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EVALUATING FILTER MATERIALS FOR E. COLI REMOVAL FROM
STORMWATER

BY
GHAEM HOOSHYARI

A thesis submitted in partial fulfillment of the requirements for the

Master of Science

Major in Civil Engineering

South Dakota State University

2017

Evaluating Filter Materials for E. coli Removal from Stormwater

This thesis is approved as a creditable and independent investigation by a candidate for the Master of Science in Civil Engineering degree and is acceptable for meeting the thesis requirements for this degree. Acceptance of this does not imply that the conclusions reached by the candidate are necessarily the conclusions of the major department.

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I would like to dedicate this thesis to my amazing wife,

and to my parent.

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LIST OF ABBREVIATIONS

Al	Aluminum
ATCC	American type culture collection
°C	Centigrade
C/C ₀	Ratio of outlet E. coli to inlet E. coli
C _e	Equilibrium E. coli concentration
C ₀	Initial E. coli concentration
Ca	Calcium
CaCO ₃	Calcium Carbonate
Cd	Cadmium
Cl ⁻	Chlorine
Cu	Copper
D	Desorption of E. coli
<i>d_e</i>	E. coli Concentration after certain desorption time
DOC	Dissolved organic carbon
EBCT	Empty bed contact time
E. Coli	Escherichia coli
Fe	Iron
g	Gram
ΔG°	Gibb's free energy change
h	Hour
H ₂ SO ₄	Sulfuric acid
K _F	Freundlich constant

K_L	Langmuir constant
k_1	Pseudo first-order equilibrium rate constant
k_2	Pseudo second-order equilibrium rate constant
KCl	Potassium chloride
L	Liter
Lead	Pb
M	Molar
mg	Milligram
Mm	Millimolar
mm	Millimeter
mL	Milliliter
MPN	Most probable number
NaHCO_3^-	Sodium bicarbonate
NaOH	Sodium hydroxide
NOM	Natural organic matter
Ni	Nickle
N_r	Number of the E. coli removed by adsorbents
OH^-	Hydroxide
q_e	Equilibrium E. coli adsorption capacity
q_{\max}	Maximum E. coli adsorption capacity
q_t	Time specific E. coli adsorption capacity
q_{total}	Total mass of E. coli adsorbed
Q	Volumetric flow rate

RO	Reverse osmosis
rpm	Revolution per minute
sd	Standard deviation
SMPs	Stormwater management practices
T	Temperature
Zn	Zinc

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ABSTRACT

EVALUATING FILTER MATERIALS FOR E. COLI REMOVAL FROM
STORMWATER

GHAEM HOOSHYARI

2017

Stormwater runoff from agricultural and urban areas carries a wide range of pollutants and pathogens that can negatively affect surface water bodies and cause significant risks to the ecosystem and public health. Bacteria is one of the pollutants carried by stormwater, and *Escherichia coli* (*E. coli*) is commonly used as a microbial pollution indicator of surface water. The aim of this research was to investigate the removal of *E. coli* from stormwater using low-cost filter materials. Two industrial byproducts (steel slag and steel chips) and two natural minerals (zeolite and limestone) with three different sizes (0.5-1 mm, 1-2 mm, and 2-4 mm) were chosen to investigate the potential of these materials as filter media for *E. coli* removal from stormwater. Batch experiments were conducted to investigate the impact of initial *E. coli* concentration, temperature, pH, particles size and mass, salt, natural organic matter (NOM), and contact time on the removal of *E. coli*. Column adsorption experiments were also performed to obtain the *E. coli* adsorption characteristics of steel chips, steel slag, limestone, and zeolite under continuous flow conditions. In addition, the *E. coli* release potential of these materials were determined. Using a desorption test, the batch adsorption results demonstrated that the maximum *E. coli* removal efficiencies of 100%, 99.5%, 86.5%, and 80.2% were achieved using steel byproducts, steel slag, limestone, and zeolite, respectively, using *E. coli* concentrations of 10^7 MPN/mL for steel chips and 10^4 MPN/mL

for steel slag, limestone, and zeolite, in the synthetic stormwater. Increasing pH from 5 to 9 resulted in a reduction in E. coli adsorption by 33.5% and 19.0% for steel chips and steel slag, respectively. As temperature increased steel chips and steel slag adsorption capacities increased. The effect of the addition of NOM on E. coli removal efficiencies was determined. The results indicated that E. coli removal efficiency were reduced by 19.77% and 41.77% for steel chips and 6.86% and 11.56% for steel slag in the presence of 20 and 50 mg/L of NOM. Moreover, E. coli release from steel chips was negligible in comparison with other absorbents. Finally, adding high salt concentrations in E. coli solution showed a significant impact in natural minerals adsorption capacities. As the salinity increased the E. coli adsorption capacity of limestone and zeolite were improved by 23.5%, and 35.5%, respectively.

1. INTRODUCTION

Stormwater runoff from agricultural and urban areas carries a large amount of pollutants and pathogens, such as heavy metals (e.g. copper (Cu), zinc (Zn), cadmium (Cd), Nickel (Ni), and lead (Pb)), nutrients (phosphate and nitrate), suspended solids (organic substance), and microorganisms (viruses and bacteria). Organic and inorganic pollutants are carried into ground and surface water by stormwater runoff, that can negatively affect natural water bodies. Stormwater runoff also causes significant risks to the ecosystem and public health (House et al., 1993; Hat et al., 2008; Tafuri and Field, 2012).

Bacteria is one of the pollutants carried by stormwater and *Escherichia coli* (*E. coli*) is commonly used as a microbial pollution indicator of surface water (CWP 1999). *E. coli* is found in the food, environment, and intestines of human and animals. A large amount of *E. coli* from animal manures and municipal disposal can enter into natural water after a heavy rainfall.

Some stormwater management practices (SMPs) have been applied to control and treat the pollutants from stormwater. Pitt et al. (2008) reported that the geographical location and land were the principal causes that affected the runoff pollutant concentrations. The removal of runoff contaminants can be achieved by a variety of technologies including infiltration, bio-retention, constructed wetlands, vegetative swales, and other engineered treatment systems (Clark and Pitt, 2012). Although many of the conventional SMPs (e.g. detention ponds) are designed to control runoff volume and remove particles from the runoff, these conventional SMPs are generally not effective in

removing other pollutants such as heavy metals, nutrients, and (*E. coli*). Infiltration-type stormwater treatment systems are able to remove different contaminants but these systems typically require a large operating footprint and have the risk of contaminating the groundwater. There is a need to develop low-maintenance, low-cost, and effective SMPs that can remove multiple contaminants in stormwater runoff.

Media filtration has received increasing attention as an effective technology that can remove microorganisms and other pollutants from stormwater runoff using a relatively small footprint. Many filter materials have been evaluated for their potential for runoff treatment. These materials include sand, modified sand, anthracite coal, modified wool, modified cotton, limestone, clay, zeolite, steel slags, iron filling, woodchips, sawdust, and tire crumbles; all of which possess good hydraulic properties and are widely available (Bailey et al., 1999; Lukasik et al., 1999; Kim et al., 2010; Wium-Anderson et al., 2012; Reddy et al., 2014a). Some studies have also demonstrated that single filter media does not have a great adsorption capacity to remove all the contaminants of concern in stormwater (Dastgheibi 2012; Wium-Anderson et al., 2012; Reddy 2013; Reddy et al., 2014). The combination of two or more media is often necessary to achieve high removal efficiency of multiple contaminants.

Seelsaen et al. (2006) recommended that different sorption media mixes (sand, compost, zeolite etc.) be used as an effective medium for the treatment of dissolved metal contaminants commonly found in stormwater. Prabhukumar (2014) performed column experiments to evaluate the contaminant removal of filter materials. The results showed that calcite was the most efficient material for nutrients and suspended solids removal, natural zeolite was highly effective in removing *E. Coli*, and iron fillings were effective

in removing nutrients and metals. Reddy et al. (2014) showed that mixed-media filtration (calcite, zeolite, sand and iron fillings) was effective for simultaneous removal of nutrients and heavy metals from stormwater runoff. These studies suggested that mixed-media filtration systems using permeable reactive materials have great potential to remove multiple contaminants in stormwater runoff.

There are different methods for *E. coli* removal such as biofiltration (Almaguer et al., 2011; Chandrasena et al., 2014), electro-coagulation (Wong et al., 2010; Delaire et al., 2013), and filtration with modified materials (Jonson and Logan 1995; Li, Y. L et al., 2012; Akhigbe et al., 2016; Shaw et al., 2016).

One of the most important methods for bacteria removal is adsorption of bacteria onto the surface of filter media and this mechanism is referred to as surface attachment. Many factors affect bacteria attachment onto porous media. In order to describe the bacterial sorption, adsorption isotherms have been developed for illustrating mass transfer between solid and liquid phases. The Freundlich isotherm, the Langmuir isotherm, and the Linear isotherm are three types of isotherms used to illustrate sorption process (Powelson and Mills, 2001; Mankin et al., 2007).

Moreover, stormwater runoff carries different types of organic compounds that can deteriorate surface water quality. Several studies have shown that natural organic matter (NOM) has some effect on bacterial cell walls, their electrophoretic mobility, and their attachment to minerals (Gerritson and Bradly, 1987). Bacteria and dissolved organic carbon (DOC) have negative charges on their surface and they can be attached to the positive charge of media surface. This mechanism results in competition between bacteria and DOC during filtration (Scholl and Harvey, 1992). Johnson et al. (1996) reported that

Suwannee river NOM could influence bacteria and filter media (quartz), thereby increasing the negative charge on both bacteria and quartz surfaces, which results in repulsive electrostatic interaction between bacteria and quartz leading to a decrease in bacteria removal. Jonson and Logan (1995) stated that bacteria removal decreased with the presence of sediment organic matter (SOM). The authors used Fe-quartz as filtration media in their study. It was shown that SOM could be attached to the positive charge of the media surface and establish a negatively charged media surface. This mechanism leads to an electrostatic repulsion between porous media and bacteria. However, bacteria removal with Fe-quartz without any competitors increased because iron-oxide raises the positive charge of the media surface.

Other factors that affect *E. coli* removal through filter media are salinity and pH. As the salinity increased, ionic strength increased, which led to enhanced attachment of negatively charged bacteria onto silica beads, quartz, and sand (Fonts et al., 1991; Mills et al., 1994). Guber et al. (2005) reported that soil and *E. coli* both have negatively charged surfaces, which make unfavorable environments in *E. coli* removal by soil. The ionic strength can be increased to reduce the electrical double layer on both media and *E. coli* cells surface, thereby allowing the *E. coli* cells to approach the media surface and Van der Waals force as an attractive power exceeds electrostatic repulsion.

Lytle et al. (1999) measured the electrophoretic mobilities (EPMs) of several *E. coli* types in different pH values (2, 5, and 6.5). The results indicated that EPM was increased with decreasing pH. Several studies also illustrated that the adsorption of bacteria onto porous media is strongly pH-dependent (Scholl and Harvey, 1992; Guber et al., 2005; Starosvetsky et al., 2012).

The aim of this research was to investigate the adsorption capacities of low-cost materials for *E. coli* removal in synthetic stormwater. Batch experiments were conducted to assess the impact of initial *E. coli* concentration, temperature, pH, particles size and mass, contact time, salt, and NOM on the *E. coli* removal. Two natural minerals (zeolite and limestone) and two industrial byproducts (steel slag and steel chips) with three different sizes (0.5-1 mm, 1-2 mm, and 2-4 mm) were chosen to investigate the potential of these materials as filter media for *E. coli* removal from stormwater.

2. MATERIAL AND METHODS

2.1 Filter materials

Four materials were selected for this study; carbon steel byproducts (steel chips), electric arc furnace steel slag, limestone, and zeolite (Fig. 2.1). Table 2.1 summarizes the characteristics of filter materials. Limestone as a sedimentary stone composed of varying crystal forms of calcium carbonate (CaCO_3) (Aziz et al., 2001) was acquired from Martin Marita Limestone Co, Inc. Natural zeolites are formed in basaltic lava and particular rocks exposed to moderate geologic temperature and pressure (D.E.W 1988). The natural zeolite from Bear River Zeolite Co, Inc. was used for this study.

Steel chips are produced by cutting, shaping, drilling, and finishing carbon steel products and were obtained from a metal machining factory located in Sioux Falls, South Dakota. Electric arc furnace steel slag is a byproduct of steelmaking and typically produced through the blast furnace. Slag is usually a mixture of metal oxides and silicon dioxide. The steel slag was collected from Nucor Steel (Norfolk, Nebraska).

All filter materials were sieved with different size sieves, then washed with distilled water water to remove any dust or very fine particles that can cause interference in adsorption experiments. Steel chips were washed with phosphate free soap to remove any possible oil on the surface. Then materials were air-dried. During the drying process, the steel chips were oxidized, forming a layer of rust on the surface. For comparison purposes, all materials were sieved to a size range of 0.5-1 mm, 1-2 mm. and 2-4 mm (Fig. 2.2), to identify the impact of material sizes on *E. coli* removal. Particle densities were determined for four materials as the amount of water volume displaced by a certain mass of material. Steel slag, limestone, and zeolite had lower particle densities between

2.18-3.57 g/cm³, while steel chips had higher particle densities between 5.20-5.91 g/cm³. PH values were measured using ASTM D4972. The steel slag and natural minerals were alkaline (pH > 8.0) while the steel chips pH was 6.3.

2.2 E. coli preparation

An Escherichia coli (E. coli) ATCC 35218 (American Type Culture Collection) as a nonpathogen and Gram-negative bacterium was used as bacteria culture in this study. E. coli was incubated in a 100 mL Luria Broth Base (Thermo Fisher Scientific, included 10 g/L of peptone, 10 g/L of sodium chloride, 5 g/L of yeast extract) at 37 °C, in a temperature controlled shaker. The culture was shaken at 150 rpm for 24 h, then the cells were centrifuged to remove growth media, and washed with buffer solution three times before re-suspending in buffer solution. The buffer solution was prepared by dissolving 1.0 mole NaHCO₃ and 0.01mole KCl in one liter distilled water. The pH value of buffer solution was adjusted to 7 using 1.0 mole H₂SO₄ solution. A spectrophotometer (Model DR400, Hach, Germany) was used for measuring E. coli cell concentration. 1.5×10^9 cells/mL was the initial E. coli cell concentration in buffer solution. Serial dilution (10-fold) were used to bring the E. coli concentration to the level of 1×10^4 MPN/mL (Most Probably Number) as an initial E. coli concentration for batch experiments. IDEXX Quanti-Tray 2000 method (i.e., most probably number, MPN) was used for accurate determination of the level of E. coli in solution. All dishes were placed in phosphate-free soap baths overnight then rinsed with reverse osmosis (RO) water. Dishes were then placed in 25% H₂SO₄ (acid bath) overnight, afterward rinsed with 18MΩ nanopure water, and autoclaved prior to use. For avoiding any contamination, before and after each

experiment all apparatus were sterilized with Steris Coverage Spray HB plus (disinfectant cleaner).

2.3 Batch Adsorption for E. coli removal

Batch adsorption experiments were conducted to evaluate the E. coli removal efficiency for each filter material at various initial E. coli concentrations, temperature, pH, particles size and mass, and contact time. The impact of salt and NOM on E. coli adsorption onto filter materials was also assessed. The E. coli removal efficiency depends on the mass of materials, time, pH, and temperature. Initial E. coli concentrations of 10^7 MPN/mL and 10^4 MPN/mL were chosen for steel chips and steel slag, limestone, and zeolite, respectively, for adsorption isotherm, kinetics, and desorption tests. An initial E. coli concentration of 10^4 MPN/mL was used to evaluate the impact of pH, temperature, salt, NOM, and continuous flow on adsorption. Although 24 hours was found to be adequate to achieve equilibrium conditions in adsorption tests, 2 hours was used for determining the impact of pH, temperature, salt, NOM, and different initial E. coli concentrations based on kinetic results. A temperature controlled orbital shaker (Model MaxQ 4000, Thermo Scientific, Waltham, MA) was used for all batch experiments (Fig. 2.3).

2.4 Adsorption Isotherms

For the adsorption isotherm tests, 0.1, 0.2, 0.5, 1, 2, 4, and 8 g (steel slag and steel chips), and 0.5, 1, 2, 4, 8, and 16 g (limestone and zeolite) with the three different size ranges (0.5-1, 1-2, and 2-4 mm) were used. Each material was placed in a 250 mL Erlenmeyer flask containing 100 mL buffer solution with an initial E. coli concentration of 10^4 MPN/mL for steel slag, limestone, and zeolite, and 10^7 MPN/mL for steel chips. A

flask with 100 mL E. coli solution without any absorbent was prepared as a control. Flasks were placed in a shaker and were continuously shaken at 100 rpm for 24 h at 20°C. After 24 h, flasks were removed from the shaker and allowed to stand for 30 min, then the solution was analyzed for E. coli concentration via IDEXX method. The following steps were included in analyzing the solution with the IDEXX Quanti-Tray/2000. First, samples were diluted (10-fold) to below 2400 MPN/100 mL (this is the highest number that the incubation tray can measure), and placed into a 100 mL IDEXX vessel containing a dechlorination chemical (sodium thiosulfate to neutralize up to 15 mg/L of chlorine). Second, one packet of Colilert reagent was added and mixed to the sample by gently inverting the bottle. After that, the solution was poured into the incubation tray and sealed by a Quanti-Tray sealer (Cat WQTs2X-115). Third, the sealed tray was placed in a 45°C incubator for 24h then the results were analyzed by counting large and small positive wells and checking with fluorescence light under a long-wave ultraviolet light which is known as an E. coli indicator (Fig. 2.4). All experiments were conducted in duplicate to ensure the accuracy of the experiments. The results of the duplicate experiments are expressed as average values and standard deviations.

The equilibrium adsorption capacity, q_e (mg/g), was calculated by the following equation:

$$q_e = \frac{V(C_0 - C_e)}{m} \quad (\text{Eq. 2.1})$$

where C_0 and C_e are the initial and equilibrium liquid phase E. coli concentrations (cells/mL), V is the volume of the solution (L), and m is the mass of the adsorbent (g).

The Langmuir model assumes monolayer adsorption on a homogeneous surface while the Freundlich model assumes multilayer adsorption over a heterogeneous surface (Foo and

Hameed, 2010). The Freundlich isotherm model was used for fitting the experimental data. The linearized form of the Freundlich model can be expressed mathematically as follows:

$$q_e = K_F C_e^{1/n} \quad (\text{Eq. 2.2})$$

where q_e = equilibrium adsorption capacity (number of E. coli cells measured as most probably number, MPN) mg/g.

K_F = Freundlich adsorption capacity coefficient.

$1/n$ = Freundlich adsorption intensity (Miller et al., 2001; Wang et al., 2002).

Freundlich coefficients (n and K_F) are expected to relate to the material size and bacteria type.

2.5 Adsorption Kinetics

The rate of E. coli adsorption on the adsorbents surface was described by adsorption kinetics. Steel chips, steel slag, limestone, and zeolite (1-2 mm) were chosen for kinetic tests based on the results of the adsorption isotherm experiments. Steel chips adsorption kinetics was evaluated at an initial E. coli concentrations of 2.0×10^7 MPN/mL, and 1.4×10^4 MPN/mL was used for steel slag, limestone, and zeolite. The sample preparation procedure was the same as the adsorption experiments and samples were collected at different time intervals of 30, 60, 120, 360, 720, and 1440 min then analyzed for E. coli concentration. The adsorption data was fitted to pseudo first-order and pseudo-second-order kinetic models which are widely used to illustrate adsorption rates. The pseudo first-order and pseudo-second-order kinetics equations can be given as:

$$\log(q_e - q_t) = \log q_e - \frac{k_1 t}{2.303} \quad (\text{Eq.2.3})$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (\text{Eq. 2.4})$$

where q_t is the adsorption capacity at time t (mg/g), t is the shaking time (h); k_1 , k_2 is the equilibrium rate constant of the pseudo first-order adsorption (h^{-1}), and the pseudo-second-order adsorption (g/mg-h), respectively.

2.6 Different Initial E. coli Concentration

Based on adsorption isotherm and kinetics adsorption tests, one gram of steel chips, steel slag, limestone, and zeolite was used for determining the adsorbent's E. coli removal capacity with different initial E. coli concentrations of 10, 100, 1000, and 10000 MPN/mL. An E. coli culture (10^9 MPN/mL) was diluted to 10, 100, 1000, and 10000 MPN/mL after harvesting the culture. Adsorbents were placed in 250 mL flasks and continuously shaken at 100 rpm with an orbital shaker for 2 h at 20°C. Then flasks were removed from shaker and analyzed via IDEXX method to obtain E. coli removal efficiency. For each E. coli concentration one control flask (with no adsorbent) was also prepared to determine the exact initial number of cells.

2.7 Temperature Impact

The impact of temperature in E. coli removal was investigated by using initial E. coli concentrations of 1.3×10^4 , 1.5×10^4 , and 2×10^4 MPN/mL for 5 °C, 20 °C, and 30 °C, respectively. One gram of two byproduct materials (steel chips and steel slag) in the size range of 1-2 mm was added to each solution. For each temperature one control flask (E. coli solution) was prepared to determine the exact initial E. coli cells number. The temperature controlled orbital was used for these experiment. After 2 h flasks were removed from shaker and samples were analyzed.

2.8 pH Impact

The effect of pH on E. coli removal was examined. Initial E. coli concentrations of 1.5×10^4 MPN/mL were used for adsorption under pH values of 5.0, 7.0, and 9.0. Steel chips and steel slag were placed in a 100 mL E. coli solution in 250 Erlenmeyer flask. Prior to starting the experiment the pH was adjusted to initial values of 5.0, 7.0, and 9.0 by using 1.0 M H_2SO_4 (sulfuric acid) and 1.0 M NaOH (sodium hydroxide). Samples were shaken for 2 h at 20 °C and collected for analyzed by IDEXX method.

2.9 Natural organic matter Impact.

The effect of NOM on the E. coli adsorption onto steel chips and steel slag was investigated. NOM stock solution was prepared by dissolving a Suwannee River NOM standard, which was obtained from the International Humic Substances Society, in buffer solution. NOM samples were prepared with E. coli solutions in concentrations of 0, 5, 20, and 50 mg DOC/L. Samples were removed from shaker after 2h and analyzed for E. coli concentration. The initial E. coli concentration of 2.4×10^4 MPN/mL was used for this experiment.

2.10 Salt Impact

One gram of absorbent (steel chips, steel slag, limestone, and zeolite) with the size range of 1-2 mm was chosen for investigating the impact of salt on E. coli adsorption. Salt solution was prepared by mixing potassium chloride (KCl) into buffer solution at concentrations of 100, 1000, and 10,000 mg Cl⁻/L. Then the E. coli solution was added to the buffer solution to reach 2.4×10^4 MPN/mL for by-product materials and 2.0×10^4 MPN/mL for natural minerals. After 2 h samples were removed from the orbital shaker and each solution was analyzed for E. coli concentrations.

2.11 Desorption of *E. coli*

To evaluate the desorption potential of the materials (steel slag, steel chips, limestone, and zeolite), 100 mL *E. coli* solution was prepared in 250 mL Erlenmeyer flasks then one gram of each material was placed in the flask. Samples were shaken at 100 rpm for 24 h at 20°C. Two different ratios of *E. coli* concentration were used; 2×10^7 MPN/mL for steel chips and 1.4×10^4 MPN/mL for steel slag, limestone, and zeolite. Once adsorption tests were finished, samples were analyzed for *E. coli* concentration. Afterward, the adsorbents were carefully rinsed with nanopure water, and buffer solution was poured into the flasks (included the adsorbents) to make the volume of solution the same as the adsorption test volume. The flasks were shaken for 24 h on the orbital shaker at 100 rpm, and samples were collected at different time intervals of 30, 60, 120, 360, 720, and 1440 min. Then *E. coli* concentration was determined.

The desorption ratio was obtained by the following equation:

$$D = \frac{d_e}{N_r} \times 100 \quad (\text{Eq. 2.5})$$

where D is desorption percentage, d_e is *E. coli* concentration after certain desorption time (MPN/mL), N_r is the number of *E. coli* adsorbed onto adsorbents (MPN/mL).

2.12 Column *E. coli* Adsorption Experiments

The column *E. coli* adsorption experiments were operated in four Omnifit® fixed-bed glass columns with 1.5 cm inner diameter and 15 cm height (Fig. 2.5). The masses of 30, 34, 27.5, and 18.6 gram of steel chips, steel slag, limestone, and zeolite, respectively, were placed into the columns to achieve a bed height of 10 cm. Experiments were conducted for five days. Effluent samples were collected at different time intervals of

0.25, 0.5, 2, 4, 8, 24, 36, 48, 96, 114, 192, 240, and 264 hours, then analyzed for *E. coli* concentration via the IDEXX method. A 12-liter buffer solution was prepared with the initial *E. coli* concentration range of 1.1×10^4 - 1.4×10^4 MPN/mL for each day. Solution was pumped from the bottom of the columns upward at flow rate of 1.76 mL/min to achieve empty bed contact time (EBCTs) of 10 min using a peristaltic pump. The EBCT in the column is calculated from the ratio of bed volume (mL) to the flow rate (mL/min), and is calculated as follows:

$$EBCT = \frac{\text{Bed Volume}}{Q} \quad (\text{Eq. 2.6})$$

Breakthrough curves were constructed to show the adsorption behavior of *E. coli* onto steel chips, steel slag, limestone, and zeolite. The breakthrough curves showed the adsorption of *E. coli* onto the fixed-bed columns and were expressed as the ratio of inlet *E. coli* concentration to outlet *E. coli* concentration (C/C_0) as a function of time.

3. Results and discussion

3.1 Adsorption Isotherm

The E. coli adsorption data for the four materials was fitted with the Freundlich isotherm model. The experimental results are shown in Table 3.1 and Table 3.2. As Table 3.2 shown, all adsorbents had high correlation coefficients (R^2) using the Freundlich model ($R^2 > 0.93$) at 20 °C, suggesting that the adsorption of E. coli can be characterized by the formation a multilayer adsorption of bacteria cells along the surface of the adsorbents with a heterogeneous energy distribution between active sites (Yousef et al., 2011). The results of the E. coli adsorption experiments fitted to the Freundlich isotherm models shown in Fig. 3.1, 3.2, and 3.3 for different size ranges of 0.5 – 1 mm, 1 – 2 mm, and 2 – 4 mm, respectively. The results showed that by decreasing particle sizes, the removal efficiency improved and also the maximum removal efficiencies were achieved for byproducts materials using the smallest size. Regarding natural minerals, limestone removed more E. coli cells in comparison to zeolite. Based on Cheung et al. (2007), general adsorption process included three steps; mass transfer of adsorbate to adsorbent across a surface binding (internal and external), a diffusion layer, and an external boundary layer. Previous studies also has shown that bacteria adsorption to filter media can be analyzed via particle diffusion (d_p) and film diffusion (d_f) (m²/min) whereby E. coli diffusion was followed by particle diffusion in the aqueous phase (Bonilla et al. 2007; Moussavi et al. 2011; Sochacki et al. 2011; lin et al. 2013; Guaya et al. 2015; Sang et al. 2016). The comparison of k_f values for all filter medias showed that the E. coli attachment to byproducts materials (steel chips and steel slag) was higher than attachment

to natural minerals (limestone and zeolite). This may be attributed to positively charged surface which can absorb negative charged E. coli, which results in removal efficiency over 97%. Hydrophobic attraction and electrostatic interaction are two forces that affect E. coli adsorption onto filter materials (Wang et al. 2012; Sang et al. 2016). Voorthuizen et al. (2001) reported that the thickness of the double layer is important for electrostatic interaction. Accordingly, electrolytes on the surface of materials, such as metal ions, cause attraction between bacteria cells and filter media (Pagnout et al., 2012). The zeta potential value of the filter materials also affects E. coli removal. It has been shown that iron filling have positive zeta potentials which can enhance the E. coli removal efficiency (Schwegmann et al. 2010; Hernovic et al. 2011, Walcaz et al. 2012).

3.2 Adsorption Kinetics

The results of kinetic experiments of E. coli adsorption to steel chips, steel slag, limestone, and zeolite are shown in Fig 3.4. Two different initial E. coli concentrations of 2×10^7 MPN/mL and 1.4×10^4 MPN/mL were used for steel chips and steel slag, limestone and zeolite, respectively. In general, all four of the selected materials exhibited fast adsorption kinetics. The adsorption rate of steel byproducts were larger than natural minerals. Steel chips and steel slag reached 93% and 84% of the 24 h adsorption rate within 6 h, respectively. However, limestone and zeolite reached equilibrium about 6 h. Table 3.3 shows the analyzed data for pseudo-first-order and pseudo-second-order. It can be seen both the correlation coefficient R^2 of orders are high. But the adsorption data fits pseudo-second-order reasonably well ($R^2 \geq 0.962$) for all cases. The k_1 values (pseudo-first-order kinetic constant) for steel chips, steel slag, limestone, and zeolite were obtained 0.813, 0.48, 0.393, and 0.37 (h^{-1}), respectively.

3.3 Different Initial E. coli Concentrations

Fig 3.5 illustrates the effect of different initial E. coli concentrations on filter materials for E. coli removal. This indicates that as C_0 changed from 10 MPN/mL to 10,000 MPN/mL, adsorbents removal percentage substantially decreased. The results showed that the E. coli adsorption capacities for steel chips, steel slag, limestone, and zeolite were decreased by 2.4%, 44.0%, 38.0%, and 71.0%, respectively, when the cells concentration increased from 10 to 10,000 MPN/mL. However increasing E. coli concentration did not have significant effect on steel chips adsorption capacities in comparison with other materials. Sang-Woo et al. (2015) examined the different adsorption capacities of natural zeolite and Mg^{2+} -modified zeolite (MMZs) for E. coli removal. The authors reported that when initial E. coli concentration increased from 10mg/L to 50 mg/L (based on dry cell mass) zeolite adsorption capacity was decreased. But MMZs have an enormous E. coli adsorption capacity compared to natural zeolite toward different initial E. coli concentration. Also, other studies have reported that with increasing the contamination concentration the removal capacities of filter media were decreased (Aziz et al. 2001; Erdem et al. 2004; Komnitsas et al. 2004; Pitcher et al. 2004; Rangsvivek et al. 2005; Aziz et al. 2008; Wang and Peng, 2010).

3.4 Temperature Impact

The extents of E. coli removal by steel chips and steel slag under different temperatures (5, 20, and 30°C) are shown in Fig. 3.6. Filter materials adsorption capacities were increased by 62.0% and 76.0% for steel chips and steel slag as the experimental temperature increased from 5°C to 30°C. The results suggested that E. coli adsorption was more favorable at higher temperatures. In stormwater treatment ponds,

water temperature will be dependent on season and location. Therefore, a field-scale filter structure for stormwater treatment will more than likely have higher *E. coli* removal efficiencies during the warmer months and lower efficiencies in the cooler months. Ishii et al. (2005) reported that by decreasing the temperature *E. coli* retention on filter materials decreased whereas by rising the temperature *E. coli* cells attachment to media increased.

3.5 pH Impact

In order to investigate the pH impact on *E. coli* removal by using steel chips and steel slag, pH values of 5, 7, and 9 were examined. The adsorption capacities of steel chips and steel slag were increased by 34.0% and 62.5%, respectively, as pH decreased from 9 to 5 (Fig. 3.7). Scholl and Harvey (1992) conducted the bacteria adsorption using the aquifer materials at pH 3.5 to 8 for uncontaminated groundwater and pH 3.0 to 9 for contaminated groundwater. Their results showed that pH increased, sorption of bacteria to media was decreased. The maximum bacteria adsorption in uncontaminated and contaminated groundwater occurred at pH 5 and 6.5, respectively. Also, several studies demonstrated that *E. coli* adsorption onto filter media increased significantly with decreasing pH from 10 to 4. By decreasing pH, hydroxide (OH⁻) ions decrease resulting in a decline in materials surface negative charge, thereby reducing repulsion force between *E. coli* and materials surface (Lytle et al. 1999; Tawfik et al. 2003; Guber et al. 2005; Starosvetsky et al. 2012; Delaire et al. 2015).

3.6 Natural organic matter Impact

The effect of NOM on *E. coli* retention on steel chips and steel slag was examined. Fig. 3.8 shows the impact of adding different NOM concentrations (0, 5, 20,

and 50 mg DOC/L) on *E. coli* adsorption. The results showed that *E. coli* adsorption capacities were reduced by 21.0% and 44.0%, and 25.5% and 43.0% in the presence of 20 and 50 mg DOC/L onto steel chips and steel slag, respectively. However, Fig. 3.8 showed that 5mg DOC/L did not influence *E. coli* removal. The main reason is described by the competitive attachment of *E. coli* cells and dissolved organic carbon (DOC) onto the positively charged adsorbents. This is similar to a study by Delaire et al. (2015) which reported a reduction in bacteria removal efficiency after adding 3 mg/L of NOM (Suwannee River fulvic acid). This is likely due to attachment of NOM on filter materials surface and increase the negative surface charge which led to a decrease in *E. coli* attachment (Song et al. 1994; Johnson et al. 1995). Also, Johnson and Logan (1996) reported that in the presence of Suwannee River dissolved organic matter in solution, bacteria attachment on quartz and Fe-quartz was decreased about 20% and 10%, respectively.

3.7 Salt Impact

The impact of adding different initial salt concentrations (100, 1000, and 10,000 mg Cl⁻/L) on *E. coli* adsorption onto four materials (steel chips, steel slag, limestone, and zeolite) was investigated. Results shown in Fig. 3.9 illustrated that as the salinity increased more *E. coli* were attachment to adsorbents. The *E. coli* adsorption capacities of steel chips, steel slag, Limestone, and zeolite were improved by 5%, 35.5%, 45.0%, and 78.0%, respectively, when the initial salt concentration increased from 0 to 10,000 mg Cl⁻/L. As can be seen in Fig. 3.9, increasing chloride concentration had a significant effect on natural minerals adsorption capacities in comparison to byproduct materials. It may be that the increase in the ionic strength caused a reduction in electrical double layer on both

media and *E. coli* cells surface, thereby allowing the *E. coli* cells to approach the filter media surface, and Van der Waals force as an attractive power exceeds electrostatic repulsion (Fonts et al., 1991; Mills et al., 1994; Guber et al. 2005). However, Starosvetsky et al. (2012) observed that high salt concentration (above 140 mM) prevented bacteria strains (*E. coli* and *Salmonella typhimurium*) attachment on media surface. Some studies also demonstrated that for certain bacteria types, high salt concentration reduced adsorption capacity and caused desorption from porous media (Daniels 1972; Wood et al. 1980).

3.8 Desorption of *E. coli*

Fig. 3.10 illustrated *E. coli* desorption of the four filter materials (steel chips, steel slag, limestone, and zeolite). The absorbents ability to keep previously adsorbed *E. coli* is critical to the success of field-scale filter, since the release of *E. coli* will increase the pathogen loading to natural water bodies. After an initial batch adsorption experiment for examining the *E. coli* removal capacity by filter media, another batch experiment was applied to investigate the de-sorbability of *E. coli* over time. Over a period of 24 h, steel chips, steel slag, limestone, and zeolite desorbed 0.04, 0.4, 3.11, and 2.73% of the attached *E. coli* cells, respectively. *E. coli* release from steel chips is almost negligible to compare with other absorbents. As already described in adsorption part, steel chips was able to remove *E. coli* up to 95%. On the other hand, natural minerals (limestone and zeolite) had more desorption capacities than steel byproducts. limestone showed faster *E. coli* release the beginning of desorption test, later the curve was flattened and reached equilibrium. However, the zeolite curve is similar to straight line suggesting desorption of *E. coli* will be continuing as time goes on. Some factors play roles in *E. coli* desorption

from filter materials as follows; the strength of surface adsorption, filter media surface charge, filter materials particle size, zeta potential, contact time, temperature, existing of any pollutants like NOM, and pH value. As already mentioned, natural minerals have negative surface charge which led to less adsorption capacity and more desorption because of surface repulsive force between *E. coli* and minerals. Also, Wang et al. (2014) concluded that with increasing of quartz sand particle size desorption ratio was improved and adsorption capacity was reduced. Large particles desorption capacity was higher than fine particles and they took longer time to reach equilibrium.

3.9 Column *E. coli* Adsorption Experiments

The breakthrough curves for steel chips, steel slag, limestone, and zeolite are shown in Fig. 3.11. All filter materials have an immediate breakthrough of *E. coli*, reaching initial removal rates of 95.7%, 51.2%, 42.0% and 21.2% for steel chips, steel slag, limestone, and zeolite, respectively. The steel slag, limestone, and zeolite curves are then characterized by a slow decrease in *E. coli* removal efficiency over time. As can be seen in Fig. 3.11 the effluent *E. coli* concentration of the natural minerals reached 95% of the influent concentration after 96 h. The steel slag showed a 95% breakthrough in the effluent after 264 h adsorption. However, the steel chips breakthrough curve exhibited no significant decrease ($C/C_0 \% < 7\%$) after 11 days. The adsorption of *E. coli* onto steel slag and, limestone and zeolite showed a traditional S-shaped breakthrough curve and reached full exhaustion ($C/C_0 \% = 100\%$) after 11 days and 4 days, respectively. In contrast, steel chips had a high *E. coli* removal until day 11, and never reached completely exhaustion. Therefore, it is possible that this is a result from the continuous

regeneration of adsorption sites due to continuous oxidation in the column and forming new Fe oxides for E. coli to bind with.

4. Conclusions

This study was conducted to investigate E. coli removal from stormwater runoff using byproduct materials (steel slag and steel chips) and natural minerals (zeolite and limestone). The results of the batch adsorption experiments and column study illustrated that industrial byproducts were more effective at removing E. coli from storm water than natural minerals. As material sizes increased E. coli adsorption capacities onto filter materials were decreased. In general, all four of the selected materials exhibited fast adsorption kinetics. The presence of varying initial E. coli concentrations (10, 100, 1000, and 10000 MPN/mL) had a large impact on E. coli adsorption onto all filter materials except steel chips. With increasing the temperature adsorption capacities were increased from 36.9% to 96.2% and 7.5% to 30.6% for steel chips and steel slag from 5°C to 30°C. As pH decreased from 9 to 5 steel chips and steel slags' E. coli adsorption capacities improved.

The results of adding salt on E. coli removal revealed a significant impact on natural minerals adsorption capacities in comparison with steel byproducts. E. coli removal efficiency were reduced by 19.8% and 41.8% onto steel chips and 6.8% and 11.5% onto steel slag in the presence of 20 and 50 mg DOC/L. The desorption results showed that E. coli release from steel chips was negligible in comparison with other absorbents since the strong chemical bonds between steel byproducts and E. coli prevented the release of E. coli. Overall, the results of this study suggest that steel byproducts are highly efficient for E. coli removal under variable environmental conditions.

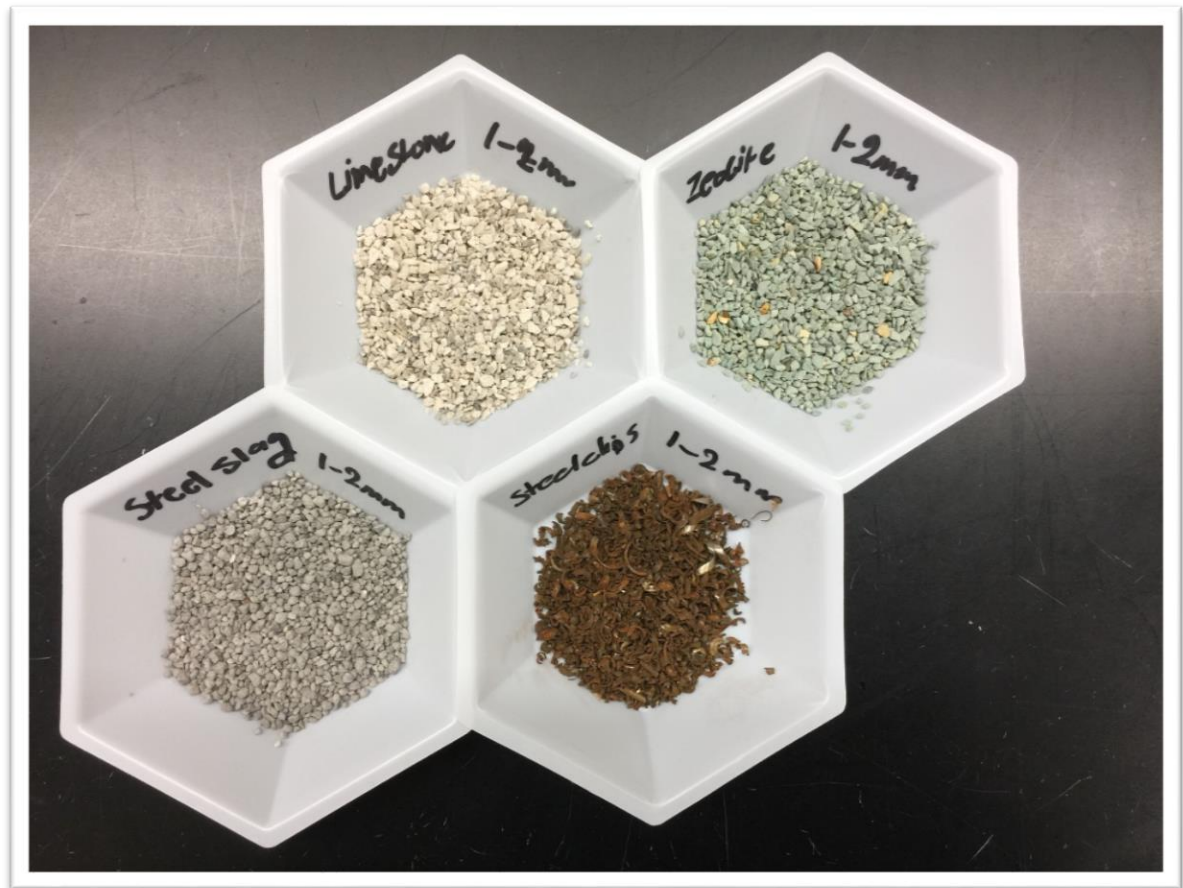
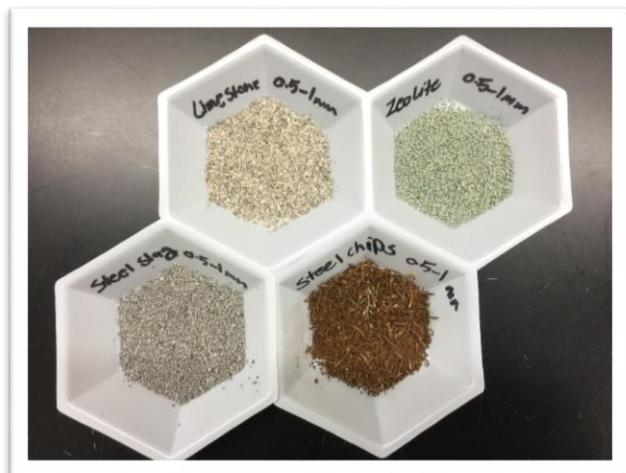


Figure 2.1: Filter materials (left to right : Limestone, zeolite, steel slag, and steel chips).



a.



b.



c.

Figure 2.2: Filter materials in size range 0.5 – 1 mm(a), 1.0 – 2.0 mm (b), and 2.0 – 4.0 mm (c).



Figure 2.3: Temperature controlled orbital shaker.



Figure 2.4: IDEXX Quanti-Tray/2000 (Quanti tray sealer, tray, vessel, and reagent).

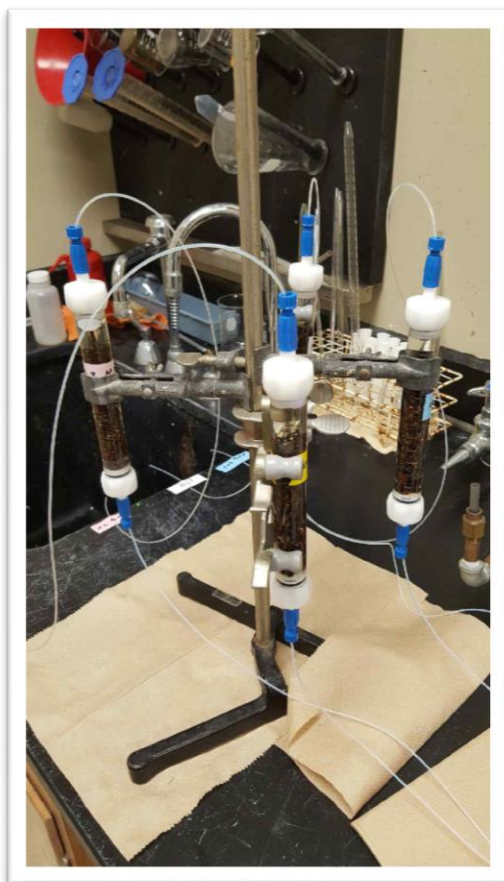


Figure 2.5: Column experiment equipment.

Table 2.1: Characteristics of industrial byproducts and natural minerals.

Material	Size^a (mm)	Particle Density (g/cm³)	pH^b
	0.5-1		
Steel chips	1-2	5.20	6.3
	2-4		
	0.5-1		
Steel Slag	1-2	3.57	10.9
	2-4		
	0.5-1		
limestone	1-2	2.76	9.0
	2-4		
	0.5-1		
Zeolite	1-2	2.18	8.0
	2-4		

^a Size ranges determined from known sieve sizes.

^b Values of pH were obtained from a 1:1 by weight slurry mixture of material and distilled water.

Table 3.1: Filter materials adsorption capacities in different size range.*

Material	Size (mm)	Mass (gram)	Time (hours)	Removal (%)
Steel Chips	0.5-1	1	24	92.0
	1-2			90.0
	2-4			87.0
Steel Slag	0.5-1	1	24	96.3
	1-2			94.0
	2-4			92.0
Limestone	0.5-1	1	24	49.0
	1-2			32.2
	2-4			13.1
Zeolite	0.5-1	1	24	32.2
	1-2			23.5
	2-4			6.9

*Experimental conditions: Initial E. coli concentration = 2.0×10^7 MPN/mL for steel chips, and 1.4×10^4 MPN/mL was used for steel slag, limestone, and zeolite; adsorption time=24h; pH=7.0.

Table 3.2: Freundlich isotherm constants parameters for the E. coli adsorption onto filter materials.*

Material	Size (mm)	Freundlich Isotherm		
		K_F	n	R^2
Steel Chips	0.5-1	0.6	0.78	0.96
	1-2	0.61	0.7	0.98
	2-4	0.66	0.52	0.97
Steel Slag	0.5-1	0.73	2.7	0.97
	1-2	0.65	2.27	0.98
	2-4	0.51	1.82	0.98
Limestone	0.5-1	0.58	1.37	0.96
	1-2	0.54	1.67	0.98
	2-4	0.39	1.48	0.93
Zeolite	0.5-1	0.69	1.86	0.99
	1-2	0.66	1.74	0.97
	2-4	0.64	1.69	0.97

*Experimental conditions: Initial E. coli concentration = 2.0×10^7 MPN/mL for steel chips, and 1.4×10^4 MPN/mL was used for steel slag, limestone, and zeolite; adsorption time = 24 h; pH = 7.0.

Table 3.3: Kinetic parameters for E. coli adsorption onto filter materials.*

Material	Size (mm)	Pseudo first-order model		Pseudo second-order model	
		k ₁ (h ⁻¹)	R ²	k ₂ (g/mg-h)	R ²
Steel Chips	1.0-2.0	0.813	0.9685	6	0.9776
Steel Slag	1.0-2.0	0.48	0.8762	2.6	0.9678
Limestone	1.0-2.0	0.393	0.7569	2.387	0.9936
Zeolite	1.0-2.0	0.37	0.6479	2.34	0.9864

*Experimental conditions: Initial E. coli concentration = 2.0×10^7 MPN/mL for steel chips, and 1.4×10^4 MPN/mL was used for steel slag, limestone, and zeolite; collected sample at 30, 60, 120, 360, 720, and 1440 min = 24 h; pH=7.0.

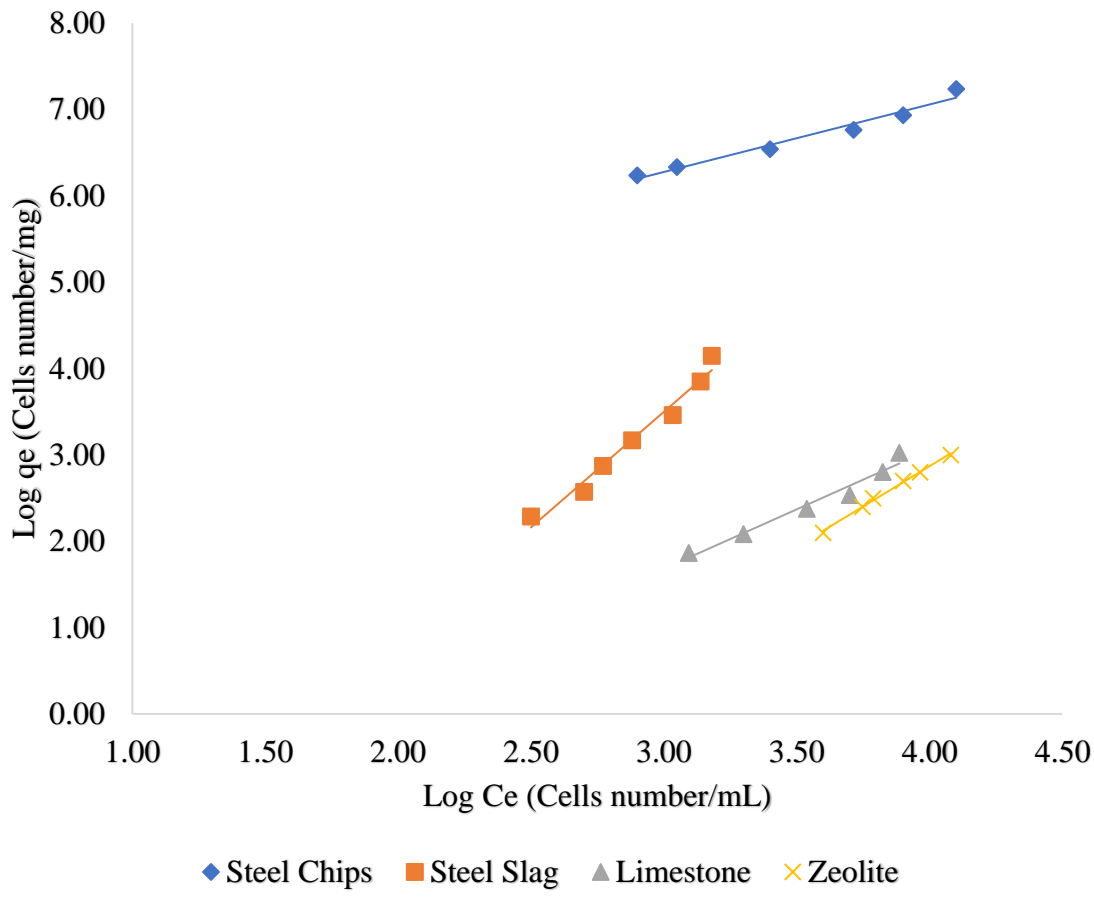


Figure 3.1: Freundlich adsorption isotherms for filter materials in size range 0.5 – 1 mm. (Experimental conditions: Initial E. coli concentration = 2.0×10^7 MPN/mL for steel chips, and 1.4×10^4 MPN/mL was used for steel slag, limestone, and zeolite; adsorption time = 24 h; pH = 7.0.)

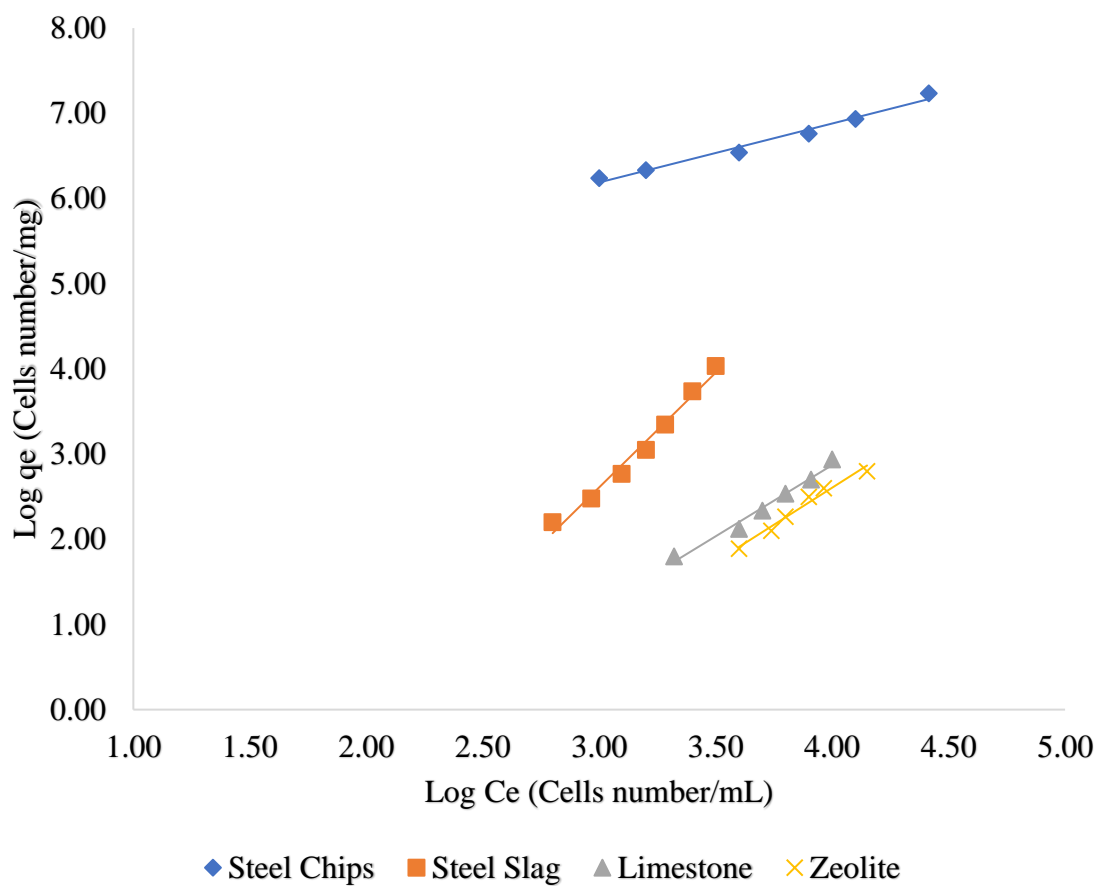


Figure 3.2: Freundlich adsorption isotherms for filter materials in size range 1 – 2 mm. (Experimental conditions: Initial E. coli concentration = 2.0×10^7 MPN/mL for steel chips, and 1.4×10^4 MPN/mL was used for steel slag, limestone, and zeolite; adsorption time = 24 h; pH = 7.0.)

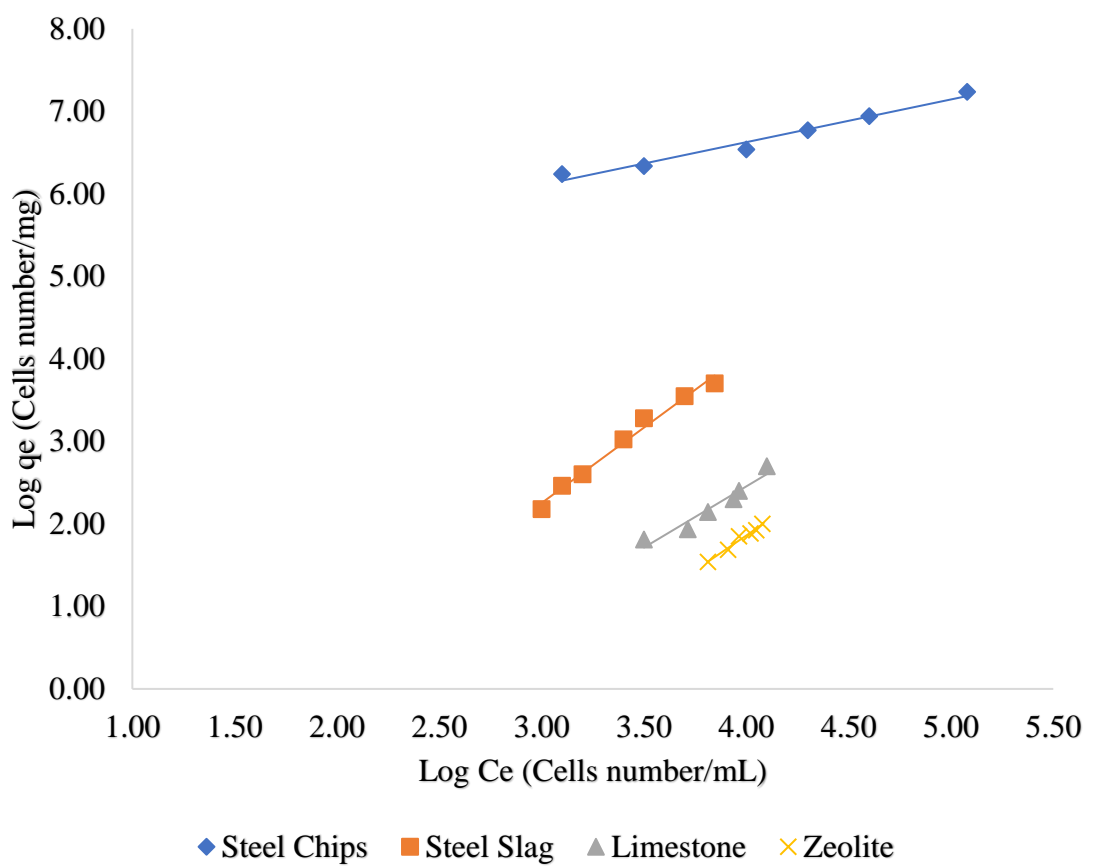


Figure 3.3: Freundlich adsorption isotherms for filter materials in size range 2 – 4 mm. (Experimental conditions: Initial E. coli concentration = 2.0×10^7 MPN/mL for steel chips, and 1.4×10^4 MPN/mL was used for steel slag, limestone, and zeolite; adsorption time = 24 h; pH = 7.0.)

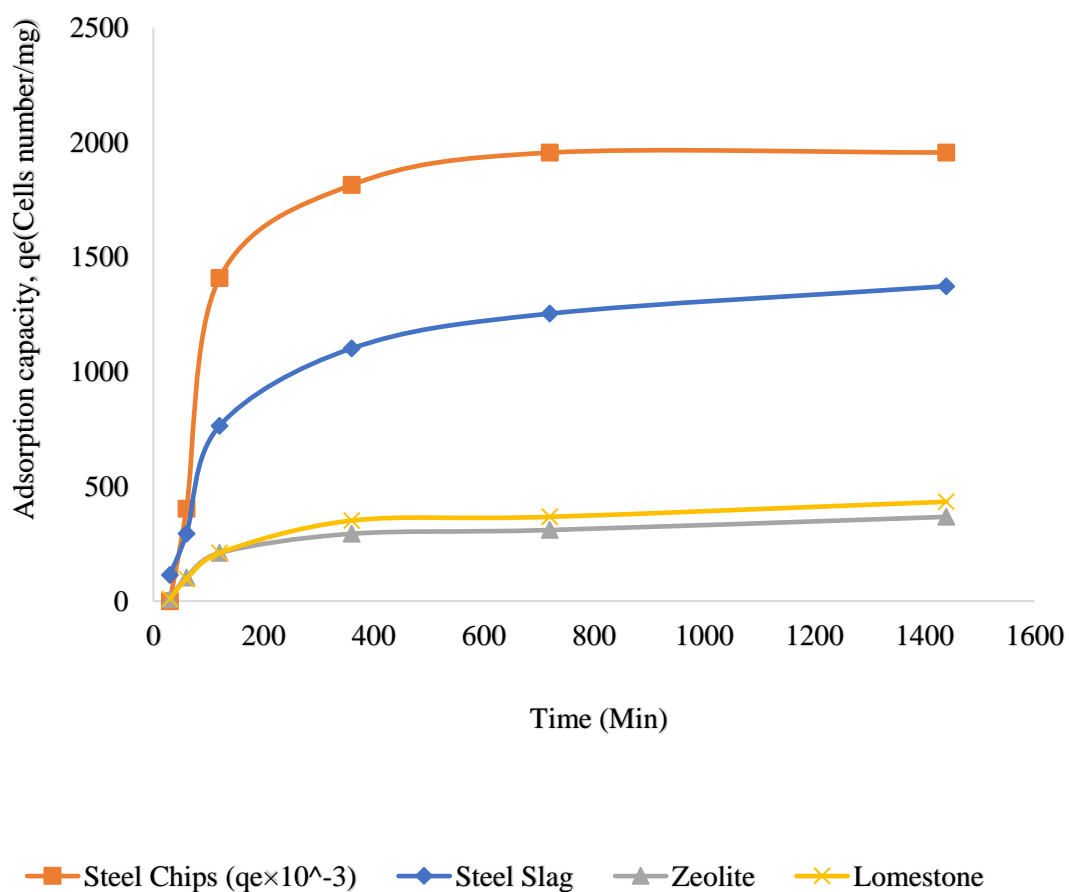


Figure 3.4: Effect of time on E. coli removal (Kinetics adsorption). (Experimental conditions: Initial E. coli concentration = 2.0×10^7 MPN/mL for steel chips, and 1.4×10^4 MPN/mL was used for steel slag, limestone, and zeolite; collected sample at 30, 60, 120, 360, 720, and 1440 min; pH=7.0.)

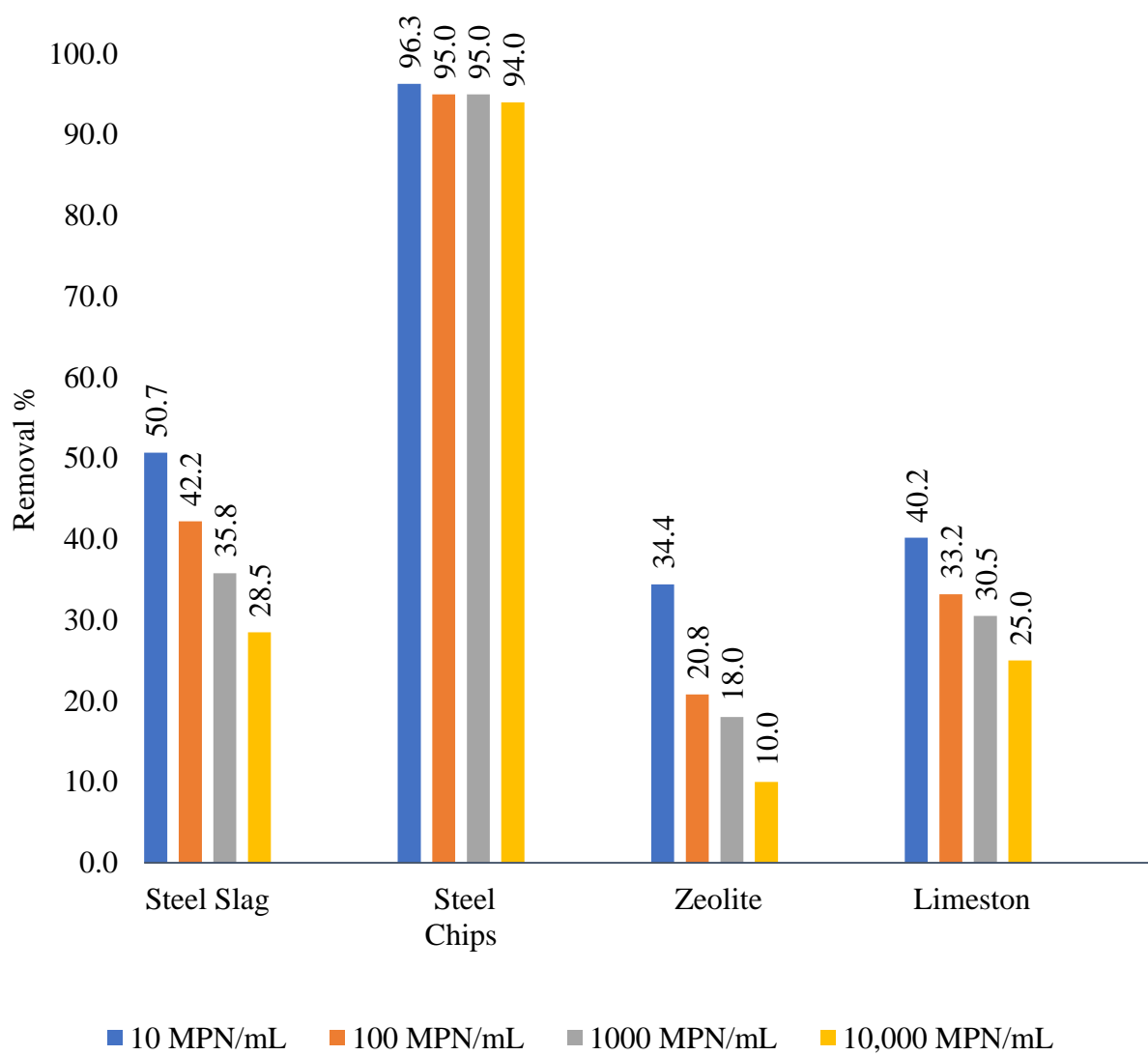


Figure 3.5: Effect of different initial E. coli concentrations on adsorption.
(Experimental conditions: Initial E. coli concentration =10.0, 100.0, 1000.0, and 10,000.0 MPN/mL, adsorption time = 2 h; pH=7.0.)

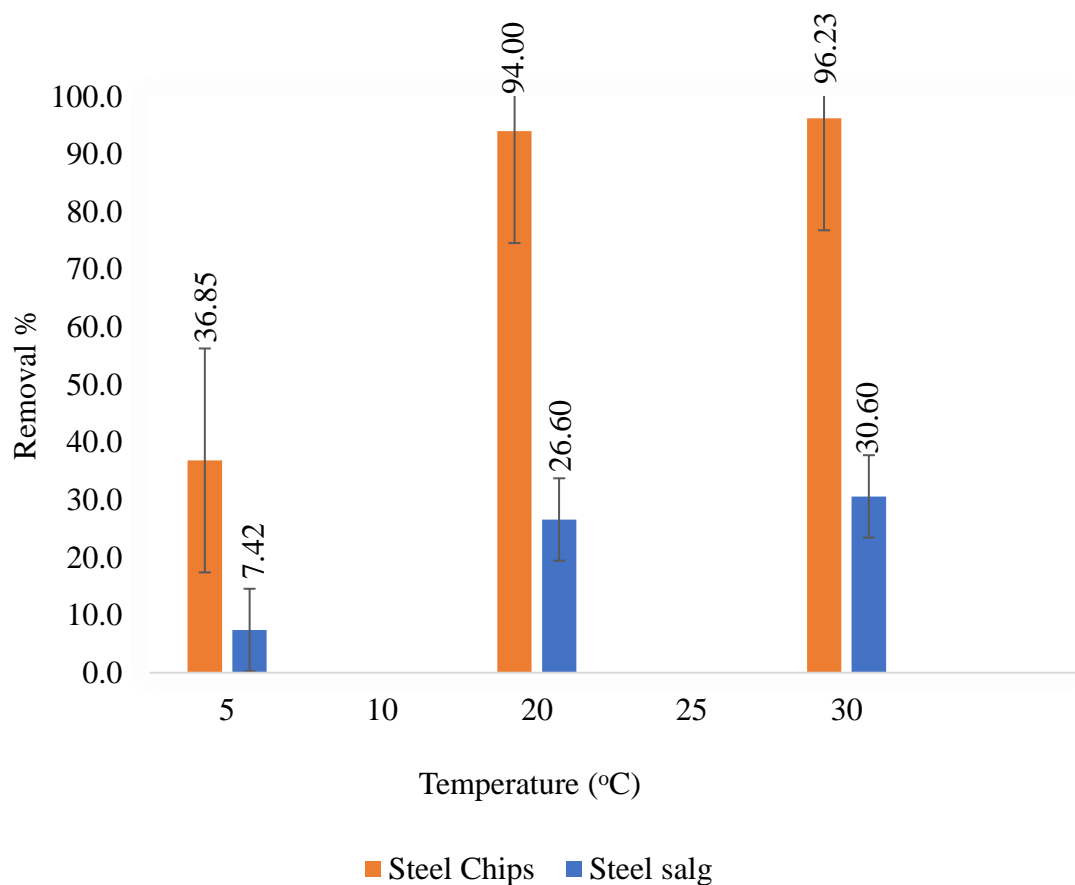


Figure 3.6: Effect of different temperature on E. coli adsorption. (Experimental conditions: Initial E. coli concentration = 1.4×10^4 MPN/mL, adsorption time = 2h; temperature = 5, 20, and 30°C; pH=7.0, error bars indicate standard deviation)

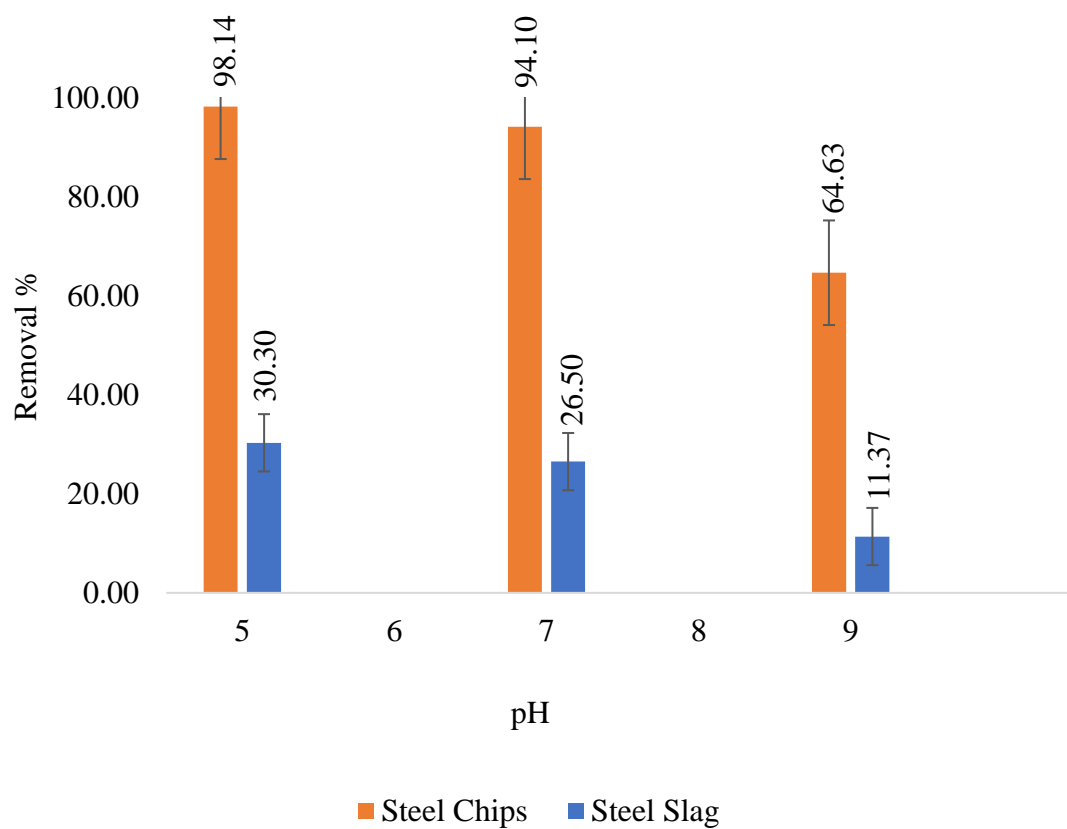


Figure 3.7: Effect of different pH on E. coli adsorption. (Experimental conditions: Initial E. coli concentration = 1.4×10^4 MPN/mL, adsorption time = 2 h; temperature = 5, 20, and 30°C; pH=7.0, error bars indicate standard deviation.)

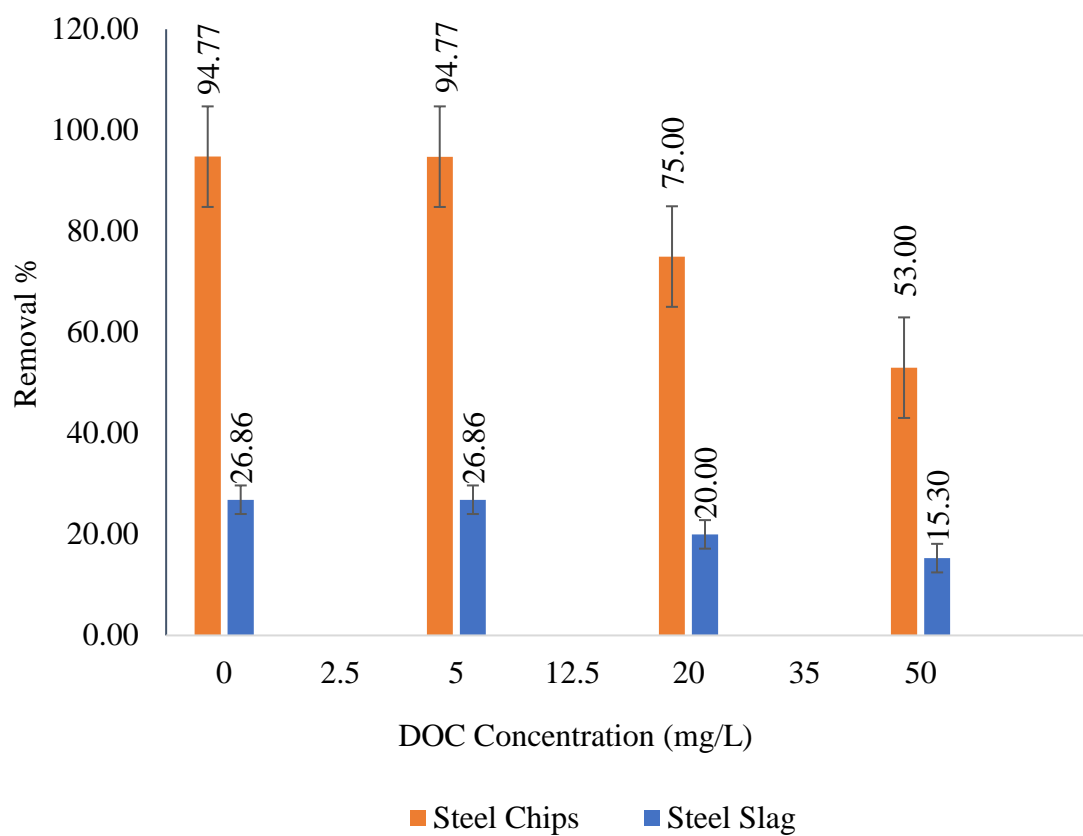


Figure 3.8: Effect of different natural organic matter concentration on E. coli adsorption. (Experimental conditions: Initial E. coli concentration = 1.4×10^4 MPN/mL, adsorption time = 2 h; temperature = 20°C; pH=7.0; error bars indicate standard deviation, NOM = 0, 5, 20, and 50 mg/L.)

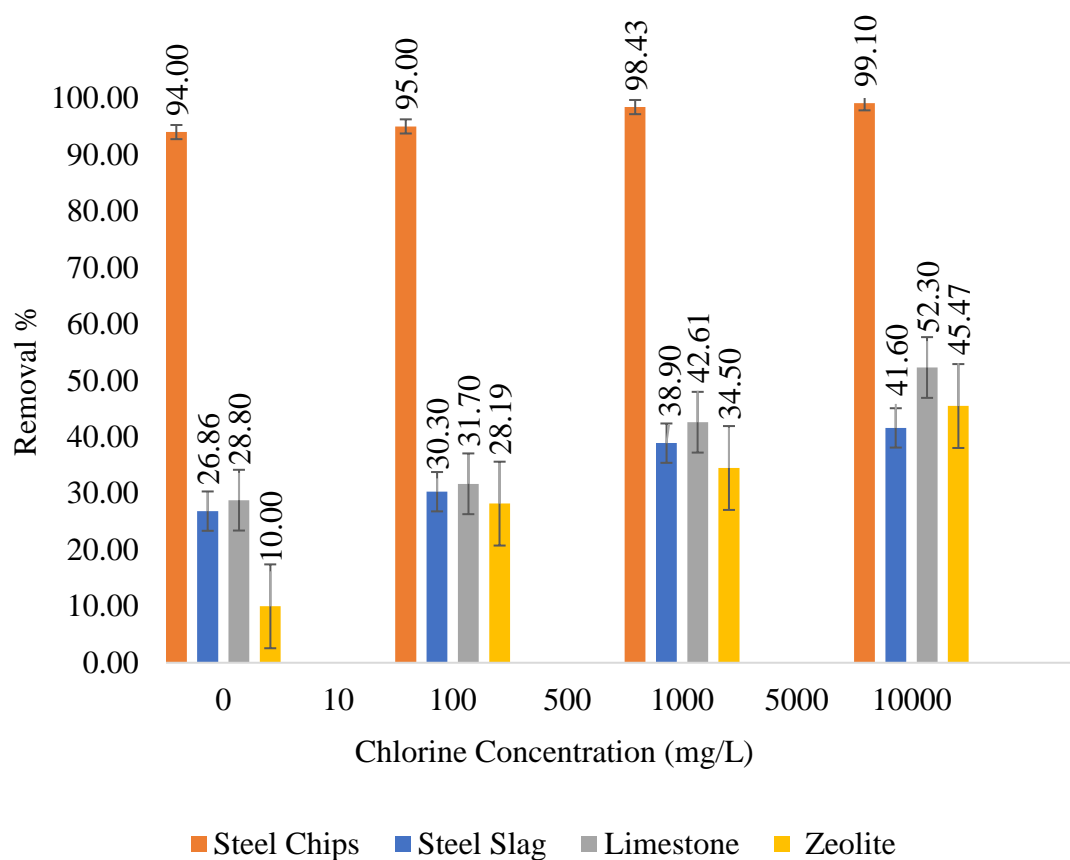


Figure 3.9: Effect of different salt (KCl) concentration on *E. coli* adsorption.

(Experimental conditions: Initial *E. coli* concentration = 1.4×10^4 MPN/mL, adsorption time = 2 h; temperature = 20°C; pH=7.0; error bars indicate standard deviation, salt concentration = 0, 100, 1000, and 10,000 mg/L.

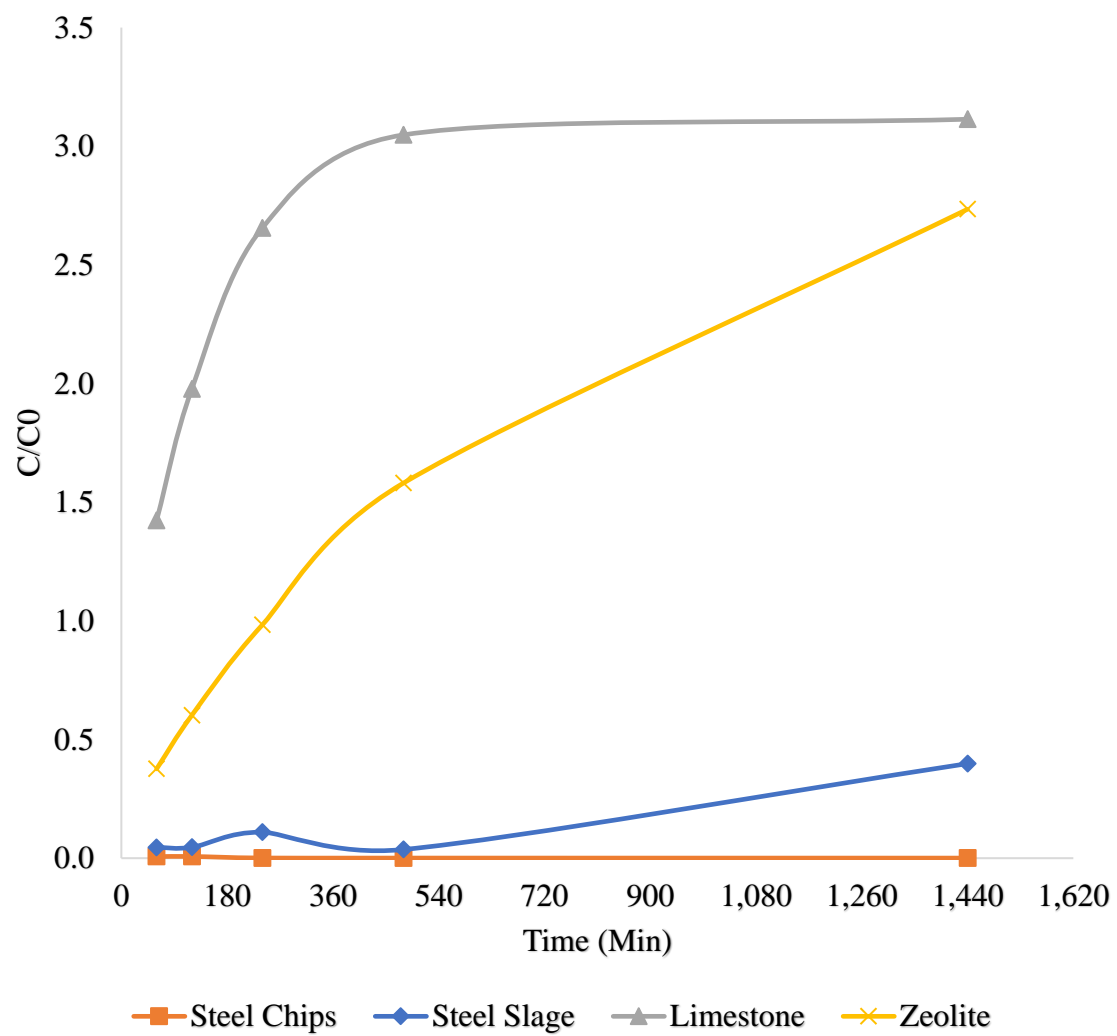


Figure 3.10: Desorption of *E. coli*. (Time = 30, 60, 120, 360, 720, and 1440 min; temperature = 20°C; pH=7.0.)

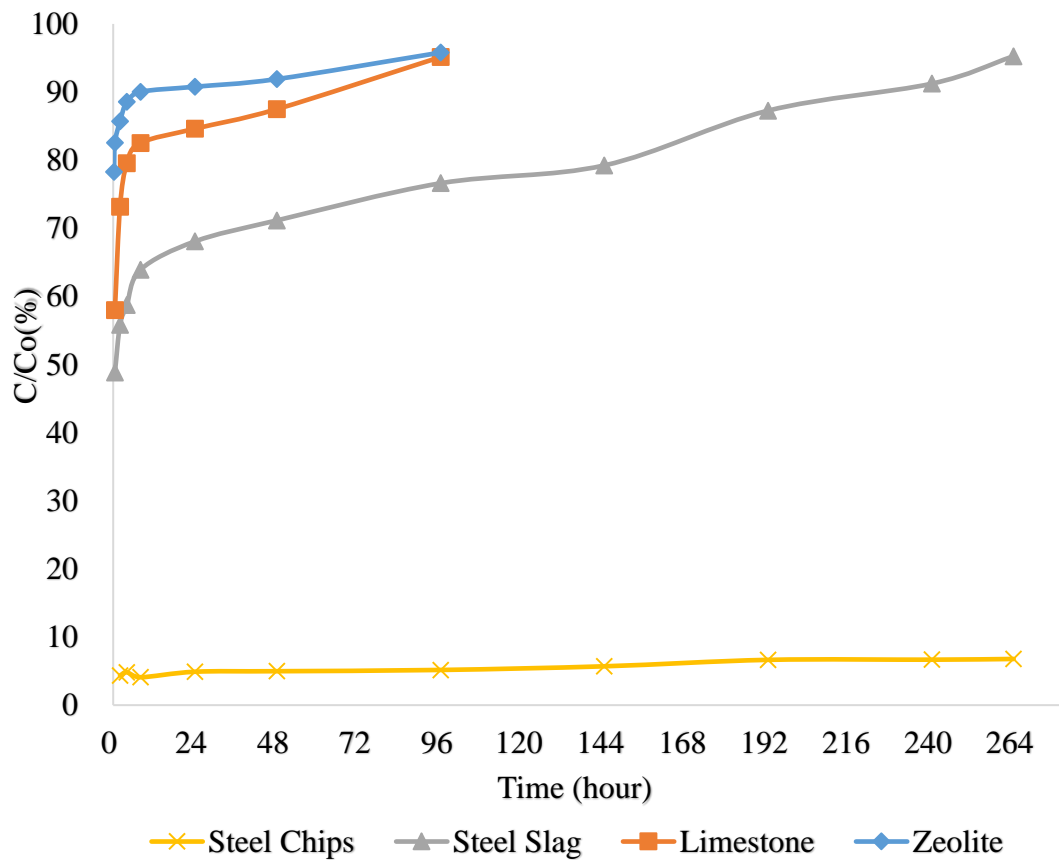


Figure 3.11: column study for *E. coli* adsorption onto filter materials. Breakthrough curves of filter materials (Initial *E. coli* concentration = 1.0×10^4 MPN/mL; EBCT=10 min; experiment duration=5 d; bed height=10 cm; error bars indicate standard deviation,)

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