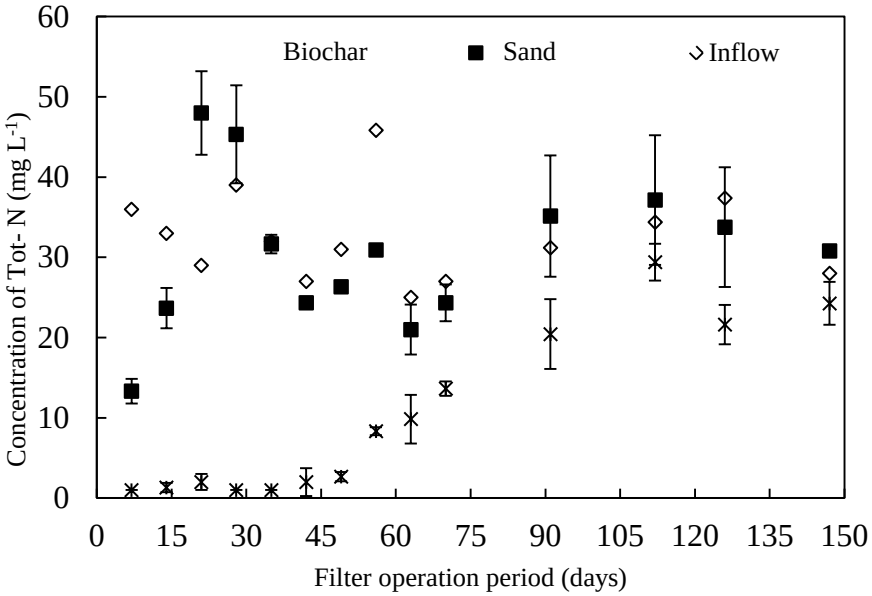

Figure 10. Treatment performance of non-activated hardwood biochar compared with sand (d_{10} = 1.4 mm in both cases) in removal of total nitrogen (Tot-N) during 6 months of filter operation at hydraulic loading rate of 37 L m^{-2} day^{-1}

4.4.3 Removal of phosphorus in biochar filters

Willow biochar showed efficient removal of phosphate (PO_4-P) and total phosphorus (Tot-P), with an average of 89±7% and 86±9%, respectively, during the two-month test period, in which the filters were operated at a hydraulic loading rate of 32 L m^{-2} day^{-1} and an organic loading rate of 20±5 g BOD_5 m^{-2} day^{-1} (Figure 11). Unfortunately, Tot-P removal in willow biochar was not studied during an extended period of wastewater treatment. The activated biochar also showed efficient removal of both Tot-P and PO_4-P during six months of fluctuating wastewater loading conditions (hydraulic load varied from 32 to 128 L m^{-2} day^{-1} and organic loading rate from of 15 to 76 g BOD_5 m^{-2} day^{-1}). The overall removal rate of Tot-P and PO_4-P during this period was 86±4% and 93±3%, respectively. In contrast, the removal of Tot-P and PO_4-P was not similarly efficient in the non-activated hardwood biochar, which showed removal of 32 to 60% (Table 7 and Figure 9). However, when the hardwood biochar was used for treatment of real wastewater, its effluent had Tot-P of <1 mg L^{-1} (Table 6). Comparing the biochar with the sand filters, it should pointed out the sand filters performed better (75-83%) than the hardwood biochar as regards PO_4-P removal (62%). However, the activated biochar achieved better removal of phosphorus than the sand filters when all filters were operated under similar hydraulic and organic loading rates (Table 8).

Adsorption is the principal mechanism for PO_4-P reduction in sand filters (Pell and Nyberg, 1989), and the capacity of the sand to bind phosphorus depends on pH and the Ca, Fe and Al concentrations in the sand (Arias et al., 2001). The characteristics of the medium (specific surface, mineral content on the surface of the particles) play a significant role in the removal of PO_4-P in wastewater filters. As shown in the elemental SEM images (Figure 6; Table 4), the surface of the hardwood biochar is poor in terms of mineral and metal content (Fe and Ca

and Mg). Thus the removal of PO_4-P was not as efficient as in the activated biochar and sand filters, which both have a richer mineral content on their surfaces than the hardwood biochar (see Table 5).

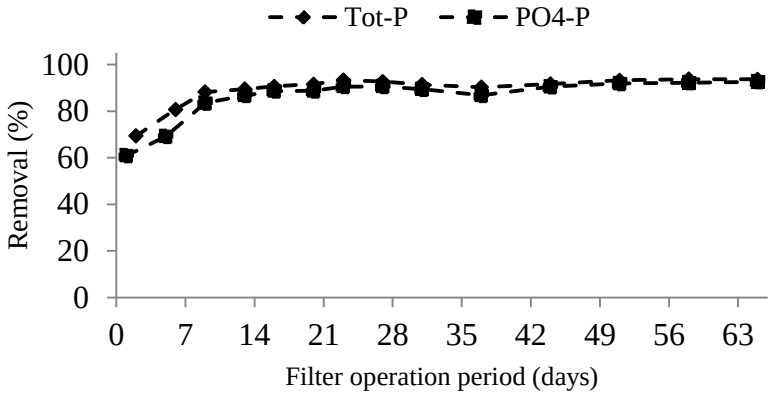

Figure 11. Removal efficiency of total phosphorus (Tot-P) and phosphate (PO_4-P) in willow biochar filters during a test period of two months of wastewater treatment. The hydraulic loading rate was 32 L m^{-2} day^{-1}, the organic loading rate 20±5 g BOD_5 m^{-2} day^{-1} and particle size 1.4 mm.

4.4.4 Performance of hardwood biochar filters at different particle sizes

The hardwood biochar showed efficient removal of organic matter (94-99% for BOD_5 and COD) for all particle sizes tested (0.7, 1.4 and 2.8 mm) when operated for 6 months at a hydraulic loading rate of 34 L m^{-2} day^{-1} and an organic loading rate of 20±5 g BOD_5 m^{-2} day^{-1}. Nonetheless, there was a statistically significant difference between the 2.8 mm filter and the other sizes (0.7 and 1.4 mm) regarding COD removal. The 2.8 mm hardwood biochar filter had the lowest efficiency (94%) of all the particle sizes tested. The larger the particle size, the larger the macropores in the filter. Under such conditions, it is likely that more pores are connected to each other, leading to some of the wastewater passing through the filter quickly without enough contact time between the filter medium and organic matter in the wastewater. These fractions of non-treated wastewater will appear in the effluent of the filters. One finding to support this assumption is that the hydraulic residence time of the 2.8 mm filters was shorter (66 h) than that of the 0.7 and 1.4 mm filters (85 and 87 h, respectively). Other evidence is provided by the finding that NH_4-N concentration was higher in the effluent of the 2.8 mm biochar filters compared with the 0.7 and 2.8 mm filters, indicating that some wastewater passed quickly through the biochar without sufficient treatment (Figure 12).

No statistically significant difference was found between the 0.7, 1.4 and 2.8 mm biochar filters in terms of their removal of Tot-N, Tot-P or PO_4-P. However, a significant difference was found regarding removal of NH_4-N in the 2.8 mm filters, which did not remove NH_4-N efficiently, as explained earlier.

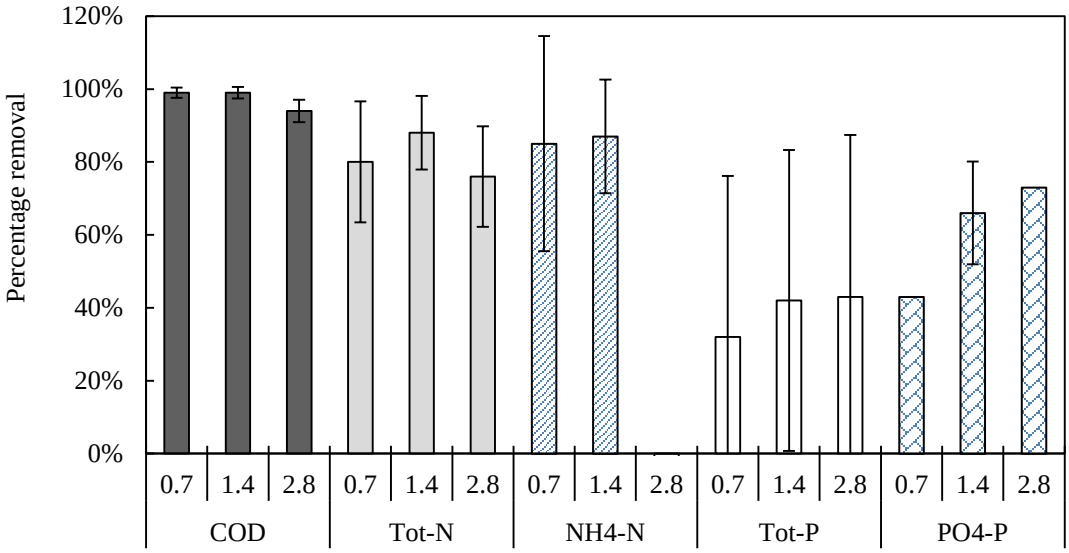

Figure 12. Performance of hardwood biochar filter in removal of different types of wastewater pollutants at a hydraulic loading rate of 34 L m^{-2} day^{-1}, an organic loading rate of 20±5 g BOD_5 m^{-2} day^{-1} and particle size 0.7, 1.4 and 2.8 mm.

When hardwood biochar filters with d_{10} of 1.4, 2.8 and >5 mm were tested at a hydraulic loading rate of 200 L m^{-2} day^{-1}, different trends were obtained regarding the relationships between pollutant removal and biochar particle size. A clear trend of decreased BOD_5 removal efficiency was found as the particle size of the biochar increased (Figure 13). Consequently, the BOD_5 removal dropped from 94% to 68% as the particle size increased from d_{10} 1.4 mm to >5 mm. The removal of NH_4-N showed a similar trend, declining from 99% at d_{10} 1.4 mm to 63% at d_{10} >5 mm, while the nitrification rate decreased from 11 mg L^{-1} for d_{10} 1.4 mm to 7.5 for d_{10} >5 mm. In contrast to the trends observed for BOD_5 and NH_4-N removal, the removal of Tot-N increased by 10% as the effective particle size increased from 1.4 or 2.8 to >5 mm (Figure 13). Under this hydraulic loading rate (200 L m^{-2} day^{-1}), Tot-N removal was 73% in biochar filters with d_{10} >5 mm compared with 62-63% for d_{10} 1.4-2.8 mm.

As discussed earlier, in an infiltration medium with large particle size, more wastewater is likely to pass through the filter pores without sufficient treatment of BOD_5, which eventually ends up in the effluent from the filter. In addition, 200 L m^{-2} day^{-1} is a high hydraulic loading rate. Thus, the infiltration rate through the filters can be expected to increase and the exchanges between mobile water in macropores and water retained in micropores will decrease (Boller et al., 1993). Consequently, saturated zones with anaerobic conditions are likely to have occurred in the biochar filters with large particle size, due to the pores filling with water. Anaerobic zones are not favourable for nitrifying bacteria (which are responsible for transforming NH_4 into nitrate (NO_3), so less NH_4 is nitrified in filters at high loading and infiltration rates. While having some BOD_5 passing through the filter is not acceptable in terms of BOD_5 removal, BOD_5 is good for enhancing nitrogen removal from the wastewater. The bacteria responsible for nitrogen removal (denitrifying bacteria) are heterotrophic species

that need organic carbon as an energy source to reduce nitrate into N_2 or N_2O under anaerobic conditions. Such conditions can be assumed to be behind the enhanced removal of nitrogen by biochar filters at d_{10} >5 mm.

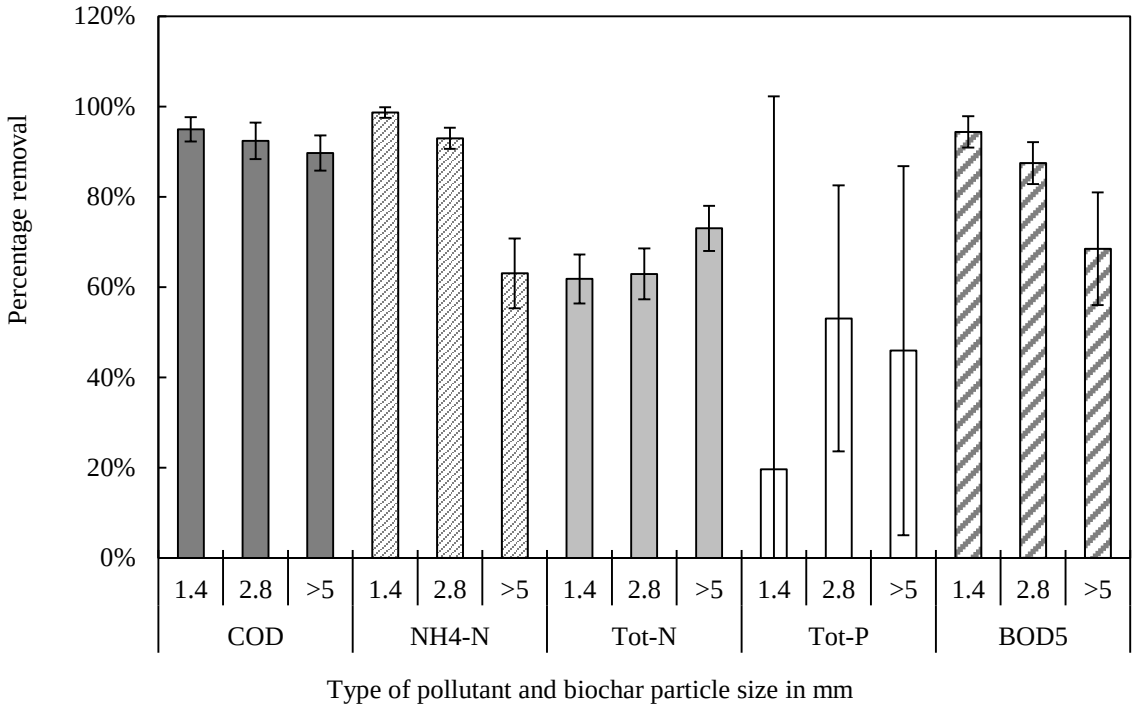

Figure 13. Performance of non-activated hardwood biochar filters in removal of different types of wastewater pollutants at a hydraulic loading rate of 200 L m^{-2} day^{-1}, an organic loading rate of 5±2 g BOD_5 m^{-2} day^{-1} and particle size 1.4, 2.8 and >5 mm.

4.4.5 Performance of biochar filters at different organic and hydraulic loading rates

The removal of COD in the non-activated hardwood biochar was similar at both the 5±2 and 20±5 g BOD_5 m^{-2} day^{-1} loading rates (95 and 99 % removal, respectively), with an effluent concentration of about 10±3 mg L^{-1} for both rates (Figure 14). Despite this small difference in percentage removal of COD, the rate of removal at the organic loading rate of 20±5 g BOD_5 m^{-2} day^{-1} was significantly higher. A possible explanation for this is that the higher organic loading provided more substrate to the biofilm developing on the surface of the biochar. When the flux of organic matter to biofilm increases, the biological activity of the microorganisms is stimulated (Wilson et al., 2011) and thereby also the mineralisation rate of organic matter (Wijeyekoon et al., 2004). However, excessive loading of the filters with high organic loading rates for an extended period might lead to failure of the filter by clogging. Moreover, under high organic loads a dense biofilm can develop and restrict the flux of substrate into the interior of the biofilm (Wijeyekoon et al., 2004). This results in microbial starvation and lost areas of microbial activity, leading to partial biofilm detachment and, consequently, emission of pollutants (Li et al., 2011).

The biochar filters showed a tendency for increased removal of Tot-N at the higher organic loading rate (20±5 g BOD_5 m^{-2} day^{-1}). However, the high variation in Tot-N removal at the lower rate (5±2 g BOD_5 m^{-2} day^{-1}) made it difficult to identify statistically significant trends in removal between the organic loading rates. Increasing amounts of influent organic matter probably caused a thick and homogeneously distributed biofilm to develop. This in turn created more anoxic sites, providing favourable conditions for denitrification, as also suggested by Gill et al. (2009). Another explanation for the increasing Tot-N reduction could be that fast-growing heterotrophic bacteria assimilated nitrogen for growth. The organic loading rates showed no significance effects in percentage removal of PO_4-P in the hardwood biochar filters (Figure 14).

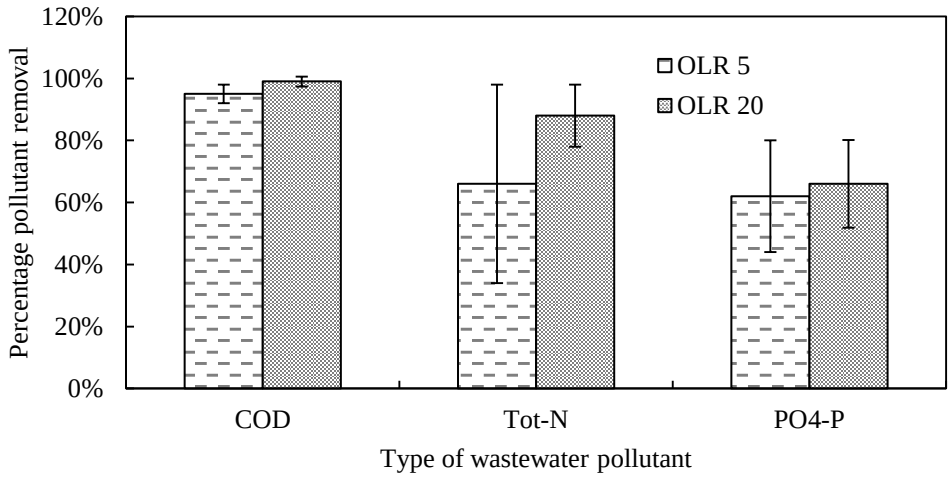

Figure 14. Performance of hardwood biochar filters in removal of different types of wastewater pollutants at two organic loading rates, 5±2 and 20±5 g BOD_5 m^{-2} day^{-1}, a hydraulic loading rate of 37±7 L m^{-2} day^{-1} and d_{10} = 1.4 mm.

As for the performance of biochar filters under different hydraulic loading rates, no significant effects or trends on percentage removal of COD, BOD_5, Tot-N and PO_4-P could be identified when the 1.4 mm hardwood biochar filters were loaded at 37±7 and 200 L m^{-2} day^{-1} and an organic loading rate of 5±2 g BOD_5 m^{-2} day^{-1} (Figure 15).

4.4.6 Clogging potential in biochar filters

As wastewater treatment in filters proceeds, biofilm, solids and dead cells from biofilm generally accumulate in the pores of the filter and decrease its porosity, which can progress to cause clogging of the pores and failure of the filter. The first sign of clogging in filters is usually failure of wastewater to percolate through the infiltration area of the filter. Early signs of clogging can be confirmed by observing the movement of water inside the filter. However, since it is a complicated procedure to observe water movement through filters, we opted to determine the time lag between wastewater application and the first outflow from the filters, i.e. the shortest residence time. The shortest residence time in activated biochar filters operated under a fluctuating loading regime (hydraulic load varied from 32 to 128 L m^{-2} day^{-1} and organic loading rate varied from 15 to 76 g BOD_5 m^{-2} day^{-1}) did not show any signs of

clogging of the filters during the six months of operation (Figure 16). In contrast, the shortest residence time in sand filters operated under the same loading regime start to lengthen within one month of operation and reached a peak after 90 days. Thereafter, sloughing of biofilm was observed in the effluent of the sand filter. This shows that activated biochar filters are likely to operate for longer periods than sand filters before they show clogging.

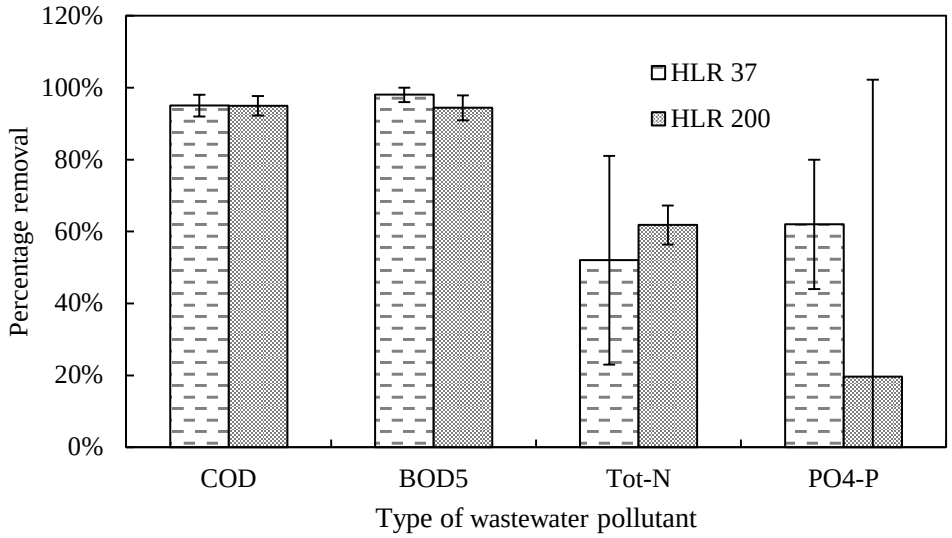

Figure 15. Performance of hardwood biochar filters in removal of different types of wastewater pollutants at two hydraulic loading rates, 37±7 and 200 L m^{-2} day^{-1}, an organic loading rate of 5±2 g BOD_5 m^{-2} day^{-1} and particle size 1.4 mm.

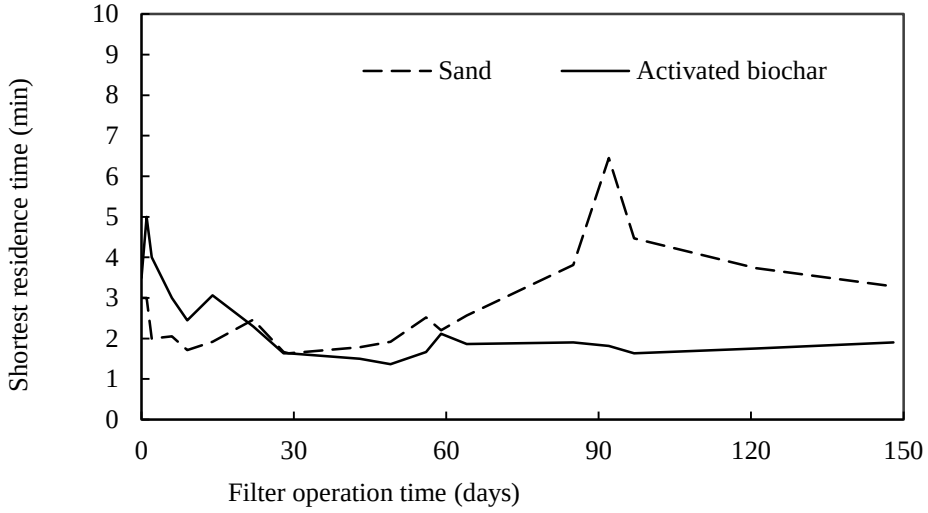

Figure 16. Shortest residence time in the activated biochar (——) and sand filters (- - -) during an operation period of 6 months under a fluctuating loading regime.

4.5 Performance of filters in removal of microorganisms

4.5.1 Removal of microorganisms in different types of biochar

Non-activated hardwood biochar and activated biochar were tested for removal of bacteria (*Salmonella* spp., *Enterococci faecalis*) and bacterial viruses (Phix-174 and MS2). Non-activated hardwood biochar filters loaded at 70 g BOD_5 m^{-2} day^{-1} and a hydraulic loading rate of 32 L m^{-2} day^{-1} achieved 2.4±1, 2.4±1.3, 0.9±0.5 and 1.4±0.8 log_{10} reduction in *Salmonella* spp, *E. faecalis*, Phix-174 and MS2 phages, respectively (Figure 17). When the activated biochar was compared with activated biochar, no significant difference in performance was found. The level of removal of *Salmonella* in the biochar can be considered good for decreasing pathogen spread if the household inhabitants are infected with *Salmonella.* The removal of *Salmonella* spp. in biochar filters is probably due to physical straining out of bacteria in the small pores of the biochar (Kristian Stevik et al., 2004).

Salmonella spp. are rod-shaped, gram-negative bacteria belonging to the Enterobacteriaceae family and are motile by their flagella (Borman et al., 1944) (Octavia & Lan, 2014). *Salmonella* are zoonotic, i.e. infect both humans and animals. By 2007, 2579 serotypes of *Salmonella* had been identified. *Salmonella* infection leads to salmonellosis, which causes gastrointestinal symptoms such as diarrhoea. Non-typhoidal salmonellosis has a low mortality rate, while the typhoidal form has a higher mortality rate (Buckle et al., 2012). In 2004, the global case mortality rate of typhoid fever was 1%, although it can be higher in specific population groups (Crump et al., 2004). *Enterococcus faecalis*, a naturally occurring bacterial species in the human intestine, can be used as an indicator organism.

It has been demonstrated previously that removal of viruses from filtration systems is likely to be adsorption-based and thus dependent on the pH of the filter medium, as the charge of viruses changes with pH (Lalander et al., 2013). The isoelectric point (ISP; the pH at which a particular molecule will carry no charge) of many viruses is in the acidic pH range, e.g. the ISP of MS2 is 3.9 (Dowd et al., 1998). The average pH of the biochar filter effluent in studies summarised in this report was 8.1, and thus the adsorption of virus to this filter medium can be expected to be low.

No particular significant reduction in PhiX-174 phage was found in the biochar filters, while about 1 log_{10} reduction of MS2 phage was achieved. One main reason for the poor removal of phages by filters is that these bacterial viruses are very small in size (24-27 nm) (Elving, 2012). Thus retention (straining) of these nanoparticles in macrofilters such as sand or biochar might not be efficient. In addition, removal of viruses from wastewater in infiltration systems can be driven by adsorption, a process which depends on the ISP of the virus and the pH of the filter.

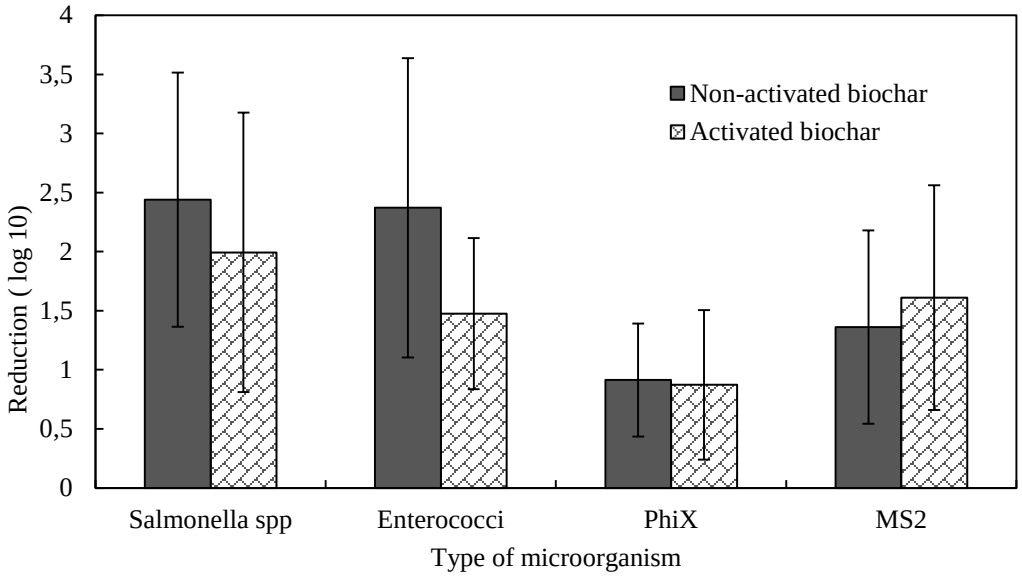

Figure 17. Removal of different types of microorganisms in activated biochar and non-activated hardwood biochar at a hydraulic loading rate of 32 L m^{-2} day^{-1}, an organic loading rate of 70 g m^{-2} day^{-1} and particle size 1.4 mm.

4.5.2 Removal of microorganisms at different biochar particle sizes

The hardwood biochar filters showed efficient removal of *E. coli* (>4 log $_{10}$ reduction) and *E. faecalis* (>4.5 log $_{10}$ reduction) when the effective size of the biochar material was small (d_{10} = 0.7 and 1.4 mm) at a hydraulic loading rate of 34 L^{-2} day^{-1} and an organic loading rate of 20 g BOD_5 m^{-2} day^{-1} (Figure 16). The removal of *E. coli* and *E. faecalis* was less efficient in the hardwood biochar filters with d_{10} = 2.8 mm. However, no statistical significant difference was found in removal of MS2 phage among the different particle sizes (Figure 18). Similar trends were observed for *E. coli* and *E. faecalis* removal in hardwood biochar with particle size 1.4, 2.8 and >5 mm, loaded at 200 L m^{-2} day^{-1} and 20 g BOD_5 m^{-2} day^{-1}. Under these loading rates, hardwood biochar with d_{10} = 1.4 mm was the most efficient in removal of *E. coli*, *E. faecalis*, yeast and Phix-174 phage (Figure 19). The 1.4 mm biochar filters achieved a 1.4, 1.5, 2.0 and 1.3 log_{10} reduction for *E. faecalis*, *E. coli*, *Saccharomyces cerevisiae yeast* and Phix-174, respectively. No statistically significant difference in removal of microorganisms was found between the hardwood biochar filters with d_{10} = 2.8 and >5 mm. It is obvious from these results that the smaller the particle size of the biochar, the better the straining of bacterial and virus particles.

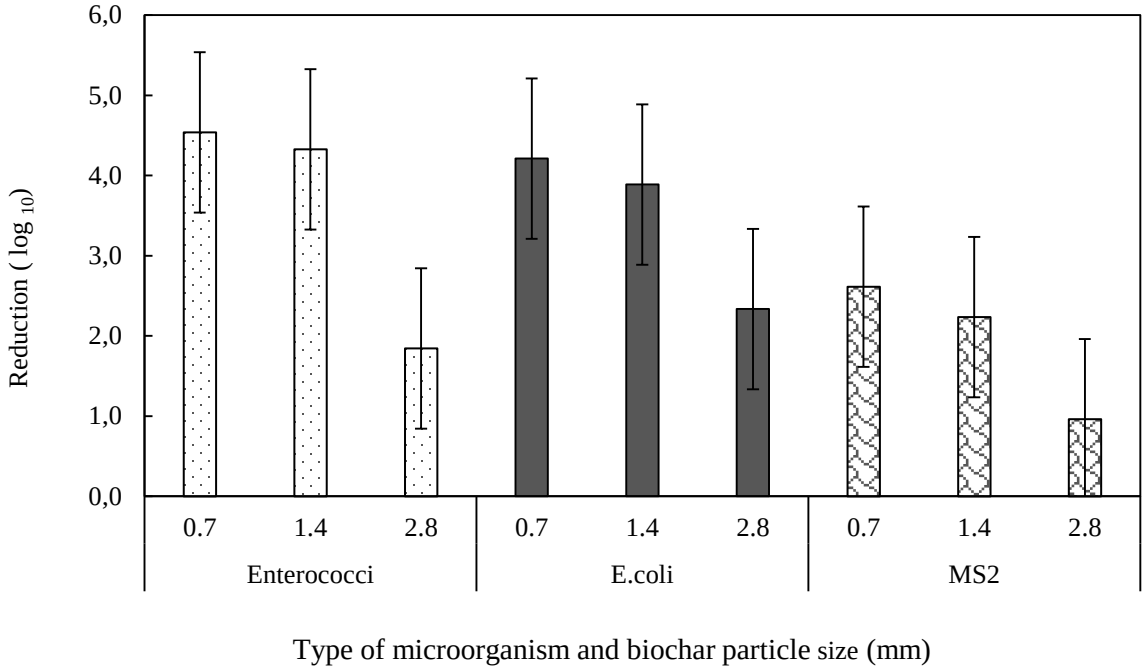

Figure 18. Performance of hardwood biochar filters in removal of different types of microorganisms at a hydraulic loading rate of 32 L m^{-2} day^{-1}, an organic loading rate of 20±5 g BOD_5 m^2 day^{-1} and particle size 0.7, 1.4 and 2.8 mm.

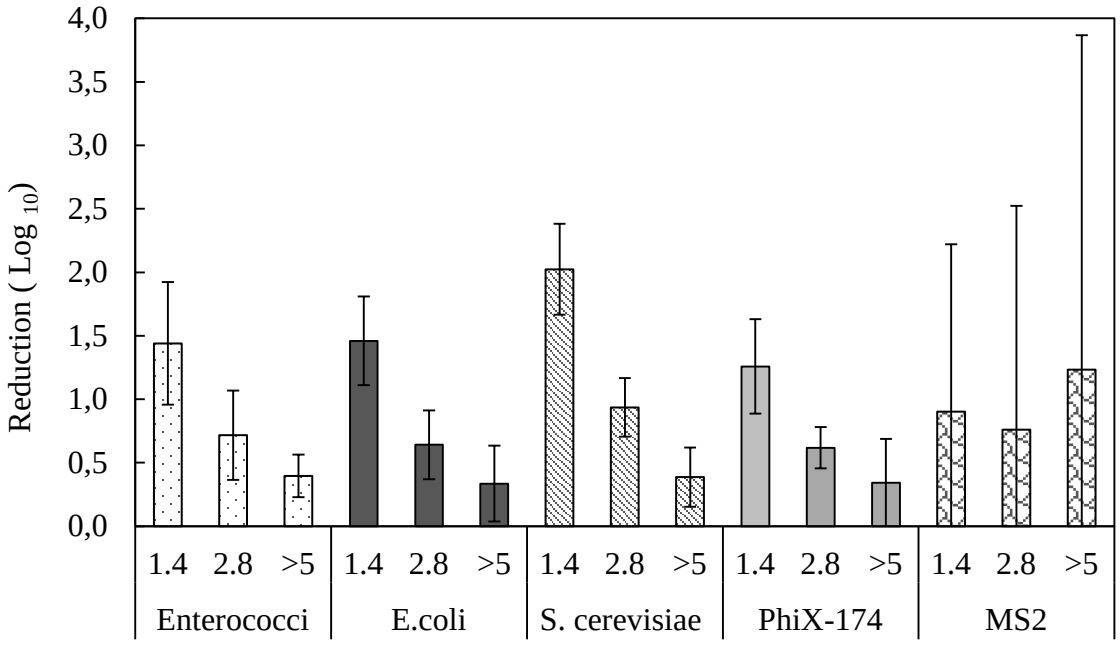

Figure 19. Performance of hardwood biochar filters in removal of different types of microorganisms at a hydraulic loading rate of 200 L m^{-2} day^{-1}, an organic loading rate of 5.2±2 g BOD_5 m^{-2} day^{-1} and particle size 1.4, 2.8 and >5 mm.

4.5.3 Removal of microorganisms at different hydraulic and organic loading rates

Hydraulic loading rate seemed to have clear effects on removal of *E. coli* and *E. faecalis* in the biochar filters. Better straining of these bacteria in the 1.4 mm biochar filters was obtained at a hydraulic loading rate of 32 L m^{-2} day^{-1} than at 200 L m^{-2} day^{-1} (Figure 20). On the one hand, at the high hydraulic loading rates the water velocity inside the filter pores might have increased, leading to washout of previously attached bacteria from the filter. On the other hand, the filters fed with the high hydraulic loading rate were subjected to a low organic loading rate (5±2 g BOD_5 m^{-2} day^{-1}), which means that the biofilm growing on the low organic substrate might be thinner than that growing on the high organic substrate (20±5 g BOD_5 m^{-2} day^{-1} at the 32 L m^{-2} day^{-1} hydraulic loading rate). With thicker biofilm, the internal pores in the filters are narrowed, which contributes to enhanced removal of bacterial particles by straining in narrow pores.

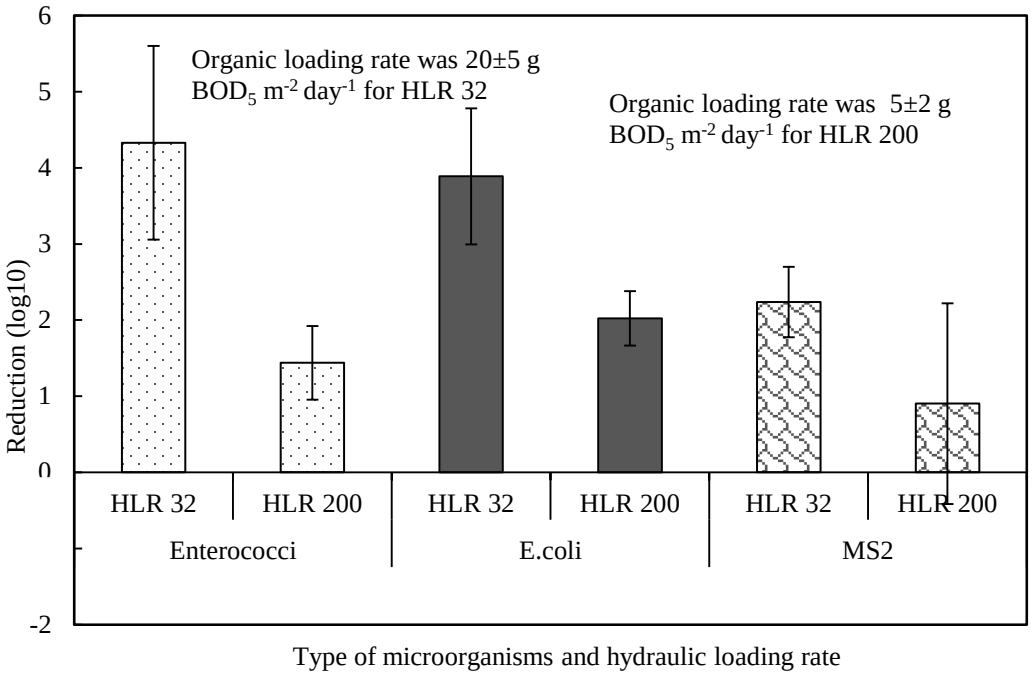

Figure 20. Performance of hardwood biochar filters (1.4 mm) in removal of different types of microorganisms at two hydraulic loading rates, HLR 32 and 200 L m^{-2} day^{-1}.

4.5 Performance of household biochar filter

The household biochar filter system was found to efficiently lower the concentration of BOD_5, TSS and NH_4, with average reduction rates of 93±7, 85±5 and 87±7%, respectively, while the reduction in Tot-P was poor (44±19%; Table 10). Over one year of operation (March 2013 to April 2014), the household biochar filter showed stable and high removal of BOD_5 and NH_4-N, while the removal of Tot-P decreased from 48±9% in April 2013 to 30±7% in April 2014, showing deterioration in Tot-P removal capacity. Adsorption and subsequent biological degradation of organic matter in biofilm is the main removal mechanism expected to occur in biochar filters. Biochar is inert material produced by

pyrolysis, which results in both large specific surface (200-800 m^2 g^{-1}) and development of micropores (Downie et al., 2009).

Table 10. Influent and effluent concentrations of physical, chemical and pathogen indicators in the influent and effluent from the biochar household treatment system and the overall percentage reduction achieved based on the mean concentrations in the influent and effluent

	Influent	Effluent	Reduction (%)
pH	8.0±0.5	7.8±0.3	
BOD_5	377±85	28±25	93±7
TSS (mg L^{-1})	118±59	17±8	86±14
Tot-P (mg L^{-1})	6.6±1.9	3.7±1.5	44±19
NH_4 (mg L^{-1})	72±14	9.2±4.6	89±25
Somatic coliphages (PFU 100 mL^{-1})	2±2	6±14	0±0.6
Male-specific coliphages (MS2) (PFU 100 mL^{-1})	<1±0	<1±0	
E. coli (log_{10} MPN 100 mL^{-1})	5.1±1.2	4.4±1.5	0.66±1.75
Faecal enterococcus (log_{10} MPN 100 mL^{-1})	0.77±0.75	2.26±0.8	-1.44±1.57
Salmonella typhi (MPN 100 $mL^{-1)}$	<1.1	<1.1	
Water generation rate (L $person^{-1}day^{-1)}$	70 (490 L day^{-1})		

The performance of the biochar filters in terms of pollutant removal in the household filter system was comparable to that obtained in the laboratory systems. In fact, the household biochar filter was designed based on the results obtained in laboratory studies regarding particle size, hydraulic and organic loading rates. The pollutant removal rates achieved by the household biochar filter were similar to those reported for a small household biochar filter (1 m × 0.6 m, diameter × height) by Niwagaba et al. (2014). Their filter removed 91±5.4 and 96±3% of COD and BOD_5, respectively, and 95±3% of faecal coliforms with a water retention time in the filter of 36 h. The removal efficiency of Tot-N, Tot-P, TSS and total dissolved solids (TDS) was 39, 30, 85 and 78.6%, respectively (Niwagaba et al., 2014). This performance is comparable to that of sand filters, probably the most common filter type used for wastewater treatment. In comparison to onsite biochar filters, onsite sand filters operated in another study at a loading rate of 24 g BOD_5 m^{-2} day^{-1} and 44 L m^{-2} day^{-1} achieved BOD_5, COD, TSS and *E. coli* removal rates of 87%, 83%, 85% and 3.42 log_{10} reduction, respectively (Assayed et al., 2010). However, their sand filters had d_{10} = 0.2 mm and were totally clogged after 6 months of wastewater treatment.

The level of *E. coli* in the effluent from the household biochar filter varied widely (by >3 log_{10} MPN 100 mL^{-1}), making it difficult to determine the reduction capacity of the filter (Table 10). The upper surface of the biochar filter was covered with only a 5 cm thick layer of gravel and the post-treatment storage tank for the filtered influent water was not sterile. This means that bacterial indicators such as *E. coli* and faecal enterococci could have entered the bed due to external contamination. Moreover, secondary growth could have occurred in the biochar filter or in the post-treatment collection tank. The structure of biochar, which is characterised by large specific surface area (200-800 m^2 g^{-1}) and high proportions of micro- and macropores (Downie et al., 2009), provides a good environment for shelter, protection

and growth of bacteria (Thies & Rilling, 2009). A number of studies have reported creation of a nutrient-enriched environment supporting high bacterial growth in biochar (Pietikäinen et al., 2000; Scholz & Martin, 1997; Shimp & Pfaender, 1982; Thies & Rilling, 2009; Van Duck, 1984). Excessive growth may lead to some bacteria being washed out with the treated water and hence occasionally more bacteria in the effluent than in the influent. The household biochar filter in the present study achieved better removal of organic matter than the sand filter described by (Assayed et al., 2010), but the sand filter was better at *E. coli* removal (3 $\log_{10}$ reduction). However, (Assayed et al., 2015) reported that removal of *E. coli* in sand filters fluctuated between 1 and 7 $\log_{10}$ reduction and was influenced by the hydraulic loading rate of the filter.

5. CONCLUSIONS

The physical and hydraulic properties (bulk density, porosity, surface area and hydraulic residence time) of different types of biochar filters suggested that they had higher potential treatment capacity than sand, both per unit volume and per unit weight. Due to their large surface area and porosity, the biochar filter media achieved pollutant removal by adsorption and biological degradation to a much larger extent than the sand filters. The properties of biochar filters were shown to improve their capacity to buffer variations in hydraulic and organic loading rates. Thus they could better handle shock loads than sand filters.

Results obtained from the laboratory-scale filters and the full-scale household filter proved that biochar filters are efficient and robust in removal of organic matter from wastewater under stable and variable loading regimes, including high and low loading rates. Different types of biochar (activated and non-activated) were equally efficient in removal of organic matter. Moreover, biochar filters with effective particle size (d_{10}) varying from 0.7 to >5 mm were all equally efficient in removal of organic matter (>90% reduction). When biochar filters are used for wastewater treatment, the removal of the organic matter starts immediately when the filters are taken into operation, without any delay period, which simplifies start-up. This is due to the large surface area, which provides immediate removal of organics by adsorption and thus allows time for initiation of biological degradation. The level of removal of organics by the biochar filters complied with the recommended levels suggested by Swedish Environmental Protection Agency (Naturs vårdverket).

Despite the biochar filters not being designed to enhance denitrification, they achieved intermediate to high (65-88%) removal of nitrogen, depending on the type of biochar and loading rate. The performance of biochar in nitrogen removal was 12-fold better than that of sand filters operated under the same conditions. Biochar particle size 1.4 mm seemed to provide the best performance under hydraulic loading rates <50 L/m^{-2} day^{-1}, which is a common loading rate for onsite wastewater treatment in Sweden. The good performance of biochar in nitrogen removal makes it a potential alternative medium for use in onsite wastewater treatment systems in environmentally sensitive areas, where at least 50% reduction of nitrogen is required. Further work is ongoing to optimise removal of nitrogen in biochar filters by testing different constructions/layering of the filters.

The removal of phosphorus varied among the types of biochar tested and most biochar filters and the household biochar showed deteriorated removal of phosphorus over time. However, work is ongoing to modify the surface of the biochar to enhance removal of phosphorus from wastewater and long series of measurements are planned to evaluate the long-term capacity of biochar filters in this respect.

The removal of bacteria and bacterial viruses was best in the biochar filters with small particle size (0.7-1.4 mm), which resulted in up to 4-5 $\log_{10}$ reductions in *E. coli* and enterococci and 2-3 $\log_{10}$ reduction in phages. However, biochar filters with larger particle size might fail to achieve such levels of reduction, especially at high hydraulic loading rates.

6. FUTURE RESEARCH AND FOLLOW-UP

The performance of the household biochar filter in removal of wastewater pollutant over extended periods of time needs to be followed up. Thus frequent collection of wastewater samples and analysis of the concentrations of the different pollutants (organic matter, phosphorus, nitrogen and bacteria) in influent and effluent are necessary. Such follow-up work will be of great value in assessing the service life span of biochar filters and the required frequency of renewal of the biochar. This activity is planned to be performed in an extension of the project.

All the biochar filter studies presented in this report were designed mainly for removal of organic matter and no consideration was taken in their design to providing optimal conditions for nitrogen or phosphorus removal. Nonetheless, the removal of nitrogen and phosphorus achieved was promising and the biochar shows high potential for better nutrient removal if the right process conditions and relevant design parameters are provided. Work on optimising the nitrogen and phosphorus removal in biochar filters has started, by modifying the filter layout, using saturation flow for nitrogen removal and functionalising the surface of the biochar (modifying it by adding chemical functional groups and impregnating it with lime and iron) to enhance phosphorus precipitation.

Development of integrated design criteria for dimensioning of biochar filters for removal of different types of pollutant is needed. Thereafter, a guiding document (e.g. design manual or guidelines) will need to be published to facilitate design and construction of biochar beds by different actors (environmental inspectors, entrepreneurs and even householders).

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