Science of the Total Environment 548–549 (2016) 100–109

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Transport and retention of bacteria and viruses in biochar-amended sand

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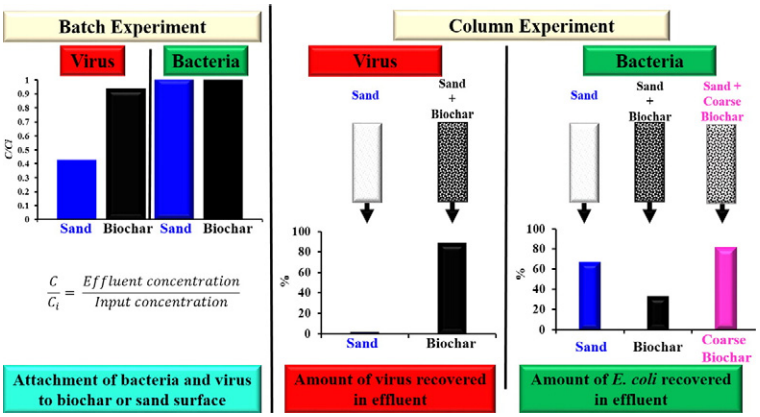
HIGHLIGHTS

- Negligible attachment of bacteria and viruses to biochar particles
- Enhanced transport of virus in the biochar-amended sand
- Enhanced retention of bacteria in biochar-amended sediment

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 13 November 2015
Received in revised form 21 December 2015
Accepted 24 December 2015
Available online 20 January 2016

Keywords:
Biochar
Porous media
E. coli bacteria
Bacteriophages MS2
PRD1
ΦX174
Straining
Attachment

ABSTRACT

The transport and retention of *Escherichia coli* and bacteriophages (PRD1, MS2 and ΦX174), as surrogates for human pathogenic bacteria and viruses, respectively, were studied in the sand that was amended with several types of biochar produced from various feedstocks. Batch and column studies were conducted to distinguish between the role of attachment and straining in microbe retention during transport. Batch experiments conducted at various solution chemistries showed negligible attachment of viruses and bacteria to biochar before or after chemical activation. At any given solution ionic strength, the attachment of viruses to sand was significantly higher than that of biochar, whereas bacteria showed no attachment to either sand or biochar. Consistent with batch results, biochar addition (10% *w*/*w*) to sand reduced virus retention in the column experiments, suggesting a potential negative impact of biochar application to soil on virus removal. In contrast, the retention of bacteria was enhanced in biochar-amended sand columns. However, elimination of the fine fraction (<60 μm) of biochar particles in biochar-amended sand columns significantly reduced bacteria retention. Results from batch and column experiments suggest that land application of biochar may only play a role in microbe retention via straining, by alteration of pore size distribution, and not via attachment. Consequently, the particle size distribution of biochar and sediments is a more important factor than type of biochar in determining whether land application of biochar enhances or diminishes microbial retention.

Abbreviations: DLVO, Derjaguin–Landau–Verwey–Overbeek theory; C_i, Initial microbial concentration; C_f, Final microbial concentration (batch); C, Effluent concentration (column); PV, Pore volumes; Φ_{max}, Energy barrier against primary minimum attachment; $\Phi_1{}^0{}_{min}$, Depth of the primary minimum; T_H, Applied hydrodynamic torques; T_A, Resisting adhesive torques; PRT, Percentage of microbes retained.

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http://dx.doi.org/10.1016/j.scitotenv.2015.12.126
0048-9697/

1. Introduction

Biochar is a stable form of carbon that is produced by pyrolysis of biomass (e.g., grass, crop or woody residue) under a limited supply of oxygen (Kumari et al., 2014; Wang et al., 2013b). Recently, biochar has gained interest due to its use as a soil amendment to simultaneously mitigate anthropogenic climate change whilst improving soil fertility and enhancing crop production (Lehmann et al., 2006; Mukherjee and Lal, 2014). Extensive studies on benefits of biochar application have been reported related to soil fertility (Doan et al., 2015; Glaser et al., 2002), physical properties (Mukherjee, 2013), microbial community and biota (Jindo et al., 2012; Lehmann et al., 2011), carbon sequestration and greenhouse gas emissions (Lehmann and Joseph, 2015; Mukherjee et al., 2014). In addition, a number of studies have shown that certain biochars are very effective sorbents, especially for chemical contaminants such as pesticides and heavy metals (Cui et al., 2016; Kearns et al., 2014; Kookana, 2010; Macdonald et al., 2015). Literature also indicates that biochar application to natural porous media (e.g., soil) may enhance pathogen retention (Abit et al., 2012, 2014; Mohanty and Boehm, 2014; Mohanty et al., 2014).

Mechanisms that control retention of microbes, and in general colloids, in porous media include attachment to and detachment from solid (collector) surfaces and physical entrapment (straining) in small pore spaces (Torkzaban and Bradford, 2016; Torkzaban et al., 2015). Colloid interactions with solid surfaces have been explained using the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory (Derjaguin, 1941; Verwey and Overbeek, 1955). DLVO theory states that the interaction energy can be quantified as the sum of van der Waals and electrostatic double layer interactions, which can be either attractive or repulsive. The strength of interaction is therefore controlled by various physical and chemical factors such as pH, ionic strength (IS), temperature, presence of organic matter, metal oxides, and multivalent-ions like calcium (Torkzaban et al., 2013; Da Silva et al., 2011; Foppen et al., 2008; Foppen et al., 2006; Furiga et al., 2010; Kim and Walker, 2009; Bradford et al., 2014; Redman et al., 2004; Sadeghi et al., 2013; Schijven and Hassanizadeh, 2000; Wong et al., 2013; Zhang et al., 2012). For example, an increase in pH, which is commonly observed in biochar-amended porous media (Mosley et al., 2015), may increase electrostatic double layer repulsion and consequently enhance transport of microbes in the porous media. Recently, nanoscale surface roughness and chemical heterogeneity on the collector (e.g. sand grains) and colloid surfaces have been shown to play a significant role in the interaction energy between a colloid and collector (Bradford and Torkzaban, 2013, 2015). It is expected that biochar particle size and their physical and chemical surface properties will be similarly important factors influencing the extent of microbial retention in biochar-amended soil.

Straining is another potential mechanism for retention of pathogens. It involves retention of colloids in smallest regions of pore space such as those formed near grain-to-grain contact points and microscopic roughness locations. Staining may also occur in pore throats that are too small to allow passage of single or multiple colloids (Torkzaban et al., 2015). It is expected that the presence of micro and macro-porous structure on the surface of biochar particles and micro-sized biochar particles (e.g. a few micrometers) can lead to an enhanced retention of colloids in biochar-amended porous media (Bradford et al., 2014; Hale et al., 2014). For example, the existence of microscale porous structures on the surface of biochar particles can create low-velocity regions where microbes can be retained via a shallow secondary energy minimum (Mohanty and Boehm, 2014). The relative importance of colloid retention by attachment and straining depends on properties of colloid (both in size and concentration), porous medium (porosity, grain size, and roughness), the hydrodynamic conditions, and the solution chemistry (Bradford and Torkzaban, 2013, 2015).

Batch and column experiments are common methods to study colloid retention in porous media. These experimental techniques offer the advantage that retention mechanisms can be examined under well-defined laboratory conditions. The solid phase in batch systems is continuously mixed and, therefore, the flow direction changes over time. This agitation facilitates collision of colloids to solid surfaces and possibly increases the attachment rate. However, this agitation also eliminates pore structure and continuously changes the applied and adhesive torques that contributes to colloid retention, especially at microscopic roughness locations on the solid phase (Treumann et al., 2014). Hence, attachment controls colloid retention in batch systems. Conversely, packed-column experiments are commonly utilized to analyze colloid breakthrough curves (BTCs) and the retention profiles. The solid phase in column experiments is stationary, colloids that are retained at locations associated with microscopic roughness, and grain-grain contacts always experience a low applied torque and a greater adhesive torque. The solid surface contributing to microbe retention is therefore expected to be greater in the column than batch systems because of attachment and straining processes (Treumann et al., 2014). Comparison of retention results from batch and column studies can, therefore, be utilized to determine the relative importance of attachment and straining processes.

Recently, a few column studies have been undertaken to investigate transport of various types of bacteria in biochar-amended porous media (Abit et al., 2012, 2014; Bolster and Abit, 2012; Chung et al., 2014; Mohanty and Boehm, 2014; Mohanty et al., 2014). Abit et al. (2012) reported that *E. coli* retention was enhanced in a high temperature pyrolyzed biochar amended-soil compared to a low temperature pyrolyzed biochar amended-soil or soil only columns. Increasing the amount of biochar in soil increased the extent of bacteria retention (Abit et al., 2012). Chung et al. (2014) reported enhanced retention of *E. coli* in sand-packed columns containing a potassium hydroxide activated (93%) or raw maize (72%) hydrochar compared to unamended sand (~30%). To understand the retention mechanism, a backwashing test was performed following the retention phase. A considerable fraction of the retained bacteria was recovered in this phase implying that straining might have been the underlying retention mechanism (Chung et al., 2014). Mohanty and Boehm (2014) reported an enhanced removal (~96%) of *E. coli* in a biochar-amended sand compared to unamended sand (~37%). However, it was observed that elimination of fine biochar particles (<125 μm) in the biochar-amended column considerably decreased the retention capacity (~62%) (Mohanty and Boehm, 2014). This limited number of studies on the efficacy of biochar on bacteria removal indicates that mechanisms and factors controlling bacteria retention in the presence of biochar are still poorly understood. Moreover, to date, no study has been published on the transport and retention of viruses in biochar-amended porous media.

The aim of this study was to gain a better understanding of the underlining mechanisms that control transport and retention of microbes (bacteria and viruses) in the biochar-amended sand. To achieve this, systematic experiments were conducted using various types of biochars, ultra-pure quartz sand, and *Escherichia coli* and phages (PRD1, MS2, and ΦX174). First, batch experiments with biochars or sand were conducted under varying solution chemistries. Batch experiments were used to specifically examine the extent of microbial attachment to biochar and sand surfaces. In addition, the impact of chemical activation of biochars on microbial attachment was examined in the batch experiments. Then, a series of column experiments using sand amended with various types of biochar were conducted to understand the combined effect of attachment and straining on the microbe retention. Comparison between batch and column experiments using viruses and bacteria helped us identify the controlling retention mechanism in the biochar-amended sand.

2. Materials and methods

2.1. Porous media characterization

Biochar samples employed in this research were obtained from feedstocks of Macadamia Shell (MS), Oil Mallee (OM), Phragmites Reed

(PR), Rice Husk (RH) and Wheat Chaff (WC). These biochars are currently being assessed for various potential applications, including: sorption of active pharmaceutical ingredients (Williams et al., 2015), mycorrhizal root colonization, growth and nutrition of wheat (Solaiman et al., 2010), immobilization of soil cadmium (Zhang et al., 2013), pH neutralization (Mosley et al., 2015), and efficiency to decrease N volatilization (Mandal et al.). Specific characteristics (feed stock type, pyrolysis temperature, pH, specific conductivity, and density) of the biochar samples are given in the supporting information (SI) Table S1. Biochar samples were crushed and sieved (under running water) to a size <2 mm and >60 μm. Therefore, we expect the presence of fine particles smaller than 60 μm was negligible in our batch tests. Particle size distribution information for the biochar samples is given in Table S2. Scanning electron microscopy (SEM) imaging (FEI Quanta 450 FEG Environmental SEM, US) was conducted on biochar samples to observe their structure before and after washing (Fig. S3).

Activation of biochar has received considerable research attention to enhancing its adsorption capacity (Chung et al., 2014). Chemical activation of biochar may affect its physical and chemical characteristics (e.g., surface area, porosity, micro-pore volume, the presence of surface charge group and iso-electric point) and, therefore, influence its efficiency for contaminant removal (Molina-Sabio and Rodríguez-Reinoso, 2004; Trakal et al., 2014). Wheat Chaff (WC) and Oil Mallee (OM) biochars were activated using the steps reported in Azargohar and Dalai (2008), which is briefly described in the SI. Hereafter, these samples are designated as $WC_{0.1\ M\ NaOH}$, $WC_{0.05\ M\ NaOH}$, $WC_{0.1\ M\ HNO3}$, $WC_{0.05\ M\ HNO3}$, $OM_{0.1\ M\ NaOH}$, $OM_{0.05\ M\ NaOH}$, $OM_{0.1\ M\ HNO3}$ and $OM_{0.05\ M\ HNO3}$.

Ultra-pure quartz sand (Charles B. Chrystal CO., Inc., NY, USA) with size ranging from 125 to 300 μm was cleaned as described by (Sasidharan et al., 2014) and used in the experiments. All five biochar samples were used for batch experiments, whereas only WC and OM biochars were used in the column studies because of the limited availability of the other biochar samples. Raw WC or OM biochar particles (grounded and dry sieved <2 mm) were mixed with sand (biochar-amended sand) to achieve a 0.1 *w*/*w* ratio (10%) for column experiments. This corresponds to a volume percentage of 50% for the biochar and sand mixture. Hereafter, these biochar-sand mixtures were designated as 'WC-Sand' and 'OM-Sand'.

2.2. Microbe preparation

Escherichia coli 13706 (ATCC 13706) was used as a surrogate for pathogenic bacteria. The bacteria sample preparation method is explained in detail in the SI. Phages (MS2, ΦX174, and PRD1) used in this study are surrogates for human pathogenic viruses (Schijven and Hassanizadeh, 2000). Characteristics of phages and their respective host bacteria are given in Table S3. The detailed methodology for bacteriophage preparation and enumeration is given in the SI.

2.3. Interaction energy calculations

The biochar was mixed in a selected electrolyte solution and filtered through a <5 μm filter. The size and zeta potential for the fine biochar fraction that passed through the filter was measured (Nano ZS, Malvern Instruments Ltd., UK). Zeta potentials for crushed sand grains, phages, and bacteria in electrolyte solutions were also measured with this instrument. Measured zeta potentials in the various pH and electrolyte solutions were used to calculate the interaction energy profile for phages and bacteria upon their close approach to sand and biochar surfaces. Sphere-plate interaction energy calculations were conducted by assuming that microbes were spherical and collector surfaces were smooth. The van der Waals interaction (*vdW*) was determined from the expression of Gregory (Gregory and Wishart, 1980). The combined Hamaker constant was estimated from the Hamaker constant of individual materials (Israelachvili, 1992). An individual value of 3.70×10^{-20} for water (Israelachvili, 1992), 6.50×10^{-20} for sand (Israelachvili, 1992), 6.19×10^{-20} for biochar (Wang et al., 2013b), 7.00×10^{-19} for *E. coli* (Capco and Yongchen, 2014) and 6.60×10^{-20} for viruses (Kavanaugh and James, 1980) were used in this study. The combined Hamaker constant was calculated to be 4.03×10^{-20} for *E. coli*–Water–Sand, 3.64×10^{-20} for *E. coli*–Water–Biochar, 4.04×10^{-21} for Virus–Water–Sand, and 3.64×10^{-21} for Virus–Water–Biochar systems. Electrostatic double layer interaction (V_{EDL}) was calculated using the Hogg–Healy–Fuerstenau expression (Hogg et al., 1966) with zeta potentials in place of surface potentials. Born repulsion was considered using the expression given by (Ruckenstein and Prieve, 1976). Hydrophobic interactions were not considered in these calculations because needed contact angle and surface tension information for the various biochar samples were not available.

2.4. Batch experiment

Batch experiments were conducted to determine the attachment behavior of phages and bacteria to sand and biochar surfaces at selected electrolyte concentration (5, 10 and 20 mM NaCl) in the absence of pore structure (e.g., the entire system is in motion). All electrolyte solutions in this study were prepared using 1 mM Tris buffer and the pH was adjusted to pH 7.2 using 0.1 M HCl. A detailed step by step method for the batch experiment is given in Section 1.4 of the SI. Triplicate measurements were performed for all experiments. Additionally, a set of control tubes with only phage or bacteria suspension were prepared to ensure the viability of these microbes over the course of experiments.

Additional batch experiments were performed to test the effect of calcium ion (5 mM $CaCl_2$ at pH 7.2) on microbe attachment to biochar samples (RH, OM, WC, and PR). The efficiency of activated WC and OM biochar to adsorb microbes was also tested using various electrolyte solutions (5, 10 and 20 mM NaCl & 5 mM $CaCl_2$).

The water samples from virus experiments were centrifuged at 1000 ×*g* for 10 min at 4 °C followed by filtrating the supernatant through a 0.45 μm syringe filter (Merck Millipore, Germany) to remove any biochar fine particles. The filtrate was enumerated for virus concentration using the method explained in Section 1.3 of the SI. This filtration step ensured that any possible interference of particle-associated viruses did not affect our results. Similarly, water samples from bacteria experiments were centrifuged at 100 ×*g* for 5 min at 4 °C, the supernatant was filtered through a 5 μm syringe filter (Merck Millipore, Germany), and the absorbance at 460 nm using a UV–Vis spectrophotometer was measured. In addition, 100 μl of the filtrate was serially diluted and spread plated to determine the CFU/mL. In both cases, the final concentration was statistically the same. Both absorbance and spread plate analysis were conducted for all the samples and the average concentration obtained from both methods was used in the determination of the cell concentration for each sample.

2.5. Column transport experiments

Columns for the transport experiments were prepared and the experiments were conducted using an electrolyte solution (1, 5, 10 or 20 mM NaCl in 1 mM Tris Buffer with pH 7.2) with suspended phage (PRD1 and ΦX174) or *E. coli* of a known C_i as explained in Section 1.5–1.6 in the SI. The effluent samples for both microbes were collected, processed and measured using the methods explained in above Section 2.4 and Sections 1.2 & 1.3 in the SI. The effluent breakthrough concentrations (BTCs) were plotted as dimensionless concentrations (C/C_i) of microbes as a function of the number of pore volumes (PVs). The total number of retained microbes during Phase 1 and 2 (N_{1+2}) was determined by calculating the difference between the number of injected microbes into the column in Phase 1 (N_{in}) and the number of microbes that was recovered in the effluent during Phase 1 and 2 (N_{out}). This information was used to calculate the percentage of retained microbes in each experiment. The survival of phages and bacteria over a

38.4 h interval was determined in the effluent from the preconditioning phase.

Statistical differences of mean removal efficiencies were identified by one-way ANOVA. The mean removal efficiencies were separated by Tukey's honestly significant difference (HSD) test ($p < 0.05$). All statistical analyzes were performed using IBM SPSS Statistics for Windows Version 22.0.

3. Result and discussion

3.1. Zeta potentials and interaction energies

Table S4 presents zeta potentials for the microbes, biochars, and quartz sand at pH 7.2 and IS of 20 mM NaCl and 5 mM $CaCl_2$. Surfaces of sand were less negatively charged than those of biochar particles at both solution chemistries. It has been reported that biochar can contain negatively charged functional groups such as carboxyl, hydroxyl, phenolic groups on its surface (Mandal et al., 2015; Mosley et al., 2015; Nartey and Zhao, 2014; Wang et al., 2013a). These functional groups are ionized and contributed to the net negative charge on the biochar surface under the tested pH conditions (Wang et al., 2013a). All microbes, sand, and biochar surfaces were more negatively charged in the presence of Na^+ than Ca^{2+} ions. Divalent cations, such as Ca^{2+}, more effectively decrease the absolute magnitude of the zeta potential than monovalent cations, like Na^+. This has been attributed to the combined effects of charge screening and binding of Ca^{2+} to anionic functional groups on natural surfaces (Sasidharan et al., 2014).

Table S5 presents interaction energy parameters, namely the height of the energy barrier against primary minimum attachment (Φ_{max}) and the depth of the primary minimum (${\Phi_1}^0{}_{min}$), for all the microbes interacting with sand and biochars at IS = 20 mM NaCl or 5 mM $CaCl_2$. A high value of Φ_{max} existed for both *E. coli* and viruses in the presence of 20 mM Na^+. The height of Φ_{max} tended to decrease with the microbe size (*E. coli* > PRD1 > ΦX174 > MS2, with *E. coli* being 30 times larger than MS2). Hence, the value of Φ_{max} was considerably larger for the *E. coli* than viruses. The value of Φ_{max} and ${\Phi_1}^0{}_{min}$ considerably decreased and increased, respectively, in the presence of Ca^{2+} compared to Na^+ electrolyte. This behavior is attributed to both microbes and collector surfaces being less negatively charged in the presence of Ca^{2+} (Table S4). Table S5 shows that the value of Φ_{max} was always greater than the average kinetic energy of diffusing microbes (1.5 kT) at all of the examined solution chemistry conditions. The Maxwellian kinetic energy model predicts that the probability for primary minimum attachment is small when $\Phi_{max} > 1.5$ kT, and approaches zero when $\Phi_{max} > 8$ kT (Chandrasekhar, 1943; Shen et al., 2007).

3.2. Batch experiments

Batch experiments over a wide range of chemical conditions were conducted to examine the extent of attachment of three different viruses (MS2, PRD1, and ΦX174) and *E. coli* to quartz sand and various types of biochars. Fig. 1 shows the normalized virus concentrations in equilibrated solutions (C_f / C_i; where C_i is the initial concentration and C_f is the final concentration) after 2 h mixing in tubes containing sand or various types of biochar at different concentrations of NaCl solution. Control tubes (without biochar or sand) confirmed stable virus concentration (i.e., no loss due to inactivation or attachment to tube wall) during the course of experiments (data not shown). It was observed that C_f / C_i reduction was negligible for all three viruses reacting with biochars in all solution chemistries. These observations clearly demonstrate negligible attachment of viruses to biochar particles. In comparison, values of C_f / C_i of the three viruses significantly decreased in tubes containing quartz sand, indicating significant ($p < 0.0002$) attachment to sand surfaces. The amount of attachment to sand grains increased with increasing IS.

Fig. S1 shows the results of batch experiments with *E. coli* reacting with various types of biochar or sand at different NaCl solution concentrations. The results show negligible *E. coli* attachment to both biochars and sand surfaces under all test conditions. It should be mentioned that all electrolyte solutions in this study were prepared using 1 mM Tris buffer and the pH was adjusted to pH 7.2 using 0.1 M HCl. Our preliminary tests, in which 20 mM unbuffered solution (without Tris) was used and the pH was lowered to 7.3 after several washing steps, showed negligible virus and *E. coli* attachment to biochar surfaces (data not shown). This result confirmed that the presence of 1 mM Tris in our buffered solution did not affect the adsorption process to biochar surfaces. Fig. S2 shows the results of batch experiments for viruses and *E. coli* in which the electrolyte solution was 5 mM $CaCl_2$. It was observed that the attachment of viruses to biochars (Fig. S2) only slightly increased ($p < 0.001$) in the presence of 5 mM Ca^{2+} in comparison with that of Na^+ solution (Fig. 1). However, virus attachment to quartz sand increased by more than 1 order of magnitude ($C_f / C_i < 0.1$) in the presence of 5 mM Ca^{2+} ($p < 0.0006$). In addition, it was observed that C_f / C_i values for *E. coli* showed little attachment to both sand and biochars under this high calcium concentration. Each of these observations will be further discussed in detail below.

Negligible attachment of *E. coli* and viruses to biochars in the various Na^+ solution chemistries was consistent with interaction energy parameters presented in Table S5. These calculations predict the presence of a sizable energy barrier against microbe attachment in a primary minimum. However, biochar surfaces are known to contain micropores of various sizes (micropores $< 2 \times 10^{-3}$ μm, mesopores 2–50 $\times 10^{-3}$ μm and macropores $> 50 \times 10^{-3}$ μm) (Downie et al., 2009; Shen et al., 2014). Fig. S3 shows representative SEM images of OM biochar confirming the presence of a large number of micro-hollow pores (1–50 μm), accessible for viruses and even *E. coli*, on the surface of biochar particles. The water velocity, and, therefore, hydrodynamic forces, is expected to be negligible in these micropores. Thus, a considerable amount of microbe attachment is expected in micropores when the adhesive energy (e.g., even a shallow secondary energy minimum) is larger than the thermal energy of diffusing microbes (1.5 kT). However, the secondary energy minimum was negligible for microbial interaction with biochar in Na^+ solutions, suggesting that the entire surface of biochar particles was unfavorable for attachment.

Negligible attachment of *E. coli* to sand surfaces agrees with previous batch studies using *E. coli* O157:H7 and *E. coli* D21g, and Ottawa and quartz sands (Bradford et al., 2015a, 2015b). Another study with carboxyl modified latex colloids (1 and 2 μm) also showed very little colloid attachment (<25%) on sand surfaces in batch systems, even when the IS was as high as 800 mM (Treumann et al., 2014). This small amount of colloid attachment was attributed to the continuous motion of sand in batch systems that altered the applied hydrodynamic (T_H) and resisting adhesive (T_A) torques with time. Both T_A and T_H are functions of the colloid radius (r_c), but T_H decreases more rapidly (proportional to r_c^3) than T_A with r_c. Consequently, nanoparticles such as viruses show higher attachment than micro-sized colloids (e.g., *E. coli*), provided the strength of the adhesion force is larger than the Brownian force (Bradford and Torkzaban, 2015).

Table S5 shows high values of Φ_{max} and negligible secondary minimum for virus interactions with sand surfaces in NaCl solutions. However, batch results show a considerable amount of virus attachment to sand grains (Fig. 1). A detachment experiment was conducted to better understand the nature of this virus–sand interaction. Following completion of a virus–sand batch experiment, the excess solution was removed and replaced with virus-free solution of the same chemical composition. The tubes were subsequently shaken for another 2 h, and the final virus concentration in the aqueous phase was measured. The virus concentration was again found to be negligible (data not shown), demonstrating that the detachment rate was very low and that attachment most likely occurred in a primary energy minimum. Recent studies have demonstrated that primary minimum attachment may occur even at low

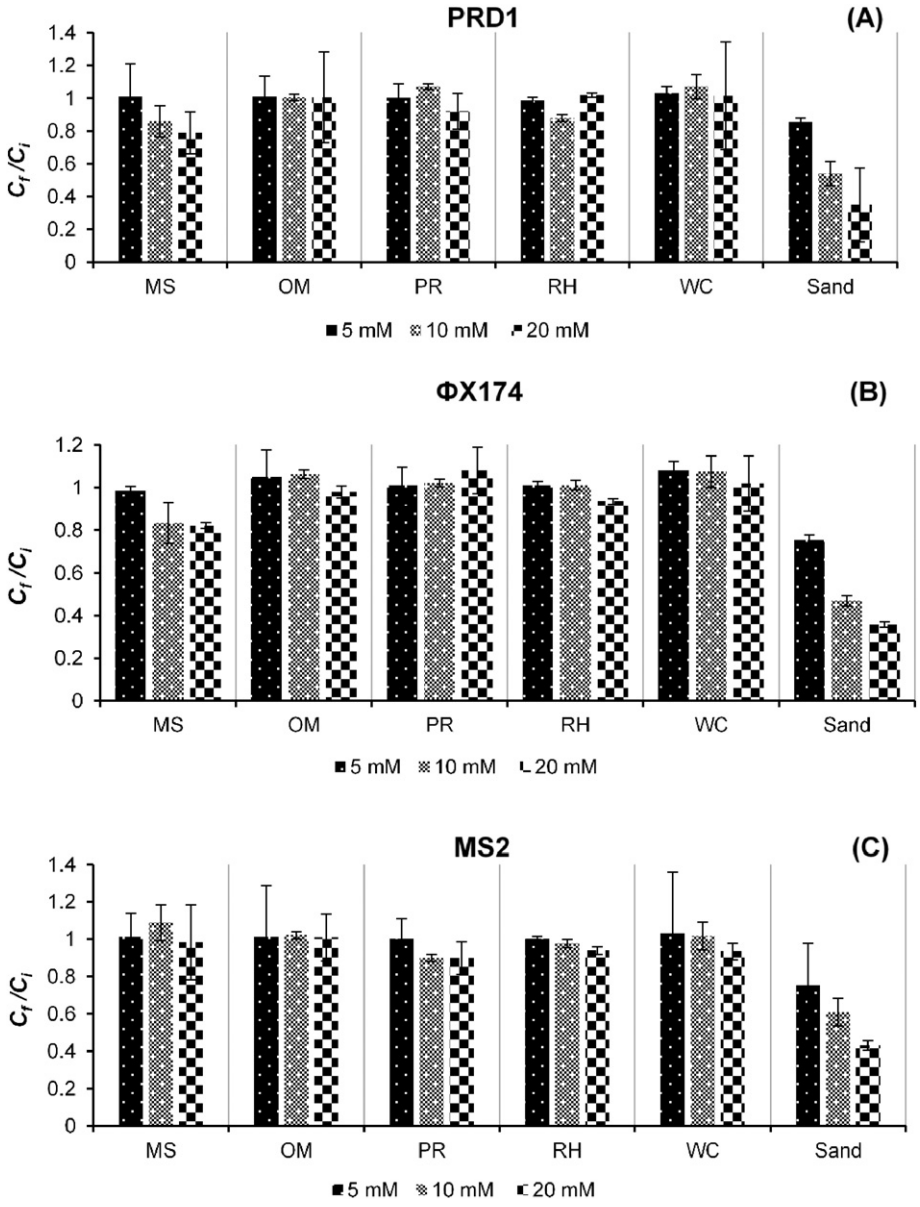

Fig. 1. Representative bar chart plot with error bar for bacteriophages: (A) PRD1 (B) ΦX174 and (C) MS2 obtained from batch experiments conducted using sand and five different biochar samples (Macadamia Shell [MS], Oil Mallee [OM], Phragmites Reed [PR], Rice Husk [RH] and Wheat Chaff [WC]) as adsorbing media. The parameters for experiment are IS = 5, 10 and 20 mM NaCl; pH = 7.2 and temperature = 18 °C. The Y-axis shows the normalized concentration C_f / C_i (C_i = initial concentration & C_f = final concentration) values. Error bars represent the standard error (n = 3).

solution IS when nanoscale surface roughness is incorporated into XDLVO calculations (Argent et al., 2015; Bradford and Torkzaban, 2013). Nanoscale roughness has been shown to reduce (or even eliminate) Φ_{max}, such that colloids can diffusive over the energy barrier. Hence, nanoscale roughness on viruses and sand surfaces provide a plausible explanation for the discrepancy in interaction energy parameters (Table S5) and batch results. Note that Φ_{max} was greater for biochar than sand (Table S5). Nanoscale roughness apparently did not reduce Φ_{max} enough to produce primary minimum interaction for microbes on the biochar.

Colloids such as *Cryptosporidium parvum* oocysts, viruses, and engineered nanoparticles have been observed to strongly attach to mineral surfaces in the presence of Ca^{2+}, even when DLVO theory predicted a substantial energy barrier (Torkzaban et al., 2013; Janjaroen et al., 2010; Sadeghi et al., 2013). In this study, the presence of Ca^{2+} only slightly enhanced attachment of *E. coli* to sand and biochar surfaces in comparison to Na^{+} (Figs. S1 and S2). In contrast, the presence of calcium in solution significantly enhanced the attachment of viruses (by more than one order of magnitude) to quartz surfaces (Figs. 1 and S2). However, much smaller amounts of virus attachment occurred on biochar samples than sand, and only slight attachment (C_f / C_i = 0.5) of MS2 and ΦX174 occurred on OM biochar. These differences in the influence of Ca^{2+} on attachment with microbe size and the solid surface can be explained in terms of nanoscale chemical heterogeneity. In particular, multivalent cations (e.g., Ca^{2+}) have been shown to strongly bind to negatively charged mineral surfaces and anionic functional groups of microbes (Torkzaban et al., 2013; Greenland, 1971; Sposito, 2008). This adsorption can create nanoscale chemical heterogeneity as a result of charge neutralization and/or reversal (De Kerchove and Elimelech, 2008; Sasidharan et al., 2014). Consistent with the experimental observations, the influence of nanoscale chemical heterogeneity on attachment has been shown to become more important for smaller colloids (such as viruses) and higher IS (Bradford and Torkzaban, 2013, 2015; Duffadar et al., 2009). Furthermore, the influence of nanoscale heterogeneity is expected to be diminished when the solid surface exhibits a greater net negative charge (biochars) because it is more difficult to

eliminate a higher energy barrier (Table S5). It should be mentioned that nanoscale chemical heterogeneity may be related to bridging complexation or "cation bridging" (Greenland, 1971). Bridging complexation occurs when anionic or polar functional groups (typically carboxylate-terminated molecules) bind with multivalent cations that are adsorbed on negatively charged surfaces (Sposito, 2008).

Additional experiments were conducted to examine the role of biochar activation on microbial attachment. A few studies have suggested that chemical activation of biochar improved its retention capacity of various contaminants (Azargohar and Dalai, 2008; Chung et al., 2014; Molina-Sabio and Rodríguez-Reinoso, 2004; Trakal et al., 2014). For example, activation with HCl led to the generation of more available sites on the surface for nutrient retention (Li et al., 2014). Activation of hydrochar with a 1 M KOH also showed an increase in *E. coli* removal by 21% in column experiments compared with that of raw hydrochar (Chung et al., 2014). Figs. 2 and S4 show batch results for three viruses and *E. coli*, respectively, on activated WC and OM biochars in the presence of 20 mM NaCl solution. The four different activation solutions (0.1 M NaOH, 0.05 M NaOH, 0.1 M HNO_3 and 0.05 M HNO_3) did not show a large influence on virus and bacteria attachment to the biochars. For example, activation of WC and OM biochar with 0.1 M NaOH did show a very slight improvement in attachment of PRD1 and ΦX174, and 0.1 M NaOH activation of OM enhanced the attachment of ΦX174 by 37% compared to the unactivated OM (Fig. 1). However, differences in microbe attachment to either activated or non-activated biochar samples were not statistically different ($p < 0.11$), and microbe attachment was always significantly ($p < 0.0001$) lower on activated biochar than quartz sand.

3.3. Column experiments

The addition of 10% *w*/*w* biochar to sand resulted in a considerable increase in total organic carbon and a negligible change in effluent pH in the biochar-amended column because of the high buffering capacity of the influent (1 mM Tris Buffer, pH 7.2) solution. Moreover, no detectable change in other water quality parameters (e.g., specific conductivity, phosphate, dissolved organic carbon) was observed in the effluent after flushing the column with 10 PVs of the background solution.

Fig. 3 presents representative effluent BTCs for PRD1 and ΦX174 from biochar-amended and unamended (sand) packed columns when the solution contained 10 mM Na^+. Here the normalized effluent concentration (C/C_i) is plotted against the number of PVs. Table 1 shows the percentage of retained (PRT) viruses for similar column experiments for the various IS levels and biochar types. Virus retention was significantly lower ($p < 10^{-7}$) in biochar-amended than unamended sand columns. Notably, the unamended (only sand) column retained ~2 log of PRD1 (~98.7%) and ΦX174 (97.4%) when the IS was 10 mM Na^+. However, always <50% of the input viruses was retained in the column when the sand was amended with biochar (Table 1). These results also show that the tailing of BTCs approached zero after a few PV injection of the virus-free solution, indicating that the detachment rate of the retained viruses was very low. Therefore, virus retention in these

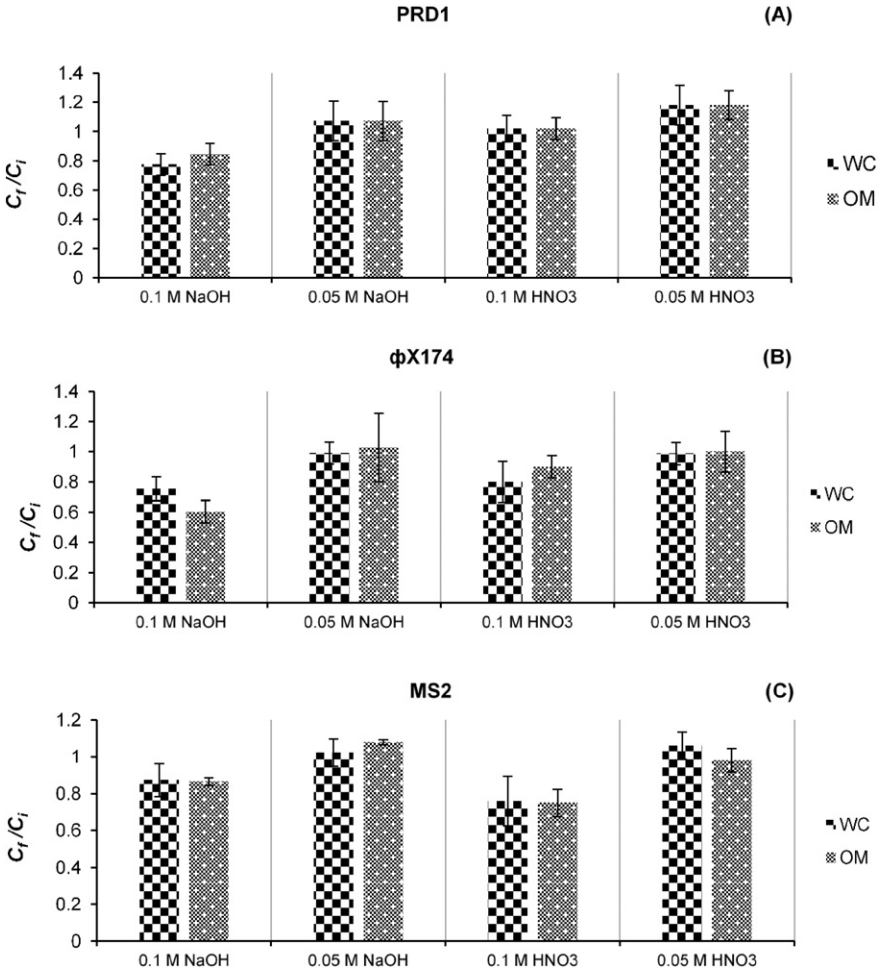

Fig. 2. Representative bar chart plot with error bar for bacteriophages PRD1, MS2 and ΦX174 obtained from batch experiments conducted using activated biochar. The biochar was activated using 0.1 M NaOH, 0.05 M NaOH, 0.1 M HNO3 and 0.05 M HNO3. (A) PRD1 (B) ΦX174 and (C) MS2 for activated biochar. The parameters for experiment are IS = 20 mM NaCl, pH = 7.2 and temperature = 18 °C. The Y-axis shows the normalized concentration C_f / C_i (C_i = initial concentration & C_f = final concentration) values. Error bars represent the standard error (n = 3). The Wheat Chaff (WC) and Oil Mallee (OM) biochar samples were used in this study.

experiments was primarily attributed to irreversible attachment to sand surfaces. These results are consistent with those obtained from the batch experiments that showed a negligible virus attachment to biochar particles and a considerable attachment to sand surfaces.

Fig. 4 presents representative BTCs for *E. coli* in biochar-amended and unamended sand columns when the solution IS = 20 mM NaCl. Table 1 provides PRT values for other biochar experiments at different IS conditions. In contrast with the results obtained for viruses, higher bacteria retention was observed in biochar-amended than unamended sand columns. For example, the addition of OM or WC biochar to sand increased retention of *E. coli* by ~60%. This result is consistent with previous studies which reported an enhanced bacteria retention after biochar addition to porous media (Abit et al., 2012, 2014; Mohanty and Boehm, 2014; Mohanty et al., 2014). The amount of bacteria retention in biochar amended sand was not dependent on the solution IS, as the PRT values were practically the same for 5 and 10 mM experiments (Table 1). Conversely, an increase in bacteria retention was observed with increasing IS in experiments with unamended sand (Table 1). Other researchers have reported a similar dependence of bacteria retention on IS in packed sand columns (Li et al., 2004; Tufenkji and Elimelech, 2004, 2005). It should be mentioned that negligible *E. coli* retention occurred in the unamended sand column when deionized water (IS = 0) was used as the background solution (data not shown), implying that physical straining was negligible.

As noted previously, interaction energy calculations presented in Table S5 indicated that attachment in the primary or secondary minimum was not expected for *E. coli* interacting with biochar and sand particles under the current experimental conditions. In addition, batch experiments showed negligible bacteria attachment to sand and biochar particles in the considered solution chemistries. The batch results demonstrated that the adhesive interaction energy between the bacteria and surfaces of sand and biochar particles was not strong enough to produce attachment. This inconsistency between the results of batch and column experiments with unamended sand (without biochar) can be attributed to the coupled effect of hydrodynamic forces and microscopic roughness on retention. In particular, colloid retention is well-known to depend on the balance of T_H and T_A at a particular location on a solid surface (Bradford and Torkzaban, 2015). Colloid retention is expected to predominantly occur at locations associated with large scale roughness, ridges, and valleys on sand grains because these locations are associated with larger T_A and lower T_H (surface topography influences the lever arms). In batch experiments, however, the direction and magnitude of T_H and T_A at a particular location on the grain surface are continuously altered with time (Treumann et al., 2014). Thus, the torque balance criterion is not be satisfied in a batch system. In contrast, the direction and magnitude of T_H and T_A are constant at a particular location on the sand surface in the static column system under steady-

Table 1
Percentage of retention (PRT) for bacteriophages (PRD1 and ΦX174) and *E. coli* in various experiments (sand, sand + biochar or sand + coarse biochar). The experiment parameters are IS = 5, 10 and 20 mM NaCl, pH = 7.2, injection pore volume = 20 PV, flow velocity = 1 m/day and temperature = 18 °C. The Wheat Chaff (WC) and Oil Mallee (OM) biochar samples were used in these studies.

Colloid	Porous media	IS	Percentage of retention
		[mM]	[%]
PRD1	Sand	10	98.7 ± 0.2
	WC-Sand		11.3 ± 2.1
	OM-Sand		50.1 ± 1.9
ΦX174	Sand	10	97.4 ± 0.4
	WC-Sand		25.9 ± 2.5
	OM-Sand		45.1 ± 2.1
E. coli	Sand	5	10.2 ± 1.9
	Sand	10	26.6 ± 1.7
	Sand	20	32.8 ± 1.2
	WC-Sand	5	64.5 ± 1.2
	WC-Sand	10	67.1 ± 1.3 ǂa
	WC-Sand	20	67.8 ± 1.3 a
	OM-Sand	5	65.2 ± 1.7 b
	OM-Sand	10	66.0 ± 1.4 b
	OM-Sand	20	67.4 ± 2.9
	$WC_{coarse\ textured}$-Sand	5	5.3 ± 1.1
	$WC_{coarse\ textured}$-Sand	10	9.6 ± 1.5
	$WC_{coarse\ textured}$-Sand	20	12.1 ± 1.1
	$OM_{coarse\ textured}$-Sand	5	8.5 ± 2.8
	$OM_{coarse\ textured}$-Sand	10	16.5 ± 1.9
	$OM_{coarse\ textured}$-Sand	20	18.5 ± 1.1

ǂ: Within the column, mean percentage of retention (PRT) values followed by the same letter are not significantly different using Turkey's HSD test at $p < 0.05$.

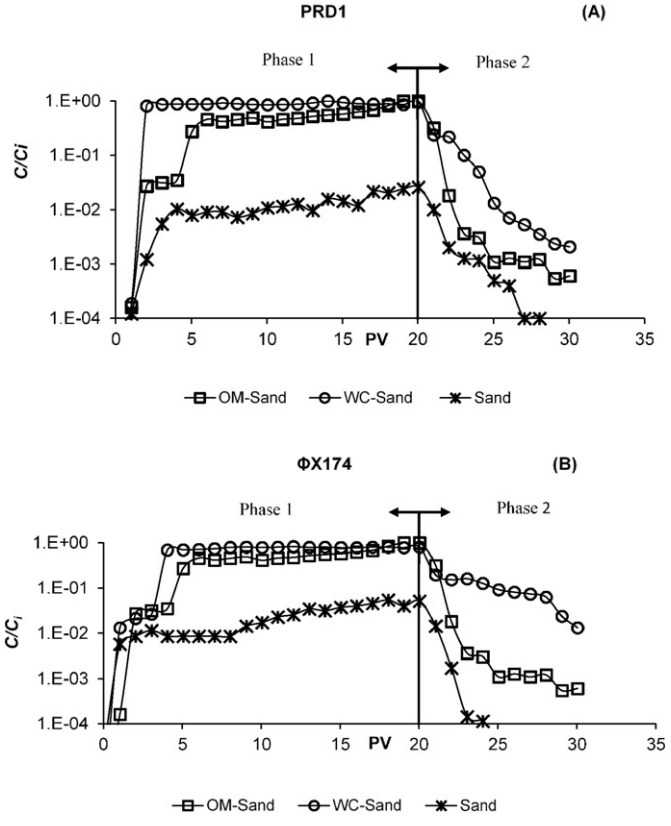

Fig. 3. Representative measured BTCs for bacteriophages (A) PRD1 and (B) ΦX174 obtained from column experiments using biochar-amended and non-amended porous media (WC-Sand, OM-Sand and Quartz sand only) at IS = 10 mM, pH = 7.2, flow velocity = 1 m/day and temperature = 18 °C. The Wheat chaff (WC) and Oil Mallee (OM) biochar samples were used in this study.

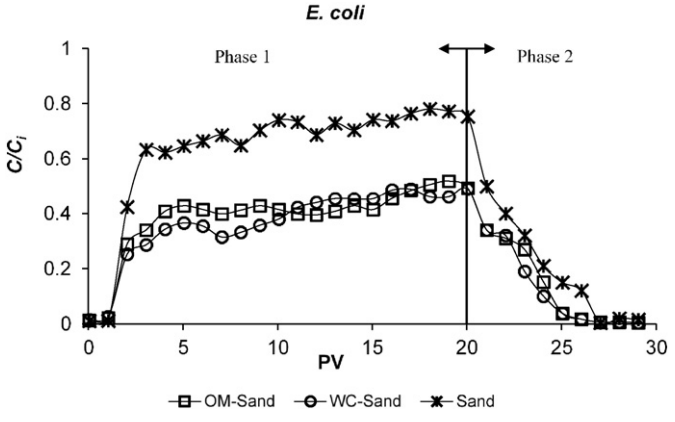

Fig. 4. Representative measured BTCs for *E. coli* bacteria obtained from column experiments using biochar-amended and non-amended porous media (WC-Sand, OM-Sand and Quartz Sand only) at IS = 20 mM NaCl, pH = 7.2, flow velocity = 1 m/day and temperature = 18 °C. The Wheat Chaff (WC) and Oil Mallee (OM) biochar samples were used in this study.

state conditions. Consequently, the negligible colloid attachment in batch experiments and the significant retention in the column experiments with sand demonstrate the importance of microscale surface roughness on bacteria retention in porous media.

In previous experiments conducted with biochar-amended porous media, enhanced bacteria retention was ascribed to increases in the specific surface area leading to increased attachment sites after the biochar addition (Mohanty and Boehm, 2014). Biochar is highly porous relative to sand, thus the surface area of biochar is at least 5 orders of magnitude larger than sand (Mohanty et al., 2014). Moreover, the enhanced bacteria retention has been attributed to stronger attachment of bacteria to surfaces of biochar particles than that of sand surfaces. Non-DLVO forces including hydrophobic and steric interactions were suggested to cause the strong attachment of bacteria to biochar particles (Mohanty et al., 2014). Hydrophobic attraction is expected to be much greater between bacteria and biochar than bacteria and sand due to the high organic carbon content of biochar (Abit et al., 2012). Thus, it has been proposed that biochar may retain *E. coli* at the primary minimum due to the increased hydrophobic interactions (Abit et al., 2012). However, strong attachment or increased attachment sites are unlikely to be the dominant mechanisms causing the enhanced bacteria retention in the biochar-amended sand in this study. If attachment was the dominant mechanism, then bacteria attachment would have been observed in the batch experiments with biochar. However, cell attachment in the batch experiments was not observed. Moreover, no attachment to biochar was observed for the three different viruses, that encompass a wide range of hydrophobicity and isoelectric points (Aronino et al., 2009; Chrysikopoulos and Syngouna, 2012; Dika et al., 2015; Schijven and Hassanizadeh, 2000).

Another explanation for the observed enhanced bacteria retention in the biochar-amended sand is physical straining. This explanation seems to be more reasonable given that biochar addition did not cause an increase in virus retention in the column experiments. Note that straining increases with the microbe size, and *E. coli* is more than 30 times larger than a virus (e.g., ΦX174). Additional column experiments were conducted to investigate whether physical straining was responsible for the effect of biochar amendment on bacteria retention. These experiments were conducted in a similar manner to others, with the exception that the crushed biochar materials were sieved to remove the fine fraction (<60 μm); e.g., only the coarse biochar fraction (60 μm–2 mm) was used to amend the sand. Fig. 5 presents BTCs for *E. coli* in coarse-textured biochar amended sand at different solution IS. Retention of *E. coli* was significantly lower ($p < 10^{-7}$) in coarse (60 μm – 2 mm) than fine (<2 mm) biochar amended sand. Note that the PRT in the coarse-textured biochar-amended sand was even smaller (~12–18%) than that of unamended sand (~33%), indicating the importance of sand surface area for bacteria retention (Table 1). A recent study found that considerable amounts of biochar micro-particles (in the order of a few micrometers) were retained at pore constrictions when a stable biochar micro-particle suspension was injected into a packed sand column (Wang et al., 2013a; Zhang et al., 2010). These results demonstrate that the fine fraction in the experiments were the dominant fraction responsible for the enhanced bacteria retention.

Pore straining is the trapping of colloidal particles, in this case, bacterial cells in the down-gradient pore throats that are too small to allow colloid passage (McDowell-Boyer et al., 1986). The magnitude of colloid retention by straining depends on both the colloid and porous medium properties (Bradford et al., 2013). Natural porous media (e.g., soil) typically exhibit a wide range in pore sizes due to variations in grain size, orientation, and configuration. Biochar also exhibits a wide range of particle sizes varying from a fraction of micrometers to a few millimeters. Recall that the results from unamended sand suggested that little straining occurred when the IS was very low (~0 mM). In contrast, when the sand was amended with biochar, a fraction of fine particles in the biochar was retained in small pores during the packing or equilibration phase. This process will decrease the

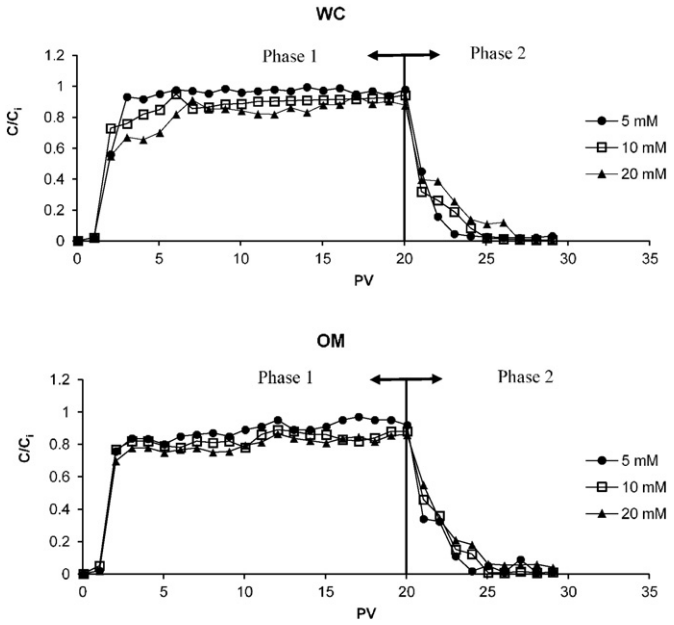

Fig. 5. Representative measured BTCs for *E. coli* bacteria obtained from column experiments using biochar-amended porous media (WC coarse $_{texture}$-Sand and OM coarse $_{texture}$-Sand) at IS = 5, 10 and 20 mM NaCl, pH = 7.2, flow velocity = 1 m/day and temperature = 18 °C. The Wheat chaff (WC) and Oil Mallee (OM) biochar samples were used in this study.

effective pore sizes of the porous media and may, therefore, increase the likelihood of subsequent bacteria retention in the narrow pores. When the colloid size is considerably smaller than the sand pore sizes (e.g., viruses), straining becomes a less dominant mechanism of colloid retention. Pore and surface (microscopic roughness and grain-grain contacts) straining of bacteria in uniformly sized sands has been shown to be an important factor affecting bacterial retention when the ratio of bacteria diameter to sand grain diameter is >0.007 (Bradford et al., 2014). In this current study, the flow velocity was constant and there was no detectable permeability reduction in the biochar-amended sand as the overall permeability was very high (50 m/day).

The effect of biochar amendment on the extent of bacterial retention has been observed to be dependent on the type of biochar and soil. For instance, Abit et al. (2014) examined the effect of biochar addition on the transport of bacteria in sand and soil columns. They found that bacteria retention decreased (13%) when a low-temperature poultry litter (LTPL) was added to a sandy loam soil. Conversely, the addition of the LTPL biochar had no major effect on the retention in a fine sand medium (7%). Moreover, the addition of a high-temperature poultry litter (HTPL) biochar to the fine sand increased the *E. coli* retention and had no discernible effect in the sandy loam (Abit et al., 2014). Note that biochar produced at higher pyrolysis temperature generally have a much greater fraction of fine particles, specific surface area and hydrophobicity (McBeath et al., 2015; Wang et al., 2013a). These observations were attributed to differences in hydrophobicity values for bacteria suspended in leachates collected from the fine sand and sandy loam amended with biochar. However, based on the results of the current study, a more likely explanation for some of the observed changes in bacteria retention after biochar addition is the potential for increasing or decreasing straining of bacteria in porous media. For example, given the size distribution of loamy sand, it is likely that LTPL biochar addition resulted in a coarser-textured porous media compared to the unamended media and, therefore, the contribution of straining was diminished. In another study by Chung et al. (2014) when the column was flushed with DI water after the retention phase, only a minor fraction (~3%) of the retained *E. coli* was released. Conversely, when the column was flushed in the reversal mode (backwashing), a considerable

fraction (~22%) of the retained bacteria was released, implying that straining was the underlying retention mechanism in the biochar-amended column.

4. Conclusion

In this study, batch experiments showed negligible attachment of viruses and bacteria to biochar surfaces before and after chemical activation. At a given chemical condition, the attachment of viruses to sand was much higher than to biochar surfaces. In this study, the column experiments demonstrated that the biochar-amendment of sand enhanced the transport of viruses. In contrast, retention of bacteria was enhanced in a biochar-amended sand column. In this study, the particle size of biochar was found to be important in retention of bacteria. The removal of a fine fraction of biochar particles (<60 μm) enhanced the transport of bacteria during column experiments. Together these results demonstrate that the enhanced retention of bacteria in the biochar-amended sand is a result of straining of bacteria in pore constrictions, grain-grain contact points, and/or microscopic roughness locations.

This study was conducted using pure quartz, which is an example for worst case scenario of microbial attachment to a collector surface. Even though the interaction energy calculations showed that experimental conditions were unfavorable for attachment, batch and column experiments showed significant ($p < 0.0001$) retention of bacteriophages to quartz surface. Natural soil and sediments typically contain metal oxides and clay particles, which will enhance the number of favorable sites available for attachment and thus increase the potential of microbial retention (Tong et al., 2012; Truesdail et al., 1998). Biochar contains a large number of nano- and micro-size biochar particles. These particles can compete for favorable attachment sites (metal oxides, clay) available on the soil surface. This process may further reduce the removal of colloids like pathogenic viruses or toxic nanoparticles, which have less negative charges than biochar particles. In addition, biochar generally tends to increase the pH of the background solution (Table S1). This effect may further reduce the retention efficiency of soil or sediments upon amendment with biochar.

Biochar application has received wide research attention, especially in agriculture and environmental fields. This study showed that the application of biochar to sediments could enhance the transport of viruses and nanoparticles. This may increase the risk of pathogen contamination in nearby drinking water wells. This study provides an important insight in the retention processes of microbes in sediments upon biochar amendment and the potential impact of biochar in facilitating microbial transport in the subsurface environment.

Funding sources

Funding for this research was provided by the National Centre for Groundwater Research and Training, an Australian Government initiative, supported by the Australian Research Council and the National Water Commission, and by CSIRO Land and Water. The work was conducted in the CSIRO Land and Water Laboratory on the Waite Campus, Adelaide, South Australia.

Acknowledgment

We like to thank Dr. Simon Toze, Dr. Jatinder Sidhu, Ms. Leonie Hodgers (CSIRO, Brisbane) and Prof Howard Fallowfield (Flinders University, Adelaide) for providing guidance in bacteriophage analysis. We like to thank Mr. Harold Rankine for assisting in the laboratory works and Mr. Toney Hirnyk for helping in laboratory set up. We also like to thank Ms. Martin Sheridan (CSIRO, Adelaide), Dr. Luke Mosely (University of Adelaide), Mr. Naser Khan (University of South Australia) for supplying the Biochar samples for us. We also thank the internal (Dr Mike Williams and Dr. Lynne Macdonald) and external reviewers who have contributed ideas and suggestions to improve this manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2015.12.126.

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