Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2019.24418

163 (2019) 270–280
September

Illite for the removal of *E. coli* by filtration in water treatment processes: a comparative study

Mun-Ju Kim[a], Chang-Gu Lee[b], Chang-Hee Lee[c], Seong-Jik Park[a,*]

[a]*Department of Bioresources and Rural System Engineering, Hankyong National University, Anseong, South Korea, Tel. +82-31-670-5131; Fax: +82-31-670-5139; email: parkseongjik@hknu.ac.kr (S.-J. Park)*
[b]*Department of Environmental and Safety Engineering, Ajou University, Suwon, South Korea, email: changgu@ajou.ac.kr*
[c]*Department of Horticultural Life Science, Hankyong National University, Anseong, South Korea, email: changheelee@hknu.ac.kr*

Received 28 January 2019; Accepted 14 May 2019

ABSTRACT

The use of illite for the removal of microorganisms from water has not been studied by other researchers. We assessed the applicability of illite for bacterial removal from water and compared it with commercially available filter media. Column experiments were performed to evaluate the bacterial removal efficiency of filter media under different pH conditions and with chlorine-free tap water. The removal percentage of illite was higher than 90% and its sticking efficiency was greater than 0.85 under all experimental conditions. The removal percentage and sticking efficiency of illite were far superior to that of the other filter media. The high bacterial removal efficiency of illite may be a result of bacterial adhesion enhancement by the Al and Fe eluted from illite. A response surface methodology was employed to optimize experimental conditions including pH, filter depth, and flow rate and to elucidate the effects and interactions of these variables. All these parameters significantly influenced the bacterial removal of illite. From the optimization study, the highest removal percentage of 99.9% was obtained under conditions of a pH of 4.02, a filter depth of 18.49 cm, and a flow rate of 0.86 mL/min. Illite is a potential filter medium for purifying water and preventing groundwater pollution from septic tanks and carcass disposal.

Keywords: Illite; Bacterial removal; Column experiment; Filter media; Optimization; Response surface methodology

1. Introduction

Waterborne diseases caused by the consumption of contaminants can result in serious public health problems [1]. Governments in many countries have attempted to provide improved drinking water sources to their people. However, 663 million people in the world still cannot access improved drinking water [2]. More people have gained access to an improved sanitation facility than to improved drinking water before. 2.4 billion people cannot still access improved sanitation facilities [3].

Microbes are a prime concern in water quality as they are responsible for two-thirds of waterborne disease outbreaks [4]. Filtration through porous media, typically sand, is the most commonly used mechanism for removal of microorganisms from water [5]. During recent years, research on bacterial transport in porous media has attracted less attention from researchers compared with that of membrane filtration. However, filtration following coagulation and sedimentation is still a widely accepted standard method for water treatment throughout the world. The point of use household water-treatment-based filtration has been considered a low-cost and effective solution to provide drinking water to low-income populations [5]. Research on bacterial transport in porous media is required to protect surface water and groundwater supplies from contamination and

* Corresponding author.

to assess the risk from microorganisms in groundwater [6]. Percolation through a natural soil profile or separate filters containing natural or engineered porous media are the most frequently used methods in decentralized systems for wastewater treatment [7]. Treatment wetlands with reactive filter media are considered an alternative to conventional intensive technical treatment of wastewater [8].

Research has been conducted on various factors influencing the transport of microorganisms in porous media: (1) physical factors, such as grain size, surface charge, and surface roughness of porous media, pore water velocity, and water content; (2) chemical factors including pH, ionic strength, competitive anions, and organic matter; and (3) biological factors such as cell type and motility, hydrophobicity, and growth phase [6,9]. Among these parameters, the physical and chemical characteristics of porous media can significantly influence bacterial transport more than other factors. Many studies have investigated the filter media effectiveness in the removal of bacteria. Straining and adsorption are the two main mechanisms responsible for immobilization of pathogenic bacteria via porous media [7]. Sand, which is commonly used in the filtration process, is negatively charged, which is unfavorable for negatively charged bacteria to adsorb on its surface because of electrostatic repulsion between the bacteria and sand surface [9]. Coating of iron and aluminum oxides on the media surface has been suggested by some researchers to improve the interaction between bacteria and media [5,10]. Some clay minerals such as montmorillonite, kaolinite, and goethite have also been investigated for bacterial removal [11,12]. In recent years, biochar-amended porous media have been evaluated for the removal of various types of bacteria by performing column experiments [13–15].

To the best of our knowledge, illite has never been used as a filter media for bacterial removal. llite is a typical 2:1 cationic-layered silicate that includes two silicon oxygen tetrahedra and an alumina oxygen octahedral. Hydrated potassium ions trapped between silicate layers prevent clay swelling [16]. The global abundance of illite is large [17], and its cost ranges from 180 to 420 US$/ton. The abundance of illite and its low cost are likely to make it a strong candidate as a filter medium for the removal of bacteria from aqueous solution. Some of the studies [17–20] have found that illite can be applied to remove the heavy metals and phosphate from aqueous solution.

Illite was compared with other commercially used filter media. Activated carbon is available for the removal of a broad spectrum of organic pollutants via adsorption because of its high specific area and it is thus widely used for drinking water treatment [21]. Anthracite is also used for drinking water treatment as a dual-filter medium with sand by placing it above sand because of its low specific gravity [22]. Birm contains MnO_2 film on its surface and has been developed for the removal of soluble iron and manganese in water [23]. Feroxer is also artificially synthesized for the removal of iron and manganese from water. In contrast to anthracite, garnet has a high hardness and specific gravity, and it is used as the lower strata in a dual-media-filter bed [22]. Hydro-filt, or pumice, is a volcanic rock with a spongy and vitreous structure and a high internal porosity because of the expansion of magmatic gases during its generation [24,25]. It is used particularly for nickel removal because of the ionic exchange with alkaline and alkaline-earth metals present in its structure [24].

The aim of this study was to investigate the feasibility of the use of illite for the bacterial removal and compare its performance with other commercially available filter media. The performance of illite in bacterial removal was compared with commercial filter media under different solution pH values and tap water. Illite, the most efficient filter media, was used to investigate the influence of pH, filter depth, and flow rate on bacterial removal and to optimize these variables. As an alternative to single-variable-at-a-time, which is a time-consuming method, we adopted a response surface methodology (RSM) to explore the relationships among several variables [26]. A Box–Behnken design was employed to optimize the experiments and elucidate the effects and interactions of pH, filter depth, and flow rate.

2. Materials and methods

2.1. Bacterial inoculum

Escherichia coli strain ATCC 11105 was grown in Lysogeny broth (tryptone 10 g/L, yeast extract 5 g/L, NaCl 5 g/L) for 48 h at 30°C. The cultured bacterial suspension was centrifuged at 3,500 rpm for 20 min in a centrifuge (Combi 514R, Hanil, Korea). Three centrifugations were repeated three times to remove nutrient media. The bacteria were suspended in deionized water or chlorine-free tap water and its concentration was adjusted to 0.5 observed density at a wavelength of 600 nm (OD_{600}) using a spectrophotometer (Optizen POP QX, Mecasys, Korea). All the instruments used during the experiment were sterilized at 17.6 psi and 121°C for 15 min to prevent contamination by microorganisms (HB-506, Han Baek Scientific Co., Korea).

2.2. Porous media

For this experiment, all filter media were purchased from a local Korean company. Illite was supplied by Medexx Co. Ltd., and sand from Joomoonjin sand. Anthracite, birm, feroxer, garnet, and hydro-filt were obtained from Baek-Seok Chemical Co. and activated carbon from the Gaya Activated Carbon Co. To remove impurities on each material, each was washed three times with deionized water and then dried at 105°C for 24 h before use.

Physical and chemical characteristics of all filter media were analyzed. Surface morphologies of the filter media were observed using a scanning electron microscope (SEM; S-3500N, Hitachi, Japan). Pore volume, pore size, and specific surface area were measured using a surface area analyzer (Quandrasorb SI, Quantachrome Instrument, USA). From the N_2 adsorption–desorption isotherms, the specific surface area was determined via Brunauer–Emmett–Teller (BET) analysis. The zeta potential was measured using a transition electrophoretic scattering photometer (ELSZ-1000, Otsuka Electronics Co., Japan) to examine the surface charge of the filter media. To determine the pH of each filter medium, it was mixed with deionized water at a ratio of 1:5, and the mixture was stirred for 1 h and then measured using a pH meter (Seven-Multi S40, Mettler Toledo, Switzerland).

X-ray fluorescence (XRF; S8 Tiger 4 K, Bruker, Germany) was used to identify the chemical composition. Elution experiments were performed to investigate the mechanism of bacterial removal using the different filter media. The results were obtained by measuring the cation concentration in the solution after 1 g of each adsorbent was reacted with 30 mL of deionized water. The cation concentrations in the extracted solution were determined using an inductively coupled plasma optical emission spectrometer (ICP-OES; Optima 8300, PerkinElmer, USA).

2.3. Column experiments

Column experiments were performed in duplicate using a polyethylene column (diameter: 16 mm; height: 150 mm) packed with filter media as shown in Fig. 1. Each material was washed three times with distilled water and dried at 105°C for 24 h. Sterile gauze was placed on the top and bottom of the column to prevent it from being dispersed and lost by flowing water. Before bacterial injection into the column, the solution without bacteria was injected using a high-performance liquid chromatography pump (Stepdos, KNF Flodos, Lucerne, Switzerland) to remove the impurities on the filter media and establish a steady flow. The concentration of all injected bacteria was adjusted to 0.5 OD_{600} and the bacterial solution was injected downward to the top of the column at a flow rate of 0.7 mL/min, which corresponded to a filtration rate of 5.0 m^3/m^2 d and a retention time of 21.1 min. The column experiments were performed at different solution pH values (4, 6, and 8) prepared from deionized water and using chlorine-free tap water. To adjust pH to 4, 6, and 8, 1 M HCl and 1 M NaOH were added to the deionized water in which the bacteria were suspended. To remove chlorine from the tap water, 0.05% Na_2SO_3 was added to the tap water according to the method suggested by Niemczewski [27]. The effluent was collected at regular intervals using a fraction collector (Retriever 500, Teledyne, Los Angeles, CA, USA). A breakthrough curve was obtained by monitoring the bacterial concentration in the effluent using an ultraviolet spectrophotometer (Optizen POP QX, Mecasys, Korea).

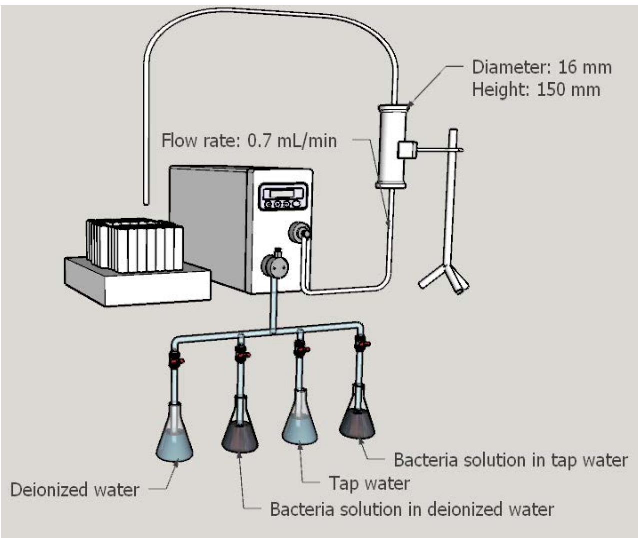

Fig. 1. Schematic diagram of column experiment.

2.4. Data analysis

The bacterial removal percentage (Re) in the effluent was quantified as follows [28]:

$$\mathrm{Re}(\%) = \left(1 - \frac{\int_0^\infty C\,dt}{C_0 t_0}\right) \times 100 \quad (1)$$

where C is the bacterial concentration in the effluent, C_0 is the initial bacterial concentration, and t_0 is the duration of bacterial injection (injection time). In colloid filtration theory, the removal of colloids from suspension via attachment to a collector can be described by the collector efficiency (C_e), which denotes the probability that a colloidal particle approaching a collector both collides with and sticks to it, as follows [29]:

$$C_e = \eta\alpha \quad (2)$$

where η is the collision efficiency, which is the probability that a colloidal particle approaching a collector collides with it, and α is the sticking efficiency, which is the probability that the particle colliding with the collector sticks to it. Collision efficiency (η) is calculated using the following equation from Tufenkji and Elemelech [30]:

$$\eta = 2.4 A_S^{1/3} N_R^{-0.081} N_{\mathrm{Pe}}^{-0.715} N_{\mathrm{vdW}}^{0.052} + 0.55 A_S N_R^{1.675} N_A^{0.125} + 0.22 N_R^{-0.24} N_G^{1.11} N_{\mathrm{vdW}}^{0.053} \quad (3)$$

where A_S is the porosity-dependent parameter, N_R is the aspect ratio, N_{Pe} is the Peclet number, N_{vdW} is the van der Waals number, N_A is the attraction number, and N_G is the gravity number. Sticking efficiency (α) is determined as follows [30]:

$$\alpha = -\frac{2}{3}\frac{d_c}{(1-n)L\eta}\ln\left(\frac{\mathrm{Mr}}{100}\right) \quad (4)$$

where d_c is the particle diameter of the collector grain, L is the filter depth, and n is the porosity. The parameters used in the calculation are η and α. Mr is the bacterial mass recovery in the effluent, which can be quantified by the following equation [28]:

$$\mathrm{Mr} = \left(\frac{\int_0^\infty C\,dt}{C_0 t_0}\right) \quad (5)$$

Statistical analysis was performed using one-way analysis of variance (ANOVA) or two-way ANOVA with repeated measures for group difference. Values of $p < 0.05$ were considered statistically significant. All statistical analyses were performed using SAS version 9.4.

2.5. Experimental design

A Box–Behnken model in the RSM study was used to optimize the experimental conditions for the bacterial removal using illite and investigate the interaction of these parameters on bacterial removal. Three independent variables, that is, solution pH, filter depth, and flow rate, were selected. Table 1 presents the actual levels and corresponding codes of the process variables. Overall, 17 sets of treatment combinations were analyzed using Design-Expert statistical software (version 7.0.0, STAT-EASE Inc., Minneapolis, Minnesota, USA). Second-order polynomials were employed to fit the experimental data obtained. Model adequacy was tested using the sequential F-test, lack-of-fit test, and other adequacy measurements. A quadratic model was generated from the data according to the following equation:

$$Y = a_0 + a_1X_1 + a_2X_2 + a_3X_3 + a_{12}X_1X_2 + a_{13}X_1X_3 + a_{23}X_2X_3 + a_{11}X_1^2 + a_{22}X_2^2 + a_{33}X_3^2 \quad (6)$$

where Y is the predicted response, that is, Re, the removal percentage; X_i represents the variables; and a_i represents the model coefficient parameters. Subscripts 1, 2, and 3 refer to pH, filter depth, and flow rate, respectively. The highest-order polynomial model that was not aliased and had a significant additional term was recommended by the statistical software. The relationship between the three independent variables and two responses was analyzed on the basis of the recommended model.

3. Results and discussion

3.1. Characterization of materials

The surface morphologies of the filter media obtained from SEM analysis are shown in Fig. 2. The surface morphology of the activated carbon is characterized by the aggregation of small particles with many pores (Fig. 2b). Birm can be seen as a cluster of small particles (Fig. 2d) but garnet has a flat structure with little elevation (Fig. 2f). Rod-like pillars were present on the surface of the feroxer (Fig. 2e) and smooth concave holes were observed on the surface of the hydro-filt (Fig. 2g). Sand has a crushed and rough surface (Fig. 2h). Illite (Fig. 2a) and anthracite (Fig. 2c) do not have distinct structure compared with other filter media.

Physical and chemical characteristics of illite are shown in Table 2. The illite used in this study has the smallest particle size and sand the next smallest. The pore volume of activated carbon and anthracite was greater than 0.1 cm^3/g, and that of activated carbon was the highest among the filter media used in this study. Garnet had the largest pore size but that of activated carbon was the smallest. Surface area was more strongly related to pore volume than particle size. The surface area of the activated carbon and anthracite

Table 1
Experimental design levels of the chosen variables

Variables	Symbol	Coded factor level		
		–1	0	1
pH	X_1	4	6	8
Filter depth (cm)	X_2	10	15	20
Flow rate (mL/min)	X_3	0.5	0.7	0.9

with a large pore volume were much larger than that of the other filter media. The zeta potential of all filter media was a negative value, indicating that the surface charge of all filter media was negative. The preparation of samples for the analysis of zeta potential could be among the reasons for the negative values of all filter media. The samples should be crushed and not be settled in quartz cell during measurement. Particle crushing results in clay minerals being negatively charged because the negative charge of the oxygen atoms at the broken edge of the tetrahedral and octahedral sheets are not shared by Si, Al, Fe, and Mg in the tetrahedral and octahedral sheets [31].

The pH of the filter media was in the following order: feroxer > birm > activated carbon > sand > anthracite > hydro-filt > garnet > illite. The low pH of illite and garnet is due to the higher amounts of aluminum dissolution, which leads to a decrease in pH [31]. XRF results show that Si is among the most abundant elements of all filter media except activated carbon, which is mainly (>90%) composed of carbon [32]. Al is an element also found in large quantities in all filter media again except for activated carbon. Garnet and birm have high contents of Fe and Mn, respectively, and are distinct from other filter media in this regard. The results of the elution experiment were significantly different from those obtained from the XRF analysis. The amount of Al in hydro-filt was higher than that of the other filter media, and the concentration of Al eluted from hydro-filt was lower than that of illite and garnet. A high amount of Fe was eluted from the activated carbon even with the small Fe amount in its composition. This finding indicates that the amount of metals eluted from a material is more dependent on the form of the metal in the materials rather than the amount of metal content.

The cost of illite was compared with that of other filter media. Illite is three times more expensive than sand but cheaper than other filter media except sand. Illite is also cheaper than anthracite, which is most commonly used in rapid sand filtration. The low cost of illite is a strong advantage as filter media for the removal of bacteria in water purification process.

3.2. Column experiment for bacterial removal

Two sets of column experiments were performed to compare the efficiency of different filter media in bacterial removal. The first sets of experiments were performed under different pH values, that is, a pH of 4, 6, and 8, and the bacterial removal efficiencies of the filter media were evaluated using chlorine-free tap water in the second sets of experiments. As shown in Table S1, a statistical analysis (two-way ANOVA with repeated measures) showed a significant difference between the groups under different pH and filter media, indicating that the removal percentage and sticking efficiency of the filter media were different depending on the pH value. The experimental results under different solution pH values are shown in Fig. 3; the bacterial removal efficiency in removal percentage and sticking efficiency are shown in Figs. 3a and b, respectively. Both the removal percentage and sticking efficiency decreased as the solution pH increased in all filter media. An observation that bacteria adsorbed on the surface of porous media

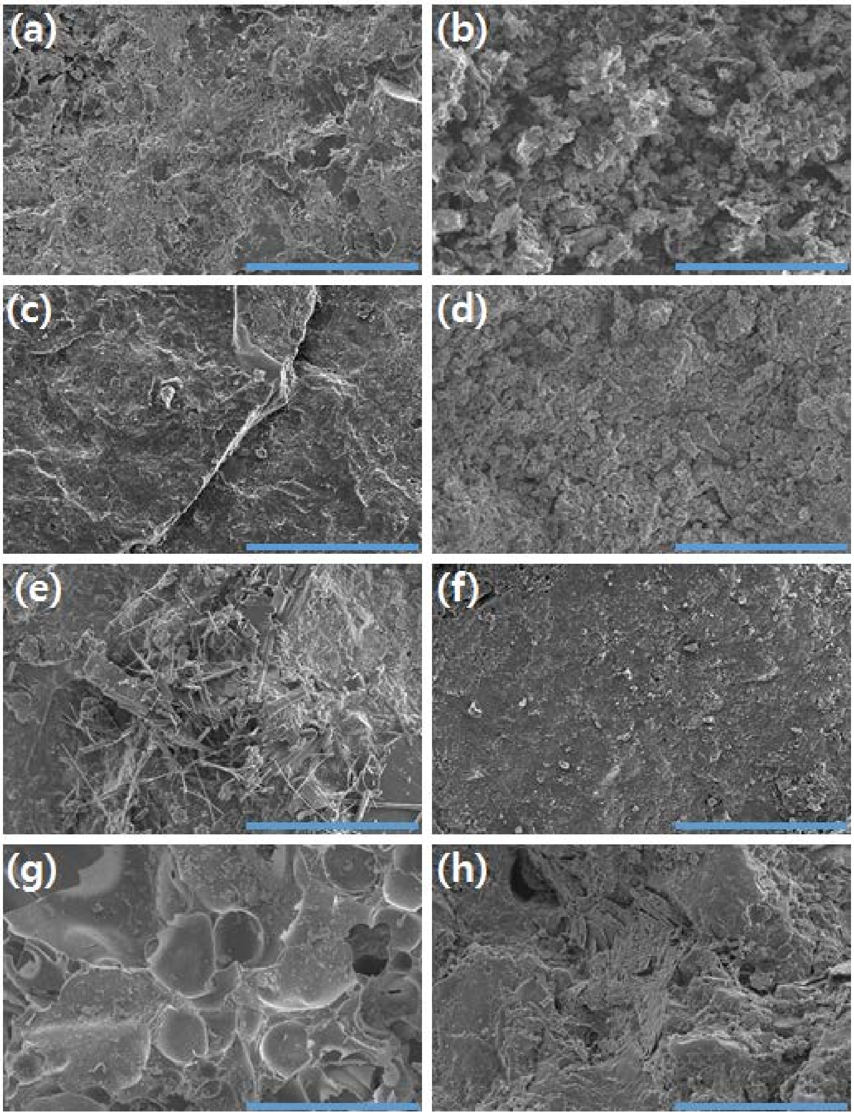

Fig. 2. SEM images: (a) illite, (b) activated carbon, (c) anthracite, (d) birm, (e) feroxer, (f) garnet, (g) hydro-filt, and (h) sand (bar scale = 5 μm).

more readily at lower pH values has also been reported in previous research. Kim et al. [33] reported that bacterial attachment to goethite-coated sand decreased as the solution pH increased. The interaction between viral particles and single-walled carbon nanotubes is repulsive at a higher pH compared with the virus isoelectric point [34]. Bacterial removal rates also decreased with increasing pH in goethite-coated sand [33], iron-coated quartz [35], and diatomite [36]. These observations can be explained by the following: in the cell wall of bacteria, various functional groups are distributed, mainly carboxyl groups (COO^-), phosphoric acid groups (PO_4^{3-}), hydroxyl groups (OH^-), etc. [37,38]. Because of these functional groups on the bacterial cell wall, the surface is negatively charged. As the pH increases, the porous media surface become more negatively charged because of the dissociation of H^+ ions from the tetrahedral or octahedral oxygen on the surface of the filter media [31]. Therefore, as the pH increases, the electrostatic repulsion between more negatively charged filter media and bacteria leads to a decrease in bacterial adhesion to the surface of the filter media.

At a pH of 4, the bacterial removal percentage of the filter media follows a decreasing order as follows: illite > anthracite > feroxer > sand > garnet > activated carbon > birm > hydro-filt. At a pH of 6 or 8, illite also has the highest bacterial removal percentage and that of hydro-filt was also the lowest. Sand was shown to have a high bacterial removal percentage, following illite, at a pH of 6 or 8. Although the sticking efficiency orders of filter media were different from that of removal percentage, illite also had the highest sticking efficiency under all pH conditions. Garnet had the second highest sticking efficiency under all pH conditions even though its bacterial removal percentage was not high compared with that of the other filter media. Hydro-filt had higher ranking in sticking efficiency than that in removal percentage. Such a discrepancy between removal percentage

Table 2
Physico-chemical properties and cost of filter media used in this study

			Illite	Activated carbon	Anthracite	Birm	Feroxer	Garnet	Hydro-filt	Sand
Physical properties	Particle diameter (mm)		0.25	1.21	0.81	0.63	1.02	1.59	1.59	0.40
	Pore volume (cm^3/g)		0.007	0.540	0.160	0.046	0.002	0.011	0.003	0.004
	Pore size (nm)		8.53	2.10	4.15	22.98	3.18	140.30	5.05	9.00
	Surface area (m^2/g)		3.43	1,023.00	152.00	7.95	2.19	0.32	2.26	1.80
	Zeta potential (mV)		–34.67	–20.28	–16.48	–14.43	–49.24	–20.06	–23.85	–19.98
Chemical properties	pH		6.04	9.58	9.15	10.00	10.78	7.06	8.37	9.20
	Elemental composition (%)	SiO_2	64.5	1.6	23.8	58.3	83.8	29.8	53.8	80.6
		Al_2O_3	16.6	0.6	11.5	11.7	3.9	15.6	20.9	7.7
		Fe_2O_3	4.3	0.1	1.8	2.1	0.4	30.7	2.5	1.4
		CaO	0.1	0.1	1.6	4.9	0.0	1.3	1.2	2.4
		K_2O	4.7	0.0	1.2	3.3	0.5	0.8	6.4	0.0
		Na_2O	0.8	0.0	0.4	2.6	0.8	0.2	8.2	1.5
		MnO	0.1	0.0	0.0	7.9	0.5	0.8	0.3	0.0
		LOI	7.5	97.0	55.8	7.2	9.7	15.8	5.2	6.2
		Others	1.6	0.6	3.9	2.0	0.3	5.0	1.5	0.2
	Element (mg/L)	Al	4.83	0.55	0.69	0.58	0.49	1.18	0.57	0.75
		Ca	0.36	0.73	0.44	0.43	0.33	1.32	0.34	0.52
		Fe	3.96	6.14	2.55	1.46	1.44	0.82	0.15	1.29
		K	1.63	0.27	3.59	0.31	1.44	0.36	1.67	0.80
		Mg	0.34	0.44	0.11	0.47	0.31	0.50	0.62	0.30
		Si	0.59	0.59	0.61	0.17	0.46	0.80	0.57	0.39
Cost (USD/ton)			180–420[a] 530[b]	1,000–1,600[a] 2,460[b]	400–650[a] 660[b]	200–500[a] 3,850[b]	1,100[b]	1,150[b]	5,500[b]	300–355[a] 180[b]

Pore volume, pore size, and specific surface were obtained using BET analysis. Elemental composition was analyzed using XRF, and the amount of metal eluted from filter media was quantified by measuring the concentration of a metal in deionized water reacted with filter media.
[a]Obtained from Alibaba (www.alibaba.com).
[b]Provided by Korean local companies.

and sticking efficiency could be due to the difference in the grain size of the filter media. From the equations of single-collector efficiency [30], sticking efficiency represents the fraction of collisions between suspended particles and collector grains that result in attachment, and the influence of grain size was excluded from this equation.

Statistical analysis (one-way ANOVA) indicated that the difference in both removal percentage and sticking efficiency of the filter media among groups was significant ($p < 0.0001$) (Table S2). Under the condition of tap water as the feed water, the bacterial removal percentage of the filter media in decreasing order was as follows: illite > sand > hydro-filt > birm > anthracite > activated carbon > feroxer > garnet (Fig. 4). Illite also proved to be the best among the filter media in this study under tap water conditions. The sticking efficiencies of illite, sand, and garnet were higher ranking, similar to the results obtained under different pH values. The pH and EC of chlorine-free tap water used in this experiment were 7.87 and 1,135 μS/cm, respectively, and its pH was near 8. Even the pH of the chlorine-free tap water was approximately 8. The removal percentage of filter media when using chlorine free-tap water was higher than that for the pH 8 solution prepared from deionized water. This result could be due to the higher ionic strength of the chlorine-free tap water as compared with that of the pH 8 solution prepared from deionized water, providing more favorable conditions for bacterial adhesion to the filter media.

Illite was found to be the most effective in bacterial removal under all experimental conditions tested. When compared with other filter media, illite has small pore volume and specific surface, which are considered important variables as an adsorbent. A high Al and Fe percentage in a filter media can contribute to bacterial adhesion by providing favorable adsorption sites such as for Al or Fe (hydr) oxide [10,39]. However, illite has a higher removal percentage and sticking efficiency than hydro-filt with a higher Al content and garnet with a higher Fe content. The amount of Al and Fe eluted from illite was much higher than that of the other filter media including hydro-filt and garnet. Free Al and Fe ions eluted from illite could create a bridge between the bacteria and illite, leading to high bacterial removal by illite. The low pH of illite is also consistent with the fact that more Al and Fe were eluted from illite. Even though the net charge of all filter media was negative, the vicinity of

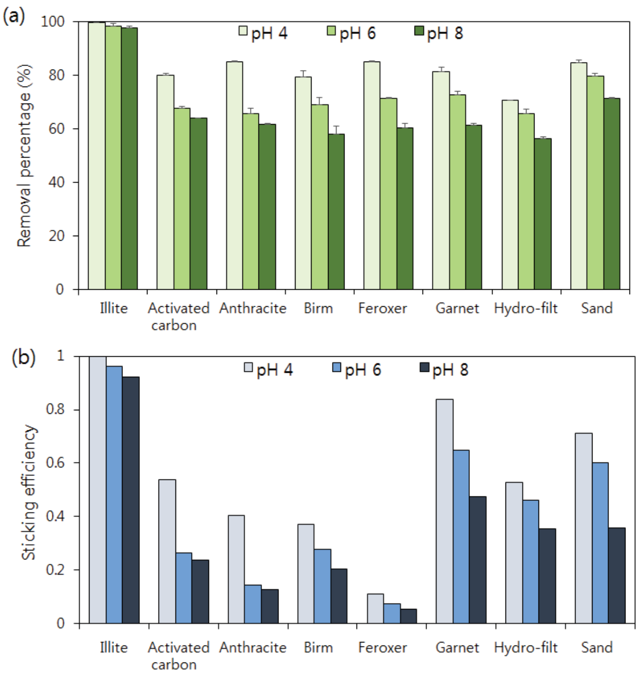

Fig. 3. Comparison of illite with other filter media in bacterial removal under different pH values (4–8): (a) removal percentage (%) and (b) sticking efficiency.

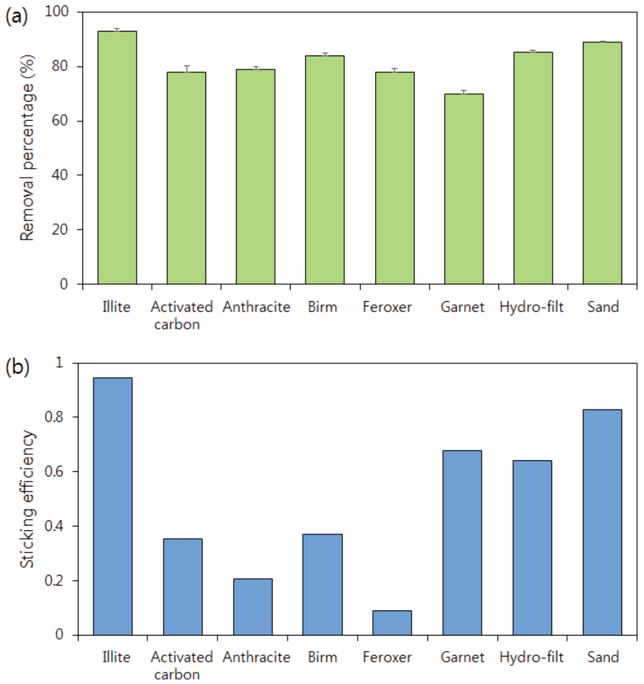

Fig. 4. Comparison of illite with other filter media in bacterial removal under chlorine-free tap water: (a) removal percentage (%) and (b) sticking efficiency.

a heterogeneous surface on illite could be positive because of its low pH [39].

3.3. Optimization studies by statistical experimental design for bacterial removal using illite

A Box–Behnken model of RSM was employed in the experiments to obtain a polynomial model from a 17-experiment design run, including five center points. The ranges and levels of three independent variables were assessed: pH, flow rate, and filter depth. *F*-value tests were performed using ANOVA to calculate the significance of each model employed in the Design-Expert statistical software. The results for Y_{Re} recommended a quadratic model as the highest-order polynomial model that satisfied the criteria such that the additional terms are significant and the model is not aliased. The respective predicted responses were obtained as follows:

$$Y_{Re} = 88.40 - 4.93X_1 + 3.17X_2 - 2.87X_3 + 2.43X_1X_2 - 1.47X_1X_3 + 2.43X_2X_3 + 2.11X_1^2 + 4.25X_2^2 + 2.54X_3^2 \quad (7)$$

The significance of the values of the model equation for removal percentage was checked by F, R^2, adjusted R^2, lack-of-fit, and adequate precision tests. It is noteworthy that we could not obtain the model equation for sticking efficiency using the Box–Behnken model of RSM. This could have been due to the fact that sticking efficiency is not dependent on two variables, that is, flow rate and filter depth, among the three independent variables used in this study. The model *F*-value, which was calculated by dividing the mean squares of each variable effect by the mean square, is shown in Table 3 with the model probability values. That for Re was <0.0001, less than 0.05, indicating that the model terms for all calculated values were significant. The goodness-of-fit for each model was tested using the determination coefficient R^2, and it was 0.992. Its nearness to 1 indicates a good model fit to the observed data. The adjusted R^2 value and the predicted R^2 value were 0.981 and 0.868, respectively, also indicating good fits. A value of (adjusted R^2 − predicted R^2) <0.20 indicates no problem with either the data or the model [40]. The value of adequate precision, which reflects the signal-to-noise ratio, was 27.20, indicating adequate signals with values greater than 4. Lack-of-fit tests used to evaluate the model adequacy obtained significant values for the model, which indicates a poor fit. The statistical results except the lack-of-fit tests showed that the constructed model for removal percentage is suitable for describing the observed data. Fig. 5 shows that the points of the predicted vs. actual plots for removal percentage were clustered along a diagonal line, indicating that the predicted values matched well with those observed.

The interactions of pH, flow rate, and filter depth affecting the removal percentage were plotted as three-dimensional (3D) response surface curves against two experimental factors while maintaining the other factor constant at its central value (Fig. 6). Table 3 also shows the significance of all individual variables. The regression model equation (Eq. (7)) presents a positive or negative influence of the variables on the performance of bacterial removal using illite.

As shown in Table 3, both first-order and second-order effects of all three variables, pH, filter depth, and flow rate, on bacterial removal percentage were significant. The negative sign of the coefficient of X_1 in the regression

Table 3
ANOVA for the RSM analysis of bacterial removal percentage

Source	Sum of squares	*df*	Mean square	*F* value	*p*-value Prob > *F*
Model	530.63	9	58.95902	92.66	<0.0001
X_1	194.39	1	194.38990	305.51	<0.0001
X_2	80.26	1	80.26445	126.14	<0.0001
X_3	65.75	1	65.75177	103.33	<0.0001
X_1X_2	23.60	1	23.59530	37.08	0.0005
X_1X_3	8.585	1	8.58490	13.49	0.0079
X_2X_3	23.69	1	23.69255	37.23	0.0005
X_1^2	18.80	1	18.80347	29.55	0.0010
X_2^2	75.95	1	75.94530	119.35	<0.0001
X_3^2	27.07	1	27.07380	42.55	0.0003
Residual	4.45	7	0.63627	–	–
Lack of fit	4.41	3	1.47021	135.91	0.0002
Pure error	0.04	4	0.01081	–	–
Corrected total	535.09	16	–	–	–

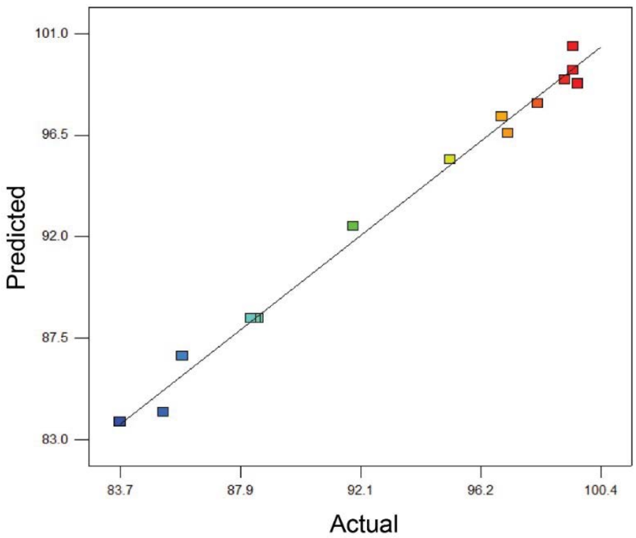

Fig. 5. Plot of predicted values vs. actual values for bacterial removal percentage (%).

model equation indicates that bacterial removal using illite decreased with an increase in pH from 4 to 8. The quadratic effect of pH (X_1^2) had significant and positive influences on bacterial removal percentage, indicating that an increase in the solution pH does not produce a constant decrease in bacterial removal. As shown in Fig. 6, the curvature effect was present at high values of pH, in which case the influence of pH on bacteria was no longer significant. The negative influence of pH on bacterial attachment to illite was previously discussed. Similar to pH, the sign of the X_3 in the regression model equation was also negative and that of X_3^2 was positive. These results indicate that the increase in flow rate negatively influenced bacterial removal but its influence decreased at a higher flow rate. This phenomenon could be explained by the increase in shear force near the surface of the porous media at high flow rates and the decrease in bacterial contact time in the column [41]. Higher hydrodynamic forces working on the surface of filter media at a higher flow rate leads to detachment of bacteria previously attached to the surface of the filter media. The decrease in contact time can negatively affect bacterial removal by decreasing the probability of bacteria to colloid with the surface of the filter media. The positive sign of X_2 indicates that the increase in filter depth increased the bacterial removal percentage. In contrast to flow rate, the increase in filter depth increased the probability of bacteria to collide with filter media. As seen in Fig. 6, the nonlinearities of all 3D response surfaces and each contour plot show a significant interaction between the independent variables and bacterial removal percentages. The nonlinear contour plots mean that there is no direct linear relationship between the selected independent variables [42]. Table 3 also shows that the interacting effects of independent variables were all significant.

The optimal values of the variables, which maximize the response, that is, Re, was tracked using the RSM. The highest Re of 99.9% was obtained under the conditions of a pH of 4.02, a filter depth of 18.49 cm, and a flow rate of 0.86 mL/min.

4. Conclusions

The applicability of illite as a filter medium for bacterial removal was assessed by comparing it with other commercially available filter media and optimizing the experimental conditions. Column experiments were performed under different solution pH values, and bacterial adhesion to all filter media decreased as pH increased. Both the bacterial removal percentage and sticking efficiency of illite were higher than that of the other filter media under all experimental conditions in this study. The higher amount of soluble Al and Fe eluted from illite and its lower pH compared with the other filter media produced favorable conditions for bacterial attachment to the surface of illite. A Box–Behnken design based on the RSM was used to explore the relationships

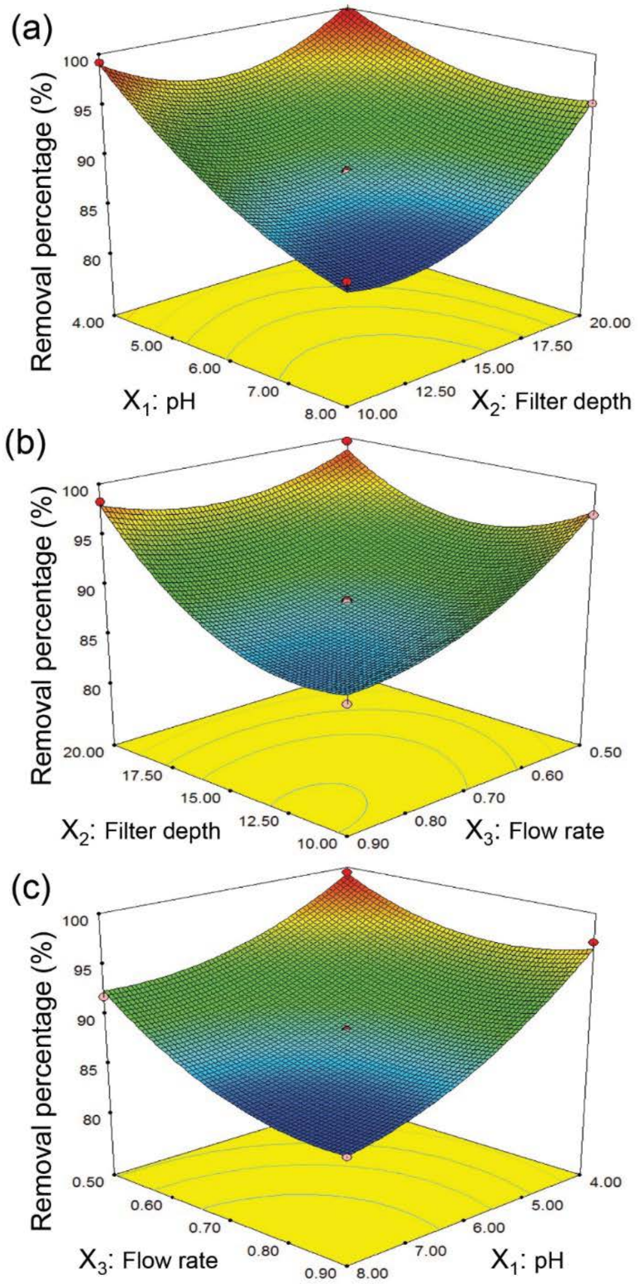

Fig. 6. Estimated response surface for removal percentage showing the effects of (a) pH and filter depth (cm), (b) filter depth (cm) and flow rate (mL/min), and (c) flow rate (mL/min) and pH.

among the three variables, that is, pH, filter depth, and flow rate, on bacterial removal using illite and to obtain an optimum of these variables. All three variables were significant for bacterial removal using illite. The optimal conditions for bacterial removal were as follows: pH of 4.02, 18.49 cm filter depth, and 0.86 mL/min flow rate. Illite proved to be an effective filter medium for water bacterial removal without further modifications. It can also be potentially used for the prevention of groundwater contamination by microorganisms from carcass disposal and septic tanks.

Acknowledgments

This work was supported by Korea Institute of Planning and Evaluation for Technology in Food, Agriculture, Forestry and Fisheries (IPET) through Advanced Production Technology Development Program, funded by Ministry of Agriculture, Food and Rural Affairs(MAFRA)(Grant No. 317017-03). This work was also supported by IPET through Animal Disease Management Technology Development Program, funded by MAFRA (Grant No. 118095-2).

References

[1] J.S. Park, J.S. Kim, S.J. Kim, E.K. Shin, K.-H. Oh, Y.H. Kim, C.H. Kim, M.A. Hwang, C.M. Jin, K.G. Na, J. Lee, E.H. Cho, B.-H. Kang, H.-S. Kwak, W.-K. Seong, J. Kim, A waterborne outbreak of multiple diarrhoeagenic Escherichia coli infections associated with drinking water at a school camp, Int. J. Infect. Dis., 66 (2018) 45–50.
[2] J. Luh, J. Bartram, Drinking water and sanitation: progress in 73 countries in relation to socioeconomic indicators, Bull. World Health Organ., 94 (2015) 111–121A.
[3] K.D. Orner, J.R. Mihelcic, A review of sanitation technologies to achieve multiple sustainable development goals that promote resource recovery, Environ. Sci. Water Res. Technol., 4 (2018) 16–32.
[4] M.F. Craun, G.F. Craun, R.L. Calderon, M.J. Beach, Waterborne outbreaks reported in the United States, J. Water Health, 4 (2006) 19–30.
[5] M.M. Ahammed, V. Meera, Metal oxide/hydroxide-coated dual-media filter for simultaneous removal of bacteria and heavy metals from natural waters, J. Hazard. Mater., 181 (2010) 788–793.
[6] H. Bai, N. Cochet, A. Pauss, E. Lamy, Bacteria cell properties and grain size impact on bacteria transport and deposition in porous media, Colloids Surf., B, 139 (2016) 148–155.
[7] T.K. Stevik, K. Aa, G. Ausland, J.F. Hanssen, Retention and removal of pathogenic bacteria in wastewater percolating through porous media: a review, Water Res., 38 (2004) 1355–1367.
[8] B. Pucher, H. Ruiz, J. Paing, F. Chazarenc, P. Molle, G. Langergraber, Using numerical simulation of a one stage vertical flow wetland to optimize the depth of a zeolite layer, Water Sci. Technol., 75 (2017) 650–658.
[9] S.J. Park, S.B. Kim, Influence of (bi) carbonate on bacterial interaction with quartz and metal oxide-coated surfaces, Colloids Surf., B, 76 (2010) 57–62.
[10] J. Lukasik, Y.-F. Cheng, F. Lu, M. Tamplin, S.R. Farrah, Removal of microorganisms from water by columns containing sand coated with ferric and aluminum hydroxides, Water Res., 33 (1999) 769–777.
[11] D. Jiang, Q. Huang, P. Cai, X. Rong, W. Chen, Adsorption of Pseudomonas putida on clay minerals and iron oxide, Colloids Surf., B, 54 (2007) 217–221.
[12] Z. Hong, W. Chen, X. Rong, P. Cai, W. Tan, Q. Huang, Effects of humic acid on adhesion of Bacillus subtilis to phyllosilicates and goethite, Chem. Geol., 416 (2015) 19–27.
[13] S.M. Abit, C.H. Bolster, P. Cai, S.L. Walker, Influence of feedstock and pyrolysis temperature of biochar amendments on transport of Escherichia coli in saturated and unsaturated soil, Environ. Sci. Technol., 46 (2012) 8097–8105.
[14] S.K. Mohanty, A.B. Boehm, Escherichia coli removal in biochar-augmented biofilter: Effect of infiltration rate, initial bacterial concentration, biochar particle size, and presence of compost, Environ. Sci. Technol., 48 (2014) 11535–11542.
[15] S. Sasidharan, S. Torkzaban, S.A. Bradford, R. Kookana, D. Page, P.G. Cook, Transport and retention of bacteria and viruses in biochar-amended sand, Sci. Total Environ., 548 (2016) 100–109.
[16] N.C. Brady, R. Weil, The Colloidal Fraction: Seat of Soil Chemical and Physical Activity, In: Elements of the Nature and Properties of Soils, 3rd ed., Pearson, London, 2014, Chapter 8.

[17] X. Gu, L.J. Evans, Modelling the adsorption of Cd (II), Cu (II), Ni (II), Pb (II), and Zn (II) onto fithian illite, J. Colloid Interface Sci., 307 (2007) 317–325.
[18] N.G. Turan, S. Elevli, B. Mesci, Adsorption of copper and zinc ions on illite: determination of the optimal conditions by the statistical design of experiments, Appl. Clay Sci., 52 (2011), 392–399.
[19] J. Chen, L.-g. Yan, H.-q. Yu, S. Li, L.-l. Qin, G.-q. Liu, B. Du, Efficient removal of phosphate by facile prepared magnetic diatomite and illite clay from aqueous solution, Chem. Eng. J., 287 (2016) 162–172.
[20] B.W. Gu, S.H. Hong, C.G. Lee, S.J. Park, The feasibility of using bentonite, illite, and zeolite as capping materials to stabilize nutrients and interrupt their release from contaminated lake sediments, Chemosphere, 219 (2019) 217–226.
[21] R.S. Summers, D.R. Knappe, V.L. Snoeyink, Adsorption of Organic Compounds by Activated Carbon, J.K. Edzwald, Ed., Water Quality & Treatment: A Handbook on Drinking Water, 6th ed., McGraw-Hill, New York, 2011.
[22] M.L. Davis, Water and Wastewater Engineering, McGraw-Hill, New York, 2011.
[23] D. Barloková, J. Ilavský, Removal of iron and manganese from water using filtration by natural materials, Pol. J. Environ. Stud., 19 (2010) 1117–1122.
[24] N. Moraci, P.S. Calabrò, Heavy metals removal and hydraulic performance in zero-valent iron/pumice permeable reactive barriers, J. Environ. Manage., 91 (2010) 2336–2341.
[25] A.S. Ruhl, N. Ünal, M. Jekel, Evaluation of two-component Fe(0) fixed bed filters with porous materials for reductive dechlorination, Chem. Eng. J., 209 (2012) 401–406.
[26] T. An, J. An, H. Yang, G. Li, H. Feng, X. Nie, Photocatalytic degradation kinetics and mechanism of antivirus drug-lamivudine in TiO_2 dispersion, J. Hazard. Mater., 197 (2011) 229–236.
[27] B. Niemczewski, Observations of water cavitation intensity under practical ultrasonic cleaning conditions, Ultrason. Sonochem., 14 (2007) 13–18.
[28] S.J. Park, C.G. Lee, S.B. Kim, The role of phosphate in bacterial interaction with iron-coated surfaces, Colloids Surf., B, 68 (2009) 79–82.
[29] T.L. Cail, M.F. Hochella Jr., The effects of solution chemistry on the sticking efficiencies of viable Enterococcus faecalis: an atomic force microscopy and modeling study, Geochim. Cosmochim. Acta, 69 (2005) 2959–2969.
[30] N. Tufenkji, M. Elimelech, Correlation equation for predicting single-collector efficiency in physicochemical filtration in saturated porous media, Environ. Sci. Technol., 38 (2004) 529–536.
[31] N.C. Brady, R.R. Weil, Elements of the Nature and Properties of Soils, No. 631.4 B733E, Upper Saddle River, Pearson Educational International, NJ, 2010.
[32] B.W. Gu, C.G. Lee, T.G. Lee, S.J. Park, Evaluation of sediment capping with activated carbon and nonwoven fabric mat to interrupt nutrient release from lake sediments, Sci. Total Environ., 599 (2017) 413–421.
[33] S.-B. Kim, S.-J. Park, C.-G. Lee, N.-C. Choi, D.-J. Kim, Bacteria transport through goethite-coated sand: effects of solution pH and coated sand content, Colloids Surf., B, 63 (2008) 236–242.
[34] A.S. Brady-Estévez, T.H. Nguyen, L. Gutierrez, M. Elimelech, Impact of solution chemistry on viral removal by a single-walled carbon nanotube filter, Water Res., 44 (2010) 3773–3780.
[35] D.A. Ams, J.B. Fein, H. Dong, P.A. Maurice, Experimental measurements of the adsorption of Bacillus subtilis and Pseudomonas mendocina onto Fe-oxyhydroxide-coated and uncoated quartz grains, Geomicrobiol. J., 21 (2004) 511–519.
[36] J.T. Gannon, V.B. Manilal, M. Alexander, Relationship between cell surface properties and transport of bacteria through soil, J. Appl. Environ. Microbiol., 57 (1991) 190–193.
[37] T.J. Beveridge, R.G. Murray, Sites of metal deposition in the cell wall of Bacillus subtilis, J. Bacteriol., 141 (1980) 876–887.
[38] J.B. Fein, D.A. Fowle, J. Cahill, K. Kenmer, M. Boyanow, B. Bunker, Nonmetabolic reduction of Cr (VI) by bacterial surfaces under nutrient-absent conditions, Geomicrobiol. J., 19 (2002) 369–382.
[39] J.W.A. Foppen, J.F. Schijven, Transport of E. coli in columns of geochemically heterogeneous sediment, Water Res., 39 (2005) 3082–3088.
[40] D.C. Montgomery, Design and Analysis of Experiments, John Wiley & Sons, Singapore, 2004.
[41] S.J. Park, C.G. Lee, S.B. Kim, Y.Y. Chang, J.K. Yang, Bacterial removal in flow-through columns packed with iron-manganese bimetallic oxide-coated sand, J. Environ. Sci. Health., Part A, 47 (2012) 1364–1371.
[42] K. Yetilmezsoy, S. Demirel, R.J. Vanderbei, Response surface modeling of Pb (II) removal from aqueous solution by Pistacia vera L.: Box–Behnken experimental design, J. Hazard. Mater., 171 (2009) 551–562.

Supplementary Information:

Table S1
Two-way ANOVA with replication for (a) removal percentage and (b) sticking efficiency of filter media under different pH values

Removal percentage					
Source of variation	Sum of squares	Degrees of freedom	Mean square	*F* ratio	*p*-value
pH	3,653.2	2	1,826.6	709.3	<0.0001
Filter media	4,438.4	7	634.1	246.2	<0.0001
Interaction	951.6	14	68.0	26.4	<0.0001
Error	61.8	24	2.6		
Total	9,105.0	47			
Sticking efficiency					
Source of variation	Sum of squares	Degrees of freedom	Mean square	F ratio	p-value
pH	0.408	2	0.204	376.8	<0.0001
Filter media	3.195	7	0.456	843.5	<0.0001
Interaction	0.136	14	0.010	17.9	<0.0001
Error	0.013	24	0.001		
Total	3.752	47			

Table S2
One-way ANOVA with replication for (a) removal percentage and (b) sticking efficiency of filter media under chlorine-free tap water

Removal percentage					
Source of variation	Sum of squares	Degrees of freedom	Mean square	*F* ratio	*p*-value
Treatment	760.1	7	108.6	41.3	<0.0001
Error	21.0	8	2.6		
Total	781.1	15			
Sticking efficiency					
Source of variation	Sum of squares	Degrees of freedom	Mean square	*F* ratio	*p*-value
Treatment	1.1685	7	0.1669	355.6	<0.0001
Error	0.0038	8	0.0005		
Total	1.1723	15			