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Long Term Evaluation of E. Coli Removal From Stormwater Using Recycled Steel Byproducts

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LONG TERM EVALUATION OF E. COLI REMOVAL FROM STORMWATER
USING RECYCLED STEEL BYPRODUCTS

BY

BRENDEN OLEVSON

A thesis submitted in partial fulfillment of the requirements for the

Master of Science

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THESIS ACCEPTANCE PAGE

Brenden Olevson

This thesis is approved as a creditable and independent investigation by a candidate for the master's 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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ABBREVIATIONS

CFU	Colony Forming Units
CEC	Contaminants of Emerging Concern
CWA	Clean water act
DBP	Disinfection By Products
EBCT	Empty Bed contact time
EPA	Environmental Protection Agency
E. COLI	Escherichia Coli
HAA	Haloacetic Acid
MCL	Maximum Contaminant Level
MCLG	Maximum Contaminant Level Goal
MPN	Most Probable Number
NOM	Natural Organic Matter
NPSP	Non-point source pollution
GM	Geometric Mean
SMP	Stormwater Management Practices
STV	Statistical Threshold Values
TOC	Total Organic Carbon

THM Trihalomethane

UV Ultraviolet Light

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ABSTRACT

LONG TERM EVALUATION OF E. COLI REMOVAL FROM STORMWATER
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BRENDEN OLEVSON

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Stormwater runoff often carries a variety of pollutants and pathogens that can endanger public health and wildlife. One contaminant of concern often highlighted in stormwater quality is Escherichia Coli (E.Coli) because its presence indicates microbial contamination in water. Different types of media filtration are being studied to reduce contaminant concentrations in stormwater. In this study, steel byproducts were examined as a potential material to use in media filtration to remove E. Coli and other contaminants from stormwater. Batch scale studies were conducted in laboratory to evaluate the E. Coli removal potential of steel chips and steel slag over a 24-hour period. Steel chips were able to remove over 99% of initial E. Coli concentrations and steel slag removed between 46% and 73% of initial E. Coli concentrations. Laboratory column studies were conducted to evaluate the performance of steel chips and steel slag under an Empty Bed Contact Time (EBCT) of 15 minutes and fixed E. Coli concentrations. Column studies examining the steel slag showed an average E. Coli removal of 25% and 18% for aged and new material respectively. Column studies examining the steel chips showed an average E. Coli removal of 83% and 45% for new and aged material respectively. Based on these results, a third column study was performed to examine the impact of steel chips to slag ratio on E. Coli removal. Ratios of 5%, 10%, 20%, and 50% steel chips removed an average of 35%, 50%, 57%, and 62% of E. Coli respectively. A pilot filter was

constructed at the inlet of a retention pond in Brookings, SD to examine the performance of steel byproducts under real conditions. In 2019 and 2020 the field filter was composed of 50% steel slag and 50% steel chips; this ratio was modified to 70% steel slag and 30% steel chips in 2021. Influent and effluent samples were collected from the filter in 2019, 2020, and 2021 to examine its effect on E. Coli, total phosphorus, orthophosphate, total nitrogen, nitrate, and iron. In 2019, the field filter removed an average of 53%, 45%, 53%, and 42% of E. Coli, total phosphorus, orthophosphate, and total nitrogen respectively. In 2020, the field filter removed average of 54%, 51%, 45%, 45%, and 8% of E. Coli, total phosphorus, orthophosphate, total nitrogen, and nitrate respectively. In 2021, the field filter removed an average of 29%, 31%, 65%, 41%, and 8% of E. Coli, total phosphorus, orthophosphate, total nitrogen, and nitrate respectively. During 2019 and 2020 the field filter increased dissolved iron concentrations in the effluent by an average of 0.27 mg/l. The 2021 field filter had a lower net average increase in dissolved iron concentration of 0.02 mg/l.

1. INTRODUCTION

1.1 Stormwater Pollution

1.1.1 Stormwater Pollution Sources

Pollution has become increasingly problematic as global populations and industrialization continue to rise. Contamination of natural water systems has been a concern for many years and countries have addressed this by enacting pollution laws. The United States first passed the Federal Water Pollution Control Act of 1948 to address water pollution. Over time these measures were determined insufficient, so congress passed a law dubbed the Clean Water Act (CWA) in 1972 (U.S. EPA 1972). The CWA established pollutant discharge limits and permits, more strictly regulated industrial wastewater, and funded efforts to promote cleaner water systems in the United States. Over time, this heavily reduced contamination levels in many water systems by monitoring the pollution sources. However, unseen manmade sources of pollution still often contaminate water systems.

Nonpoint source pollution (NPSP) is a major source of pollution in stormwater, nonpoint meaning it does not originate from a single distinguishable source. It is difficult to track and control NPSP because it comes from a seemingly infinite number of sources. All unregulated activities like fertilization, improperly managed land use, walking pets, small chemical spills, etc. contribute to NPSP. These activities contribute to NPSP by combining with runoff generated during storm events. Stormwater runoff occurs when precipitation events cause an excess amount of water to flow over surfaces. This is

caused by large rain events or periods with high snowmelt. When water flows over surfaces it often picks up pollutants and contaminants which can pose a threat to water systems. Section 319 of the amendments to the CWA in 1987 tries to address NPSP by providing yearly funding for states to reduce contamination levels in their water systems (U.S.EPA 1987).

The agricultural industry is often viewed as a large contributor of nonpoint source pollution. In 2012 in the United States there was 915 million acres of farmland, encompassing 40 percent of the continental United States (USDA 2014). This area is used for many different practices, but the majority is for livestock and crops. Modern farming practices are reliant on a variety of nutrients, pesticides, and other chemicals to achieve desired crop and livestock yields. When rainfall occurs, stormwater is washed over farms, bringing nutrients, bacteria, metals, pesticides, herbicides, and other contaminants into bodies of water. Studies have found that as runoff intensity increases, contaminant concentrations also increase (Thurston-Enriquez et al. 2005, Lui et al. 2014, Yao et al. 2021). Contaminants are difficult to trace, and therefore hard to regulate.

Urbanization has also led to an increase in the amount of nonpoint source pollution found in water supplies. As of 2018, 55 percent of the world's population lives in urban areas, and this is expected to grow to 68 percent by 2050 (United Nations, 2018). Urbanization leads to a higher total area of impervious surfaces like streets, parking lots, buildings, and sidewalks. These surfaces do not allow the runoff to soak into the ground which creates higher volumes of runoff water. Runoff from urban areas often carries a variety of pollutants that are detrimental to ecosystems. The increased runoff volumes can overwhelm natural water systems and lead to a steep decline in its quality. A

study conducted on the lakes of Bengaluru, India in 2021 directly correlated a decline in water quality with heavy urbanization of the city. It found very poor water quality in the urban watersheds nearest to the city of Bengaluru (Birawat et al. 2021). This study is indicative of potential water quality issues as large cities grow.

1.1.2 Stormwater Composition and Contaminants

The composition of stormwater runoff is dependent on several factors, including: the specifics of the rain event, how the area is used, and what systems are in place to manage it. Stormwater composition therefore varies around the world, but some constituents are common. Pollutants often found in stormwater systems include nutrients, suspended solids, microorganisms, and contaminants of emerging concern (CECs).

Excess nutrients like nitrogen and phosphorus are common in water systems near areas with large fertilizer use. Abundance of them promotes growth of plants and algae which can be detrimental to ecosystems. Nutrients discharged into water systems have been a major concern worldwide since the 1970s, after an increase in the use of fertilizers following rapid urbanization and industrialization after World War II (Bonsdorff, 2021). During this time, scientists found that the increased plant and algae growth leads to eutrophication, the process of which removes oxygen from the water and kills large populations of wildlife. Eutrophication can ultimately lead to dead zones in water systems in which no organisms reliant on oxygen can survive.

Suspended solids are commonly transported through the physical erosion of soils through storm events. As rain falls and runs off impermeable surfaces it often picks up sediment and fine particles that are brought into stormwater runoff. In small amounts

these are harmless to ecosystems, but large quantities varying from the normal concentrations can affect plants and wildlife. Increased suspended solids may deposit in sensitive ecosystems, blocking out sunlight or creating sediment build ups. Reduced levels of sunlight and sediment build up can damage ecosystems by changing the natural balance of organisms.

Microorganisms are common in the natural environment and are important to maintain the balance of ecosystems. While many are harmless, some microorganisms are pathogenic meaning they can cause disease. Pathogens may cause serious harm to human and animal health depending on the type and their abundance. Common health risks associated with contaminated water may include diarrhea, fever, vomiting, and intestinal pain. Highly contaminated waters may have more serious health complications including organ failure or death. Animal or human waste collected by the runoff during storm events often increases the risk of pathogens in water supplies. Faults in wastewater infrastructure like worn or improperly placed wastewater lines can also contribute to this. Stormwater often recharges aquifers, while microorganisms are often removed by the fine soils protecting them, large concentrations of microorganisms can impact them. A 2021 study found five of 12 microbial targets of concern detected more than once at multiple sampling points in a water table 30 feet below ground (De Lambert et al. 2021). This study shows the impact of human activities on groundwater, as groundwater sources traditionally have little to no microorganism populations.

Contaminants of emerging concern (CEC) have been a relatively recent area of study. As new chemicals are created, new chemical pollutants are as well. New advancements in pharmaceuticals, personal care products, industrial products, and other

chemicals have led an increase in the number of CECs in stormwater. A study in Minneapolis, Minnesota examined the abundance of CECs in stormwater, investigating 384 known CECs. It found that 123 CECs were present in 36 stormwater samples at nine sampling sites (Fairbairn et al. 2018). Contaminated stormwater often reaches surface and groundwater sources. Studies show that large concentrations of CECs that pose a risk to environmental health have been found in surface waters across the United States. These include chemicals like polycyclic aromatic hydrocarbons, pesticides, tire wear particles, and pharmaceuticals (Masoner et al. 2019, Saifur and Gardner 2021). Groundwater supplies recharged by runoff are impacted less by CECs, as they often have fine soils protecting them. However, trace concentrations of CECs can still be found in wells and drinking water supplies. CECs pose significant threats to the environment and human health because many of their effects have not been formally studied. Some may pose short term health risks and may be relatively harmless while others may have detrimental long-term health effects.

1.1.3 Stormwater Management Practices

To manage stormwater and protect ecosystems from sudden contaminant influxes, there are several stormwater management practices (SMP) frequently used. The most common are detention and retention ponds. Both are designed to hold runoff from precipitation events to prevent sudden flooding or pollution in natural water systems. The key difference between them is detention ponds are designed to hold water for short periods and reintroduce it into nearby water systems, whereas retention ponds hold the water indefinitely. These practices are important because they reduce the amount of nonpoint source pollution in natural water systems. Detention ponds gradually introduce

runoff into lakes, rivers, streams, or aquifers so that changes in water quality are less drastic. Large sudden changes in contaminant levels like nutrients, microorganisms, suspended solids, or toxins would have a negative impact on wildlife. Retention ponds do not reintroduce the runoff into natural systems through a direct outlet but often self-treat the water over time due to large microorganism communities existing within them. A specific type of retention pond called a bioretention pond is also frequently used. These operate similarly to normal retention ponds but have vegetated areas that help the treatment of the stormwater.

Wetlands are another common SMP. Natural and constructed wetlands use a variety of plants, animals, and other organisms to control the quality of stormwater introduced to them. They do this through processes like sedimentation, adsorption and retention, biological degradation, and plant uptake. After flowing through the wetland, water is often discharged into infiltration basins. In these, the water continues being treated as it flows through layers of permeable soils to eventually reach the water table.

1.2 E. Coli occurrence and treatment technologies in stormwater

1.2.1 E. Coli Occurrence

1.2.1.1 Background and Explanation

Escherichia coli (E. Coli) refers to a group of bacteria, common in animal intestines and many places in the environment. E. Coli is commonly used to indicate potential health risks with bodies of water used for human purposes. Fecal coliforms are used to quantify the safety of bodies of water and are typically measured using E. Coli as an indicator organism. To limit pollutants and substances of concern in water systems the United States Environmental Protection agency (U.S. E.P.A) passed the CWA (Clean Water Act). In addition to this document the U.S. EPA published *Ambient Water Quality Criteria for Bacteria*, which recommended levels of E. Coli between 100 and 126 CFU per 100 mL in fresh recreational waters (U.S. E.P.A). The regulations and recommendations passed relating to E. Coli show the level of risk it poses to public health.

Table 1. U.S. E.P.A recreational water recommendations

Criteria	Recommendation 1		Recommendation 2	
Elements	Estimated Illness Rate 36/1,000		Estimated Illness Rate 32/1,000	
Indicator	GM (cfu/100 mL)	STV (cfu/100 mL)	GM (cfu/100 mL)	STV (cfu/100 mL)
Enterococci (marine/fresh)	35	130	30	110
E. Coli (fresh)	126	410	100	320

1.2.1.2 Sources of E. Coli

Contaminated food and water sources are common causes for E. Coli outbreaks. Water sources are often contaminated when exposed to high concentrations of fecal matter. This can come from a variety of sources like polluted wastewater, stormwater, or agricultural runoff. The polluted water finds pathways into rivers, lakes, streams, ponds, and groundwater supplies where it is detrimental to water quality. A 2017 study done on the impact of combined sewer overflow examined this. The study found that as a wastewater plant discharged 4.3 percent of its untreated wastewater into the local river, E. Coli concentrations increased from 3.5×10^0 MPN/100mL to 2.8×10^3 MPN/100mL (Mascher et al. 2021). A different study on a wastewater treatment plant examined the effect of treatment lagoons on the local groundwater wells. It found that E. Coli and total coliforms exceeded drinking limits in most wells (Barakat et al. 2019). Both studies highlight how contaminated water can negatively impact clean sources of water.

Raw foods like salads and undercooked meats are also common causes for E. Coli outbreaks. Vegetables like lettuce and other leafy greens are often contaminated with E. Coli after exposure to runoff or irrigation with fecal contamination. Leafy greens have been connected to E. Coli outbreaks in many instances and have been well documented. A study from 1996 highlighted the problem with E. Coli in the food service industry by examining vegetables and salads. Approximately 14 percent of the tested vegetable salads contained partially pathogenic bacteria (Lin et al. 1996). Meat and poultry are typically cross contaminated by liquid from other meats infected by E. Coli. A 2018 study examined how faulty meat packaging impacted the E. Coli found on different surfaces. It found that more than 60 percent of poultry packages had meat juice on them, the majority

contaminated with E. Coli (Chen et al. 2018). To reduce to risk of E. Coli infection from foods, proper washing and cooking is recommended.

1.2.1.3 Health Risks

Commonly perceived as dangerous to human health, many strains of E. Coli are harmless. However, some strains are pathogenic meaning they cause disease. There are several strains of E. Coli that may have adverse effects on human health like E. coli O157:H7. Common side effects include nausea, stomach problems, or diarrhea. Severe side effects are less common but at-risk populations such as children and the elderly can develop kidney failure that may lead to death (Poolman 2017, Mayo 2020, CDC 2020, WHO 2021).

E. Coli can typically be treated with antibiotics at hospitals. However, this practice has led to long term issues. Widespread use of antibiotics to treat E. Coli cases has led to many strains becoming resistant. This leads to concern over how to effectively treat and inactivate E. Coli in the future. Studies have found high percentages of antibiotic resistant E. Coli strains in supermarket foods, wastewater treatment plants, hospitals, and the environment (Korzeniewska et al. 2013, Rasheed et al. 2014, Olorunmola et al. 2013). The study shows that antibiotic resistant E. Coli are becoming more frequent which may pose future risks to the public.

1.2.2 Conventional Treatment

1.2.2.1 Disinfection

Chlorination is widely used to remove pathogens like E. Coli from water and wastewater systems. It can be administered as chlorine gas, hypochlorite, or other solid

and liquid forms. By oxidizing cellular material, chlorine inactivates dangerous microorganisms in water supplies. Currently, chlorine is one of the most cost-effective disinfection techniques. Relatively inexpensive and easy to transport, many facilities find it suitable for treatment. Chlorine also provides residual treatment in water systems, prolonging disinfection after leaving the treatment facility. A key disadvantage of treatment is the formation of disinfection by products (DBPs). Formed from chlorine reacting with natural organic matter (NOM), and other compounds of interest like bromide and iodide; they can pose serious risks to human health. Some classes of DBPs are regulated, like trihalomethanes (THM) and haloacetic acids (HAA). Other classes exist but are unregulated due to a lack of information on them and the small quantities they occur in. Some DBPs have been associated with increased risks of different types of cancer in humans. Chlorine itself is toxic to many aquatic organisms even at low concentrations and therefore cannot be discharged into natural water systems.

Ultraviolet light (UV) is another common disinfection technique, with comparable operation costs and disinfection effectiveness as chlorine. Ultraviolet radiation emitted by mercury arc lamps damage organisms by penetrating the cell wall, disabling the ability to reproduce. Disinfection effectiveness is dependent on characteristics of the water being treated. Water with high levels of suspended solids (SS) or turbidity may see lower reduction in organisms being targeted. This treatment is a physical process rather than chemical so handling and storage of toxic chemicals like chlorine is eliminated. This also eliminates DBP formation, but UV light is unable to provide residual disinfection in distribution systems.

Ozone naturally exists in the Earth's atmosphere but can be difficult to produce due to it being an unstable gas. It acts as a strong oxidant in water and wastewater treatment and is effective in destroying viruses and bacteria. When treated with ozone, direct oxidation of the organism's cell wall occurs which effectively destroys them. Ozone is less common in treatment facilities due to its relatively high operation costs. Technology associated with ozonation can be complex and ozone must be created on site due to its instability. Operation of ozone equipment is energy and maintenance intensive. Therefore, many facilities opt for traditional chlorination or UV light for disinfection processes.

1.2.2.2 Retention Ponds

Traditionally viewed as a "low tech" treatment option, retention ponds have consistently proved effective in removing high percentages of bacteria. The efficiency of bacteria inactivation is closely tied to many factors including but not limited to temperature, pond depth, environmental conditions, dissolved oxygen, turbidity, and pH. General removal of coliforms in retention ponds often exceed 90 percent when designed properly (Struck et al. 2006).

While many factors impact the overall removal, it is thought the driving mechanism is sunlight-mediated disinfection (Passos, Dias, and Sperling. 2020) This is a similar process used in UV treatment but does not require energy or costs to operate. With sunlight as the disinfectant, turbidity negatively affects removal by limiting light penetration. This limits the removal in stormwater, as influent runoff often contains high concentrations of NOM and TOC. Retention ponds only release water through evaporation and infiltration, therefore having an infinite detention time. The drawback of

this is retention ponds are therefore limited in their capacity and may overflow during large storm events.

1.2.2.3 Detention Pond

Detention ponds are like retention ponds in that they use the same mechanisms to treat introduced water. However, detention ponds have a limited detention time. The water released from detention ponds may have slightly higher quality than the influent, but treatment is limited compared to retention ponds. Rather, detention ponds are designed to limit the sudden quantity of stormwater entering receiving waters. Despite minor treatment of the water being received, detention ponds serve to reduce sudden pollution and flooding from stormwater into water systems.

1.2.2.4 Wetlands

Wetlands refer to a distinct ecosystem that is characterized by saturated land, seasonally or year-round. They are often categorized by type, including marshes, swamps, bogs, or fens (U.S. EPA 1990). Constructed wetlands are artificially designed to replicate these ecosystems and are used as SMPs. It is difficult to determine the exact efficiency of wetlands in removing bacteria due to their complexity and biodiversity. For example, a 2-year pilot-scale study using a horizontal flow constructed wetland removed 3.44-3.74 log units of E. Coli from the primary effluent of a wastewater system (Zurita and Carreon-Alvarez. 2015). Many studies have found similarly high removal exceeding 90 percent removal of E. Coli (Karim, Glenn, and Gerba. 2008. De Amorim et al. 2019. Abunaser and Arwa. 2020). However, some wetland studies have indicated significantly lower E. coli removal potential. One such study only saw a 6.7 percent removal rate of E.

Coli (Lamori et al. 2019). The discrepancies between studies are likely due to varying wetland natures, with different plant, soil, and animal life existing in each.

1.2.3 Emerging Technology

1.2.3.1 Biofiltration

Biofiltration refers to using a bioreactor containing living organisms to remove pollutants from water. It is frequently used in water and wastewater treatment by employing microorganisms to remove target pollutants. Biofiltration is not limited to microorganisms and can be applied to larger organisms as well. Recent developments have been made using plant and aquatic life as effective E. Coli removal tools in small scale studies. In 2018, a study examining the biofiltration capabilities of *Corbicula Fluminea* showed high removal capabilities for E. Coli, heavy metals, and other common contaminants in stormwater. After 48 hours, there was zero E. Coli detected using initial concentrations of 1000 CFU/ mL. The clams only contained 1-2% of the initial E. Coli in their soft tissue, indicating bioprocessing as the main removal mechanism. E. Coli removal increased exponentially with the number of clams in the stormwater (Gomes et al. 2018). Similar conclusions were found in a 2016 study on zebra mussels. After 24 hours, E. Coli was completely removed from sample volumes (Mezzanotte et al. 2016).

Plant species have also shown potential in removing E. Coli concentrations from water. A 2017 study tested the biofiltration capability of 17 native Australian species. Nine of these reduced E. Coli populations and inhibited growth. Each plant also reduced nitrogen and other nutrient concentrations in the water (Shirdashtzadeh et al. 2017). These plants could work in tandem with specialized green wall materials to further

reduce E. Coli concentrations. Hydraulically slow media like coir, rockwool, and fyto-foam have shown up to 80 percent E. Coli removal; fast media like perlite, vermiculite, growstone, expended clay, or river sand showed up to 20 percent (Prodanovic et al. 2017). Used properly, biofilters would be an ecologically friendly approach to reduce E. Coli concentrations in stormwater.

1.2.3.2 Biochar

Biochar is pyrolyzed biomass, a charcoal-like material, that is that is produced from organic materials by decomposing them under high temperatures. Physically biochar has high porosity, surface area, and water holding capacity. Relatively inexpensive to produce, it is often added to soils or containers to increase adsorption capacity for targeted constituents. Recent studies have been performed examining its use in E. Coli removal. In 2014, a study amended sand filters at 5% by weight with biochar and found this to increase the E. Coli removal rate. Influent with high Natural Organic Matter (NOM) concentrations introduced into the sand filter reduced the removal efficiency. But even with these reductions the biochar amended filter still performed better than the sand filter alone (Mohanty et al. 2014). Biofilms form naturally through the filtration process in biochar amended filters and these can enhance or impair the removal potential of the filter. Like the introduction of high NOM concentrations, biofilms often decrease the performance of biochar amended filters but still see higher removal than solely sand filtration (Mohanty et al. 2014, Afrooz and Boehm 2016). Over time, amended filtration systems see little physical degradation but decrease in removal capacity. Intermittent rain events are thought to change the biofilm inside the filter, reducing capacity in the long term (Mohanty et al. 2014, Kranner et al. 2019). Restricted

long-term use limits the efficacy of biochar amended filters, but they demonstrate high E. Coli removal potential under ideal conditions.

1.2.3.3 Coated Sands

Slow sand filtration was one of the first technologies used to effectively remove pathogenic bacteria from surface water. Removal is highly dependent on the composition and grain size of the sand, but quartz and silica sands are often used. Sand filters demonstrate a wide range of removal capabilities, removing high percentages of NOM, metals, TOC, pathogens, and more. Under many conditions sand filtration is highly effective in removing E. Coli, even in the presence of other pollutants (Fernandez 2019). Despite high removal potential, they are prone to fouling and clogging which may diminish adsorption capacity and requires either backwash or replacement of the filter. To improve the bacteria removal potential of sand filters under a wider range of influent conditions, other media can be added. Iron coated sands have been examined as a potential addition to sand filters. Studies indicate that the addition of iron coated sands to traditional sand filters increases the removal of E. Coli and other bacteria (Aal et al. 2009, Park et al. 2011, Marik et al. 2019, Kulkarni et al. 2020) This is likely due to the anti-microbial effects demonstrated by iron oxides and iron nanoparticles. The physical coating, disruption of cell membrane, and generation of reactive oxygen species from iron are thought to inactivate many pathogens with relatively short contact times (Diao and Yao 2009, Li et al. 2018, Gabrielyan et al. 2019). Aside from the reasons mentioned above, iron coated sands are thought to remove E. Coli due to their net positive charge. E. Coli is a gram-negative bacterium and therefore attracted to the iron coated sands which ensures their attachment in the filter.

1.2.3.4 Steel Byproducts

Two common byproducts from steel production are steel chips and steel slag, both of which are easily obtained and inexpensive. The application of these materials in removing E. Coli has been researched in recent years. The results from the research done on steel byproducts has shown that steel chips and steel slag have different E. Coli removal potentials. In 2017 a batch scale adsorption study found steel chips to remove 94% of E. Coli over 2 hours with original concentrations of 10^4 MPN/ml. Under the same conditions steel slag removed 28.5% (Hooshyari 2017). High E. Coli removal by the steel chips was also recorded during column studies performed in 2019. These found that the largest size range of chips (4-9mm in diameter) removed 60% of E. Coli and the smallest (0.5-1mm in diameter) removed nearly 100% at an initial concentration of 10^6 MPN/ml (Dai 2019). However, these removal percentages decrease in the presence of higher concentrations of NOM, DOC, and other contaminants. Therefore, actual removal percentages in stormwater are typically lower than those using pure water with E. Coli (Hooshyari 2017, Dai 2019). In 2019, a pilot study using a field filter consisting of a combination of steel slag and steel chips examined the real stormwater removal by steel byproducts. It found that the filter could consistently remove 50% of E. Coli over several different runoff events. One concern highlighted from the field filter was an increase of 0.5 mg/L in dissolved iron content after the filter (Neville, 2019). Overall, steel byproducts appear to be a promising low-cost media that can remove E. Coli from water.

1.3 Research objective

The main objective of this study was to provide a recommendation for future field application of steel byproducts in E. Coli removal in stormwater. Previous studies have shown steel byproducts have potential in removing high percentages of E. Coli from stormwater through lab scale and field scale testing. An evaluation of the long-term performance of steel byproducts was required to recommend future applications of steel byproducts in E. Coli removal in stormwater. To accomplish this, lab scale testing and field scale testing were done. For lab scale testing, batch and column studies were performed to examine the impact of aging and steel byproduct ratio on E. Coli removal. Using this information, field scale testing was performed using varying steel byproduct ratios to reduce agglomeration issues while maintaining E. Coli removal and filter longevity. The ability of the field filter to remove other target contaminants like total nitrogen, nitrate, total phosphorus, orthophosphate, and dissolved iron was also examined.

2. MATERIALS AND METHODS

2.1 Lab study

2.1.1 Introduction

To evaluate the long-term performance of steel chips and steel slag, column studies and batch studies were performed in laboratory. This was done by examining the difference in performance between new and aged materials. Aged steel byproduct material was gathered from a previous field filter site after two years in use. The lab studies examined the removal potential of 2-4 mm in diameter and 4-9 mm in diameter new and aged steel chips and steel slag, for a total of 8 testing materials. E. Coli concentrated water was used to simulate E. Coli concentrations in stormwater. The column studies pushed this water through columns packed with steel byproducts at a designated flow rate. The batch studies contained a beaker with a set amount of E. Coli concentrated water and weight of steel byproduct material. Both studies examined the performance of aged versus new steel byproduct material.

2.1.2 E. Coli preparation

E. Coli was used in the lab studies to simulate varying E. Coli concentration conditions in runoff events. To grow E. Coli a container of 100 mL Luria Broth Base (LB, Thermo Fisher Scientific, 10 g/L peptone, 10 g/L sodium chloride, 5 g/L yeast extract) was made at 25 grams of broth per liter of water. The container was inoculated using concentrated E. Coli from a frozen stock maintained in a -20 °C freezer. A Thermo Scientific MaxQ 4000 Benchtop Orbital Shaker was set at 37 °C for 24 hours at 150 rpm

with the 100 mL container inside. During culturing, a 1L buffer solution was made using 1.0 mM NaHCO_3 (0.84 g/L), 0.1 mM KCL (0.7455 g/L) and 1.2 mM H_2SO_4 (0.49 g/L).

The cultured E. Coli was then evenly separated into three 50 mL centrifuge containers and spun at 4000 rpm at 20 °C for 10 minutes. The containers were then removed, and the liquid phase was discarded. Each was refilled with 30 mL of buffer solution and the E. Coli was re-suspended in the buffer by lightly swirling the containers. They were then reinserted into the centrifuge and this process was repeated a total of three times. After the third time, E. Coli in each container was resuspended with buffer solution and poured into a 100 mL glass flask. To estimate the approximate number of E. Coli cells per mL, a HACH DR/400 OU spectrophotometer was used. The E. Coli cells were measured at UV600. E. Coli concentrations using this process fell within a range of $1.0\text{-}1.6 \times 10^9$ cells/mL on the spectrophotometer. The 100 mL container of E. Coli stock was stored at room temperature in a room with no direct sunlight for up to 5 days.

2.1.3 Laboratory Materials

The steel byproducts used in the experiment were obtained from steel manufacturing and recycling facilities. The steel slag was collected from waste material provided by Nucor Steel in Norfolk, NE. The slag was the byproduct of blast furnaces melting scrap metal and fluxes to create steel. The steel chips were collected from Alter Metal Recycling in Marshall, MN. They are exclusively carbon steel chips and are a byproduct of the physical processes like machining, grinding, and milling of finished steel products at various plants.

Steel slag and steel chips were grouped into different size ranges and physical conditions. This included: 2-4 mm new, 2-4 mm aged, 4-9 mm new, and 4-9 mm aged. Figure 1 and Figure 2 show the new and aged steel byproducts after sorting them according to their size and respectively material. To group the new materials into the proper size range, sieves were used. Raw materials were sieved into 0.5-1 mm, 1-2 mm, 2-4 mm, and 4-9 mm groups; the 0.5-1 mm and 1-2 mm groups were discarded. This was done for the new steel chips and new steel slag separately. Once separated, the new steel chips were then cleaned to remove excess oils and chemicals used in the manufacturing process. They were rinsed with nanopure water and washed in a phosphorus free soap bath for 24 hours. After 24 hours they were again rinsed with nanopure water and left out to dry for 24 hours. The new steel slag was rinsed with nanopure water and dried for 24 hours to remove attached fine particles; additional cleaning was deemed unnecessary.

Aged materials were gathered from an existing field filter created by SDSU students Blake Jorgensen and Dr. Abdoul Kouanda after two years in use. The existing filter was a mixture of 50% steel slag and 50% steel chips. Approximately 10 gallons of material was removed from the filter. Sieves were used to separate the mixed materials into different size groups. After sizing the materials, chips and slag were separated by hand by examining the physical properties of each piece. All materials were then rinsed thoroughly with nanopure water to remove dirt and other particles loosely attached to their surfaces.



Figure 1. New and aged steel slag materials.

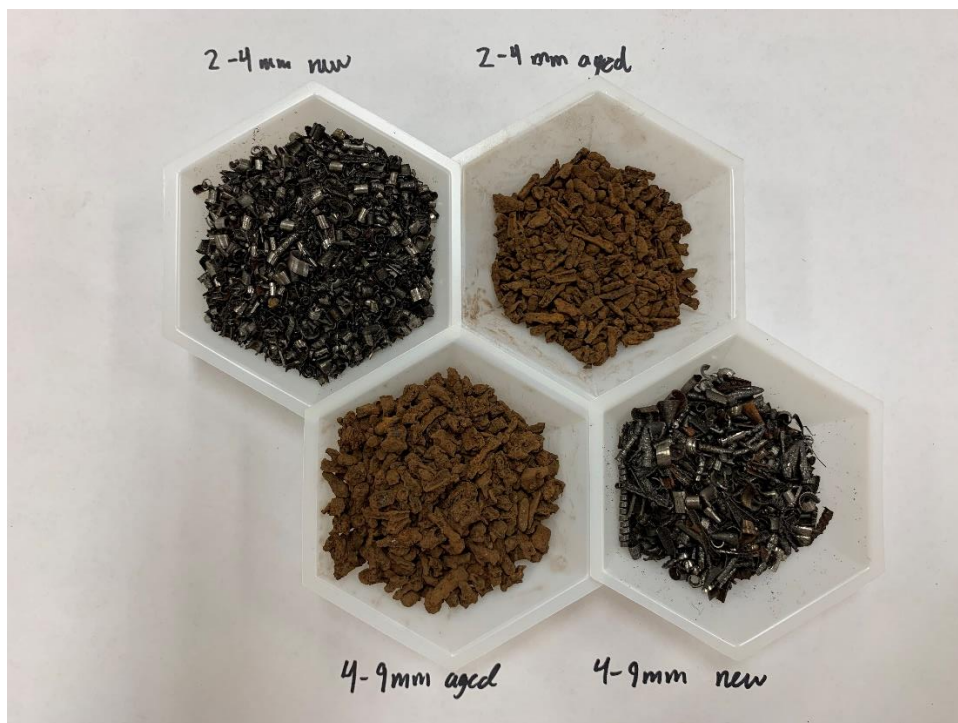


Figure 2. New and aged steel chip materials

2.1.4 Batch Study

Each of the 8 tested material's E. Coli removal capabilities were examined by placing one gram of material in 8 separate 250 mL Erlenmeyer flasks. E. Coli was prepared using the procedure outlined in the E. Coli preparation section. Flasks containing steel slag were given initial E. Coli concentrations of 10^4 MPN/mL by diluting the prepared E. Coli stock. Those containing steel chips were given a concentration of 10^7 MPN/mL. Two blanks were prepared containing no material but given concentrations of 10^4 MPN/mL and 10^7 MPN/mL. All flasks were filled with 100 mL of E. Coli spiked nanopure water. After flask preparation, each was placed on a Thermo Scientific™ MaxQ™ 4000 Benchtop Orbital Shaker set to 20 °C at 100 rpm for 24 hours. This setup can be seen on Figure 3. One mL of sample was taken from each of the 10 flasks at times of half hour, one hour, two hours, three hours, 12 hours, and 24 hours to determine E. Coli Concentrations using the IDEXX method outlined in this paper.

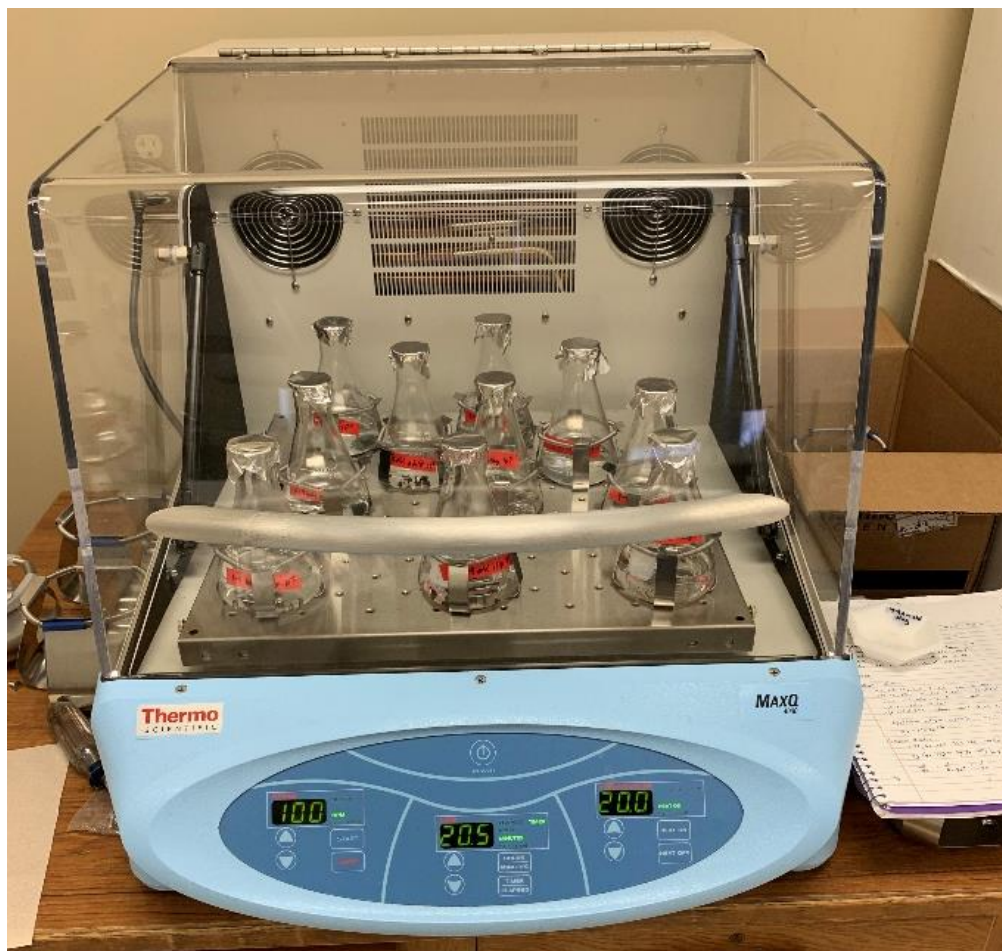


Figure 3. Thermo Scientific™ MaxQ™ 4000 Benchtop Orbital Shaker setup used for batch study and kinetics testing.

2.1.5 Column studies

Steel byproduct E. Coli removal capabilities were examined by pushing water containing fixed E. Coli concentrations through columns with packed material. The experiments were conducted using Omnifit® fixed-bed glass columns with a 15 cm height and 1.5 cm inner diameter. Three column experiments were performed: exclusively steel slag, exclusively steel chips, and a mixture of both. To pack the materials, approximately 1 cm of height was added and then the side of the column was

gently tapped to settle them. This was done to prevent any breaking of the material through normal compaction efforts, as this would reduce the material size. All column experiments used a flow rate of 1.18 mL/min to achieve an EBCT (Empty bed contact time) of 15 minutes. The EBCT is the ratio of bed volume to flow rate as shown:

$$\text{EBCT} = \frac{\text{Bed Volume (mL)}}{\text{Flow } Q \left(\frac{\text{mL}}{\text{min}}\right)}$$

Two Masterflex® L/S® peristaltic pumps were used to achieve $\pm .02$ mL/min of the desired flow rate. To check the actual flow rate in the pumps, quantities of water from each were weighed after 10 minutes of flow to determine the actual mL/min rate. Flow rates were then adjusted based on this by tightening or loosening the tubing.

E. Coli was prepared and measured as mentioned in the E. Coli preparation and IDEXX sections of this paper. To prepare adequate quantities of E. Coli spiked influent water at the given flowrate, 5 gallon glass “carboy” containers were used. Each was filled with 18 liters of nanopure water and then made into a buffer solution using 1.0 mM NaHCO_3 (0.84g/L), 0.1 mM KCL (0.7455g/L) and 1.2 mM H_2SO_4 (0.49g/L). E. Coli stock was added until the desired concentration was achieved after the buffer chemicals were added. To sample the influent and effluent, 20 mL was taken in small glass vials starting at the beginning of the sampling period. Influent was poured directly from the “carboy” container and effluent was taken from the ends of the plastic tubing after the columns. Figure 4 shows how the column experiment was set up for each of the column studies performed.



Figure 4. Reactor setup for column experiments.

Table 1. Characteristics of steel slag in the column study

Material	Size	Porosity (%)	Packing Density (g/cm ³)	Particle Density (g/cm ³)
Aged	2-4mm	0.47	1.74	3.29
	4-9mm	0.51	1.56	3.17
New	2-4mm	0.49	1.50	2.95
	4-9mm	0.52	1.32	2.76

Table 2. Characteristics of steel chips in the column study

Material	Size	Porosity (%)	Packing Density (g/cm ³)	Particle Density (g/cm ³)
Aged	2-4mm	0.69	1.10	3.51
	4-9mm	0.70	1.00	3.32
New	2-4mm	0.70	1.48	4.97
	4-9mm	0.78	1.04	4.64

Table 3. Characteristics of the steel byproducts mixture in the column study

Material	Steel chip percentage (%)	Porosity (%)	Packing Density (g/cm ³)	Particle Density (g/cm ³)
Aged	5	0.49	1.73	3.38
	10	0.51	1.67	3.42
New	20	0.54	1.60	3.46
	50	0.58	1.56	3.71

2.2 Field Study

2.2.1 Filter materials

Two types of steel byproducts were used, steel chips and steel slag. Both are a result of the steel manufacturing process in steel mills or manufacturing shops. Slag refers to the leftover impurities from the molten steel in the melting process. The composition of the slag is dependent on the type of steel being manufactured but typically consists of carbon, silicon, manganese, phosphorus, iron, lime, and dolomite. The steel slag was obtained from Nucor Steel in Norfolk Nebraska. Chips refer to the excess material from the manufacturing of steel. Formed from physical processes like cutting and shaping steel into finished products. The steel chips were obtained from Alter Metal Recycling in Marshall Minnesota and came from carbon steel.

2.2.2 2019-2020 Field Study

The field filter was installed for a previous study in 2018 (Neville 2018). It is in a residential area near Camelot Intermediate School in Brookings, South Dakota at the intersection of Camelot and Breckenridge Drive. The dimensions were designed to be 5 feet wide, 6 feet long, and 8 inches in height. These were chosen to fit the retention pond inlet. The field filter fitted to the inlet can be seen on Figure 5. A wider view of the site conditions for the field filter can be seen on Figures 6 and 7. The base structure was created by Bend Rite Custom Fabrication Inc and was made of A36 ¼ inch mild steel. Originally the filter contained 25% large steel slag (4-9mm), 12.5% small steel slag (2-4mm), 50% large steel chips (4-9mm), and 12.5% small steel chips (2-4mm). With this

percentage, the filter had to be remediated following each rain event due to agglomeration of the rusting materials.

To address the agglomeration issues the 2019 and 2020 field filters were reduced to 50% steel chips and 50% steel slag. All existing material was removed from the 2018 field filter and solely new material was added in 2019. The 2020 field filter continued to use the same materials from the 2019 field filter, no materials were taken or added during this timeframe. During rain events water samples were collected using clean 1 L plastic bottles for both the influent and effluent. Influent samples were collected a foot above the filter in the pond inlet just below the surface level of the stormwater. Effluent samples were collected in the center of the backend of the filter by holding the plastic bottle flush to the flow holes. In total four rain events with at least five sampling points were collected during the late spring to early fall months for each of the two filter years. Key water quality parameters tested include E. Coli, total phosphorus, orthophosphate, nitrogen, nitrate, and dissolved iron.



Figure 5. Pilot scale steel byproduct filter May 2020.



Figure 6. Site location facing Southwest direction.



Figure 7. Site location facing the Northeast direction.

2.2.3 2021 Field Study

To conduct research on the removal potential of aged materials used in the field filter and to reduce filter agglomeration, small quantities were taken in August of 2020 to perform lab studies. Based on these, the new ratios for the field filter were chosen to be 30% steel chips and 70% steel slag. Both categories consisting of approximately 50% 2-4 mm and 50% 4-9 mm sized pieces. A higher ratio of slag to chips was desired to counteract the agglomeration seen in previous studies. To create the new filter, all-aged materials were removed from the original filter using pickaxes and shovels. New slag was added to the filter to reduce the existing steel chips to slag ratio. Old and new material was added in layers and then mixed in the filter to achieve the desired ratio. Figures 8, 9, 10, and 11 show the different steps of the filter redesign in May of 2021. Sampling was done in the same manner done in 2019 and 2020. The different water quality parameters

examined were also the same including E. Coli, total phosphorus, orthophosphate, total nitrogen, nitrate, and dissolved iron. Duplicates for samples were done for the first and last samples of each storm event to ensure accurate testing methodology. All duplicates were within 15 percent of initial results.



Figure 8. Process of adjusting the steel byproduct ratio for the pilot scale steel byproduct filter.



Figure 9. Process of adjusting the steel byproduct ratio for the pilot scale steel byproduct filter.



Figure 10. Adjustment of steel byproduct ratio in field filter.



Figure 11. Pilot scale steel byproduct filter after ratio adjustment.

Table 4. 2021 Runoff event characteristics

	5/25/2021	7/10/2021	7/11/2021	7/25/2021
Start Time	8:30PM	9:00AM	11:00AM	10:45PM
End Time	9:30PM	11:15AM	12:30PM	12:00PM
Temperature (°F)	62	64	65	74
Total Rainfall (in)	0.8	1.7	1.2	0.6
Sampling Interval (min)	10	10	15	10

2.3 Analytical Methods

2.3.1 IDEXX

Accurate E. Coli concentrations were determined using the Coli-ert 18 method, giving MPN (most probable number)/mL. Samples being tested were diluted enough to fall within an expected range of 100 to 2000 MPN/ 100 mL. The dilutions were then added to 100 mL containers provided by IDEXX and one packet of Colilert reagent was added to each. Containers were then capped and rotated until the Colilert reagent had dissolved in the diluted sample. All contents were then added to a Quanti-Tray*2000 and sealed using an IDEXX Quanti-Tray*sealer. Any prepared Quanti-trays* were placed in an oven at 35 ± 0.5 °C for 24 hours. Trays were read out using blacklight in a dark room and using calculation instructions provided by IDEXX.

2.3.2 Colorimetric methods

Five water quality parameters were tested to examine the quality of influent and effluent stormwater from the field filters. Dissolved iron, total phosphorus, nitrate, orthophosphate, and total nitrogen were measured using Hach Company colorimetric methods. This was done using a DR/4000U Spectrometer manufactured by Hach Company. A 0.45 um filter was used to filter stormwater samples prior to measuring each of the five water parameters. Filtration removed constituents in the water that could interfere with the colorimetric results. Dissolved iron was measured using the FerroVer® Method (Hach 2014). Total phosphorus was measured using the PhosVer®3 with Avid Persulfate Digestion Method (Hach 2017). Nitrate was measured using the Cadmium Reduction Method (Hach 2014). Orthophosphate was measured using the PhosVer3®

(Ascorbic Acid) Method (Hach 2017). Total Nitrogen was measured using the Persulfate Digestion Method (Hach 2014).

2.3.3 Cleaning and Sterilization

All materials used during the preparation of E. Coli stock were cleaned using an autoclave. Outside of E. Coli preparation different cleaning procedures were used for equipment and glassware. All dishes were submerged in mixture of 1/3 phosphate free soap and 2/3 nanopure water bath for 24 hours. Glassware was removed and rinsed with reverse osmosis water, resubmerged in a 25% sulfuric acid bath for an additional 24 hours, later rinsed with 18M Ω nanopure water, and set out to dry. Plastic containers did not receive an acid bath and were instead rinsed with 18M Ω nanopure water after the soap bath and dried. All surfaces were cleaned using Steris Coverage Spray HB plus, prior to and after using, to avoid any contamination in laboratory.

3. RESULTS AND DISCUSSION

3.1 Batch study

The results from the general batch study can be found on Figures 12 and 13. These show the E. Coli concentrations over a 24-hour period. Duplicate samples were taken for influent and effluent samples. All were within 15 percent of initial samples but are not represented on the figures seen. The steel chips removed higher percentages of E. Coli, with an influent concentration several orders higher than that of the steel slag. In 24 hours both sizes of new chips effectively removed 100.00 percent of E. Coli and both sizes of aged chips removed 99.70 percent of E. Coli. Seen on Figure 12 the new chips reach a high removal percentage more rapidly, both reaching 99 percent removal within 3 hours. This is due to the rapid oxidation of the new chips. Seen in Figure 14, the Erlenmeyer flasks containing the new chips turn a brown and orange color. The interaction between the ferric oxide created through rusting and the E. Coli disrupts the E. Coli cell walls and their ability to reproduce. Studies examining the bactericidal properties of ferric oxide nanoparticles have concluded that they are able to effectively deactivate many strains of bacteria including E. Coli (Li et al. 2018, Gabrielyan et al. 2019, Diao and Yao 2009). The aged steel slag still removes high percentages of the E. Coli through adsorption. The negatively charged E. Coli attach themselves to the positively charged surface of the steel slag where they are effectively removed.

The slag removed lower percentages of E. Coli at an influent concentration of approximately 10^4 MPN/ml. The 2-4 mm aged, 4-9 mm aged, 2-4 mm new, and 4-9 mm new removed 73.2, 71.6, 58.7, and 46.3 percent respectively. The result for the steel slag batch study can be seen on Figure 14. Unlike the steel chips, the aged material performed

better than the new material. This can again be attributed to the interaction of the ferric oxides with the E. Coli. The aged material has ferric oxide attached to its outside, likely from interaction with the steel chips during the time in the field filter. The new steel slag does not oxidize over the 24-hour period, limiting its removal mechanism to solely adsorption.

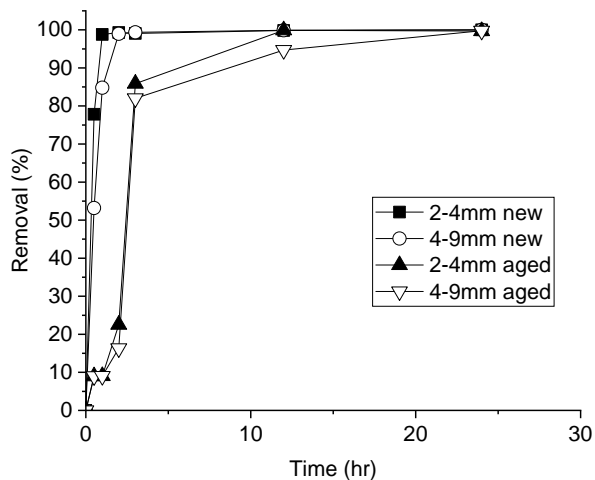


Figure 12. Steel chips batch study results.

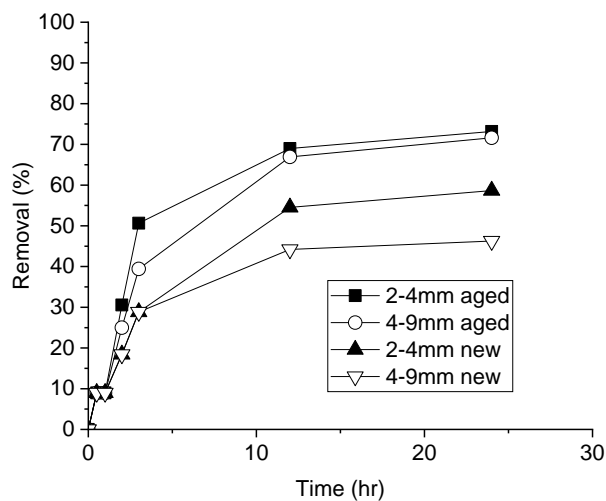


Figure 13. Steel slag batch study results.

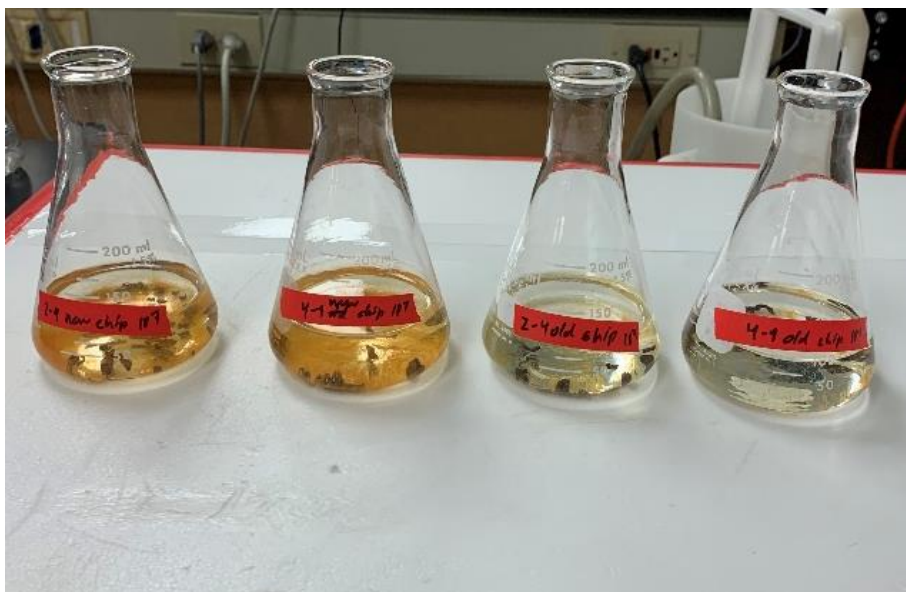


Figure 14. Erlenmeyer flasks with 1 gram of material after 24 hours in the Thermo Scientific™ MaxQ™ 4000 Benchtop Orbital Shaker (from left to right 2-4mm new chips, 4-9mm new chips, 2-4mm old chips, 4-9mm old chips).

3.2 Steel chips

New steel chips removed a higher percentage of *E. Coli* than the aged steel chips during the column study. The 2-4 mm new, 4-9 mm new, 2-4 mm old, and 4-9 mm old removed 80, 50, 30, and 10 percent of *E. Coli* respectively. The results for the steel chip column study over the 8-day sampling period can be seen on Figure 15. Higher removals are seen within the first hours of the experiment, but removal capacity stabilizes after day two. With increasing media size, removal rates decreased for both the old and new chips. Smaller particles have larger surface areas for the volume they occupy which results in both greater adsorption capacity and oxidation potential. Similar studies found that small chips (1-2 mm diameter) can removal more than 90 percent of *E. Coli* (Initial *E. coli* concentration = 1.0×10^4 MPN/mL; EBCT=10 min) (Hooshyari 2017) and that 2-4 mm

and 4-8 mm steel chips remove 70% and 60% of initial E. Coli concentrations (Initial E. coli concentration = 1.0×10^6 MPN/mL; EBCT=10 min) (Dai 2019). Duplicate samples were taken for some influent and effluent samples to ensure accurate results. All were within 20 percent of initial samples but are not represented on the figures seen.

Adsorption and oxidation are likely the two processes that remove E. Coli from influent water in this study. Adsorption is the physical process that occurs when the E. Coli attaches to the surface of the steel chips. The negatively charged E. Coli are attracted by the positively charged iron in the steel chips, resulting in surface attachment. This is likely the major removal mechanism for the aged material as it has less material available to oxidize. The difference in removal potential between the new and aged chips is likely due to the oxidation of the new. The bottom section of the new chips undergoes a color change and becomes visibly more brownish orange. It is likely that the oxidation process removes E. Coli more effectively than adsorption in this situation. Studies have shown that iron oxide nanoparticles can exhibit bactericidal properties. These nanoparticles not only inhibit the E. Coli growth process but also cause direct bacterial cell death (Diao and Maosheng 2009, Li et al. 2018, Gabrielyan et al. 2019). The significantly higher E. Coli removal rate of the new chips is likely due to the initial oxidation of the material.

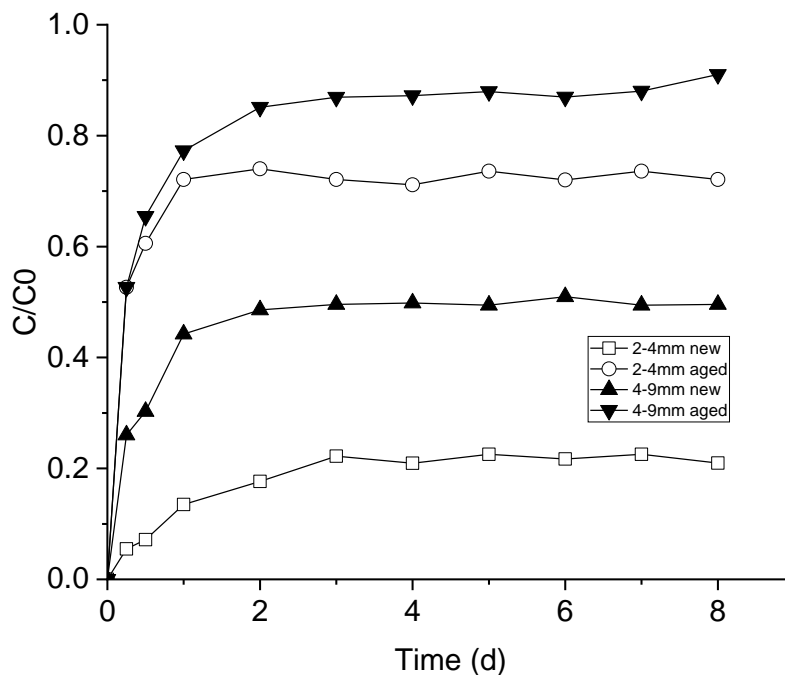


Figure 15. E. Coli removal by steel chips in column study. Experiment conditions: flow velocity= 1.18 mL/min, EBCT=15 minutes.

3.3 Steel slag

The steel slag demonstrated the opposite of the findings for the steel chips; in that the aged material performed better than the new. Duplicate samples were taken for some influent and effluent samples to ensure accurate results. All were within 20 percent of initial samples but are not represented on the figures seen. Column studies performed showed the 2-4 mm new, 4-9 mm new, 2-4 mm old, and 4-9 mm old removed an average of 17, 13, 25, and 21 percent of E. Coli respectively. Results from the steel slag column study can be seen on Figure 16. Based on the trends seen, both the new and aged steel slag continue to lose removal potential over the 8-day sampling period; never reaching a

plateau like the steel chips. Slag shows lower removal rates than that of chips, likely due to its inability to oxidize in the columns. This is because slag has a lower iron content.

Previous studies have shown that adsorption is the major removal mechanism used by slag in removing particles (Hooshyari 2017, Dai 2019, Neville 2019). The aged slag removes higher percentages of E. Coli because it also uses the interaction of ferric oxide particles to remove E. Coli. Ferric oxide particles are also more prevalent on the outside of the aged slag from its interaction with the steel chips. As explained previously, iron oxide particles have demonstrated bactericidal properties in some studies. The difference between the E. Coli removal for aged and new material is not drastic, but the column studies indicate that as slag ages its E. Coli removal capacity increases.

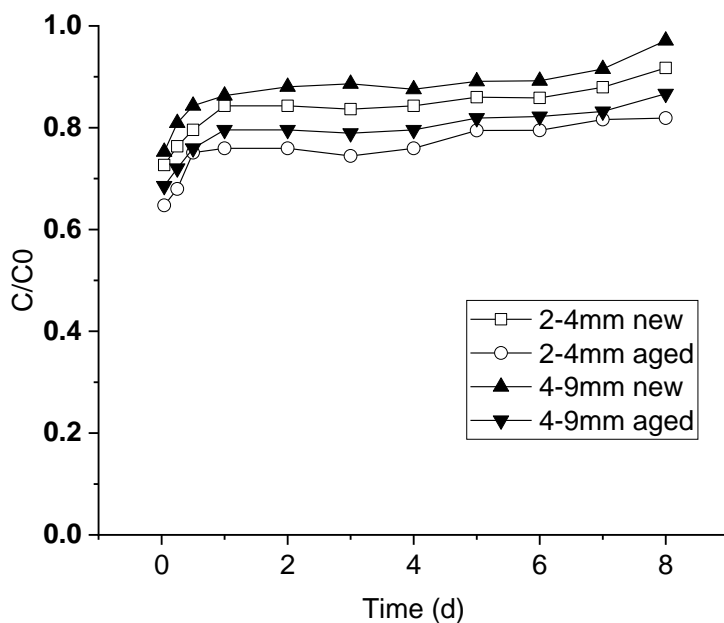


Figure 16. E. Coli removal by steel slag in column study. Experiment conditions: flow velocity= 1.18 mL/min, EBCT=15 minutes.

3.4 Steel slag and steel chip ratios

Figure 17 shows the E. coli removal of different mixtures of steel slag and chips in a column study. A lower ratio of chips to slag was desired to help with agglomeration problems in the pilot study from previous years. The 5, 10, 20, and 50 percent steel chips removed an average of 35, 49, 57, and 62 percent of E. Coli over a span of 8 days. These results indicate that even low quantities of chips present in the mixture can significantly improve E. Coli removal. Figure 18 shows the mixed materials prior to being placed in the columns. As seen, the lower ratios of 5% and 10% have fewer steel chip pieces. Despite this, the E. Coli removal is increased from solely slag being present. This is likely due to the rapid oxidation of steel chips. A color change to a brownish orange is seen in each of the four different ratios.

A lower ratio of chips to slag would help with filter agglomeration problems without significantly reducing performance. After wet and dry periods during the 2019 and 2020 the media would form large masses, resulting in reduced flow rates through the filter. Filter remediation was effective but took significant effort. Overall, the results from the mixture column study showed that even low steel chip percentages present can improve E. Coli removal.

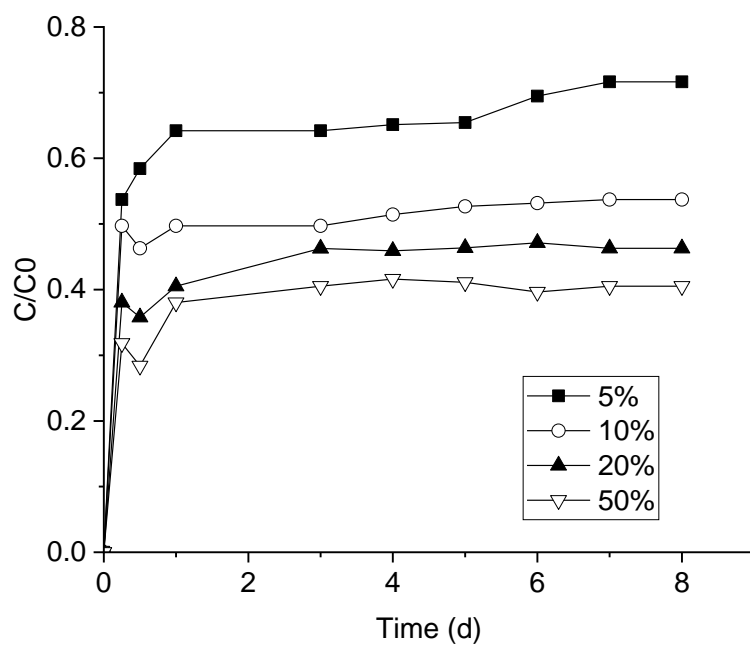


Figure 17. E. Coli removal by steel slag and steel chip mixtures in column study.

Experiment conditions: flow velocity= 1.18 mL/min, EBCT=15 minutes.

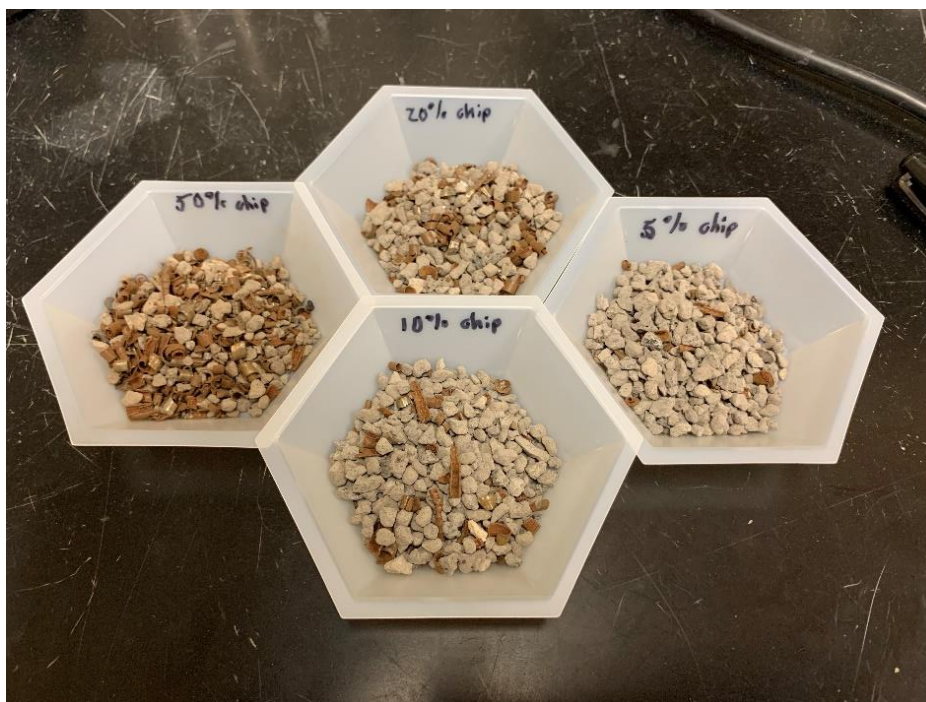


Figure 18. Varying ratios of steel byproducts.

3.5 SEM analysis

The surface structure of the new and aged steel chips and slag were observed using Scanning Electron Microscope (SEM) analysis in the South Dakota State University Daktronics building SEM laboratory. Figures 19, 20, and 21 show these SEM images at varying settings. Each figure illustrates the difference between the new and aged material. The steel chip's surface is originally smooth with few deformations but after two years it more closely resembles the surface of the steel slag. This explains the decrease in E. Coli removal potential seen in the aged chips. As they age, their removal mechanism likely shifts from the interaction of oxide species with E. Coli to more adsorption. Oxidation of the iron in the chips is likely plays a significant role in removing E. Coli from the stormwater. The slag's surface also becomes worn after two years, but not to the same extent as the chips. Deterioration of the slag surface may result in similar or increased removal of E. Coli due to an increase in the cavities at the surface as well as newly attached oxide species. The SEM images taken further prove the findings of the lab testing done.

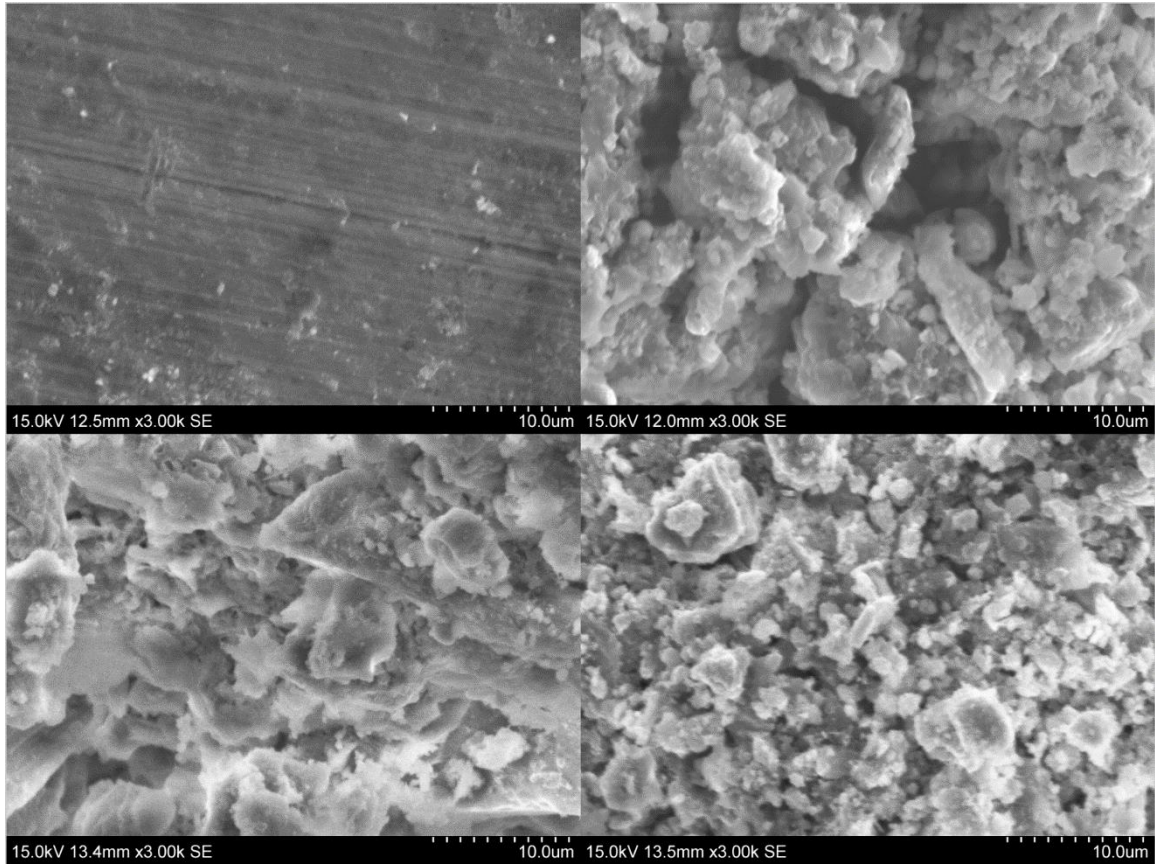


Figure 19. Images of steel byproduct surfaces using a scanning electron microscope at 10um. (Top left new chips, top right old chips, bottom left new slag, bottom right old slag)

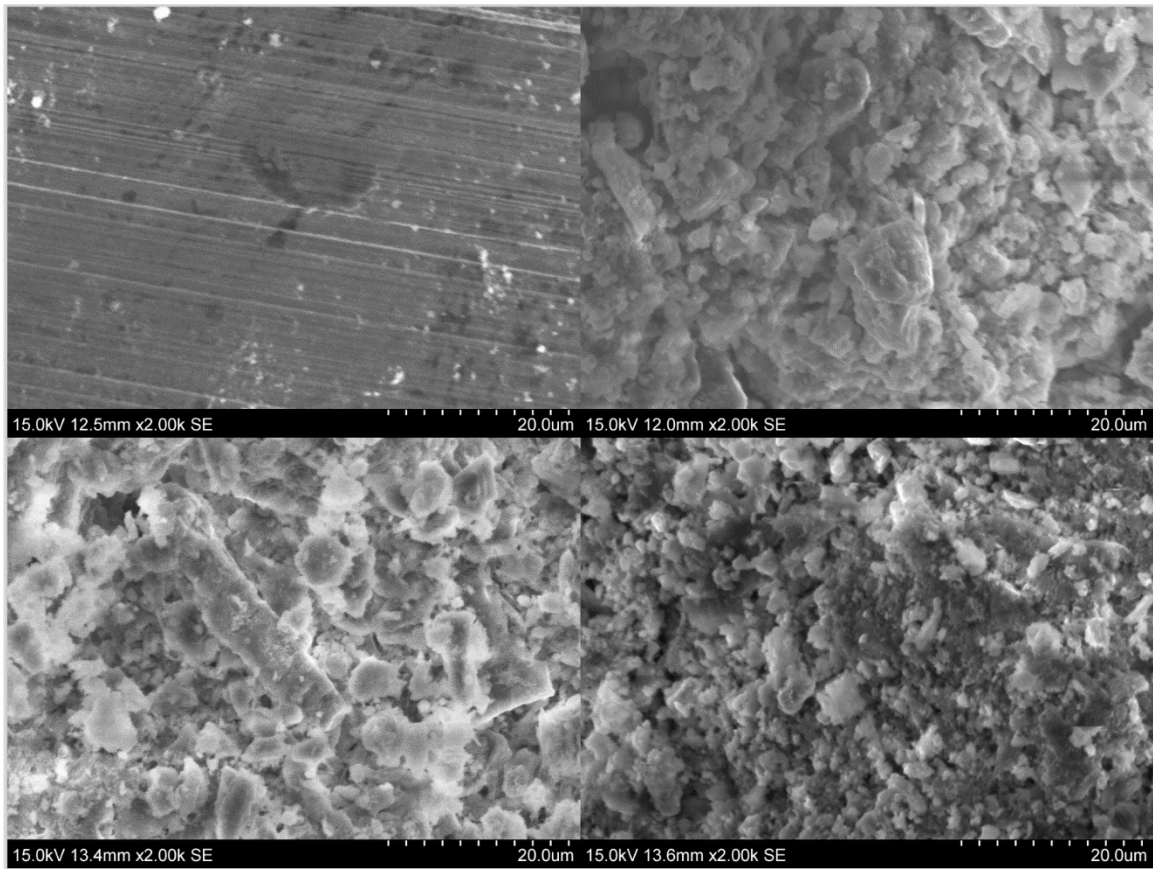


Figure 20. Images of steel byproduct surfaces using a scanning electron microscope at 20um. (Top left new chips, top right old chips, bottom left new slag, bottom right old slag)

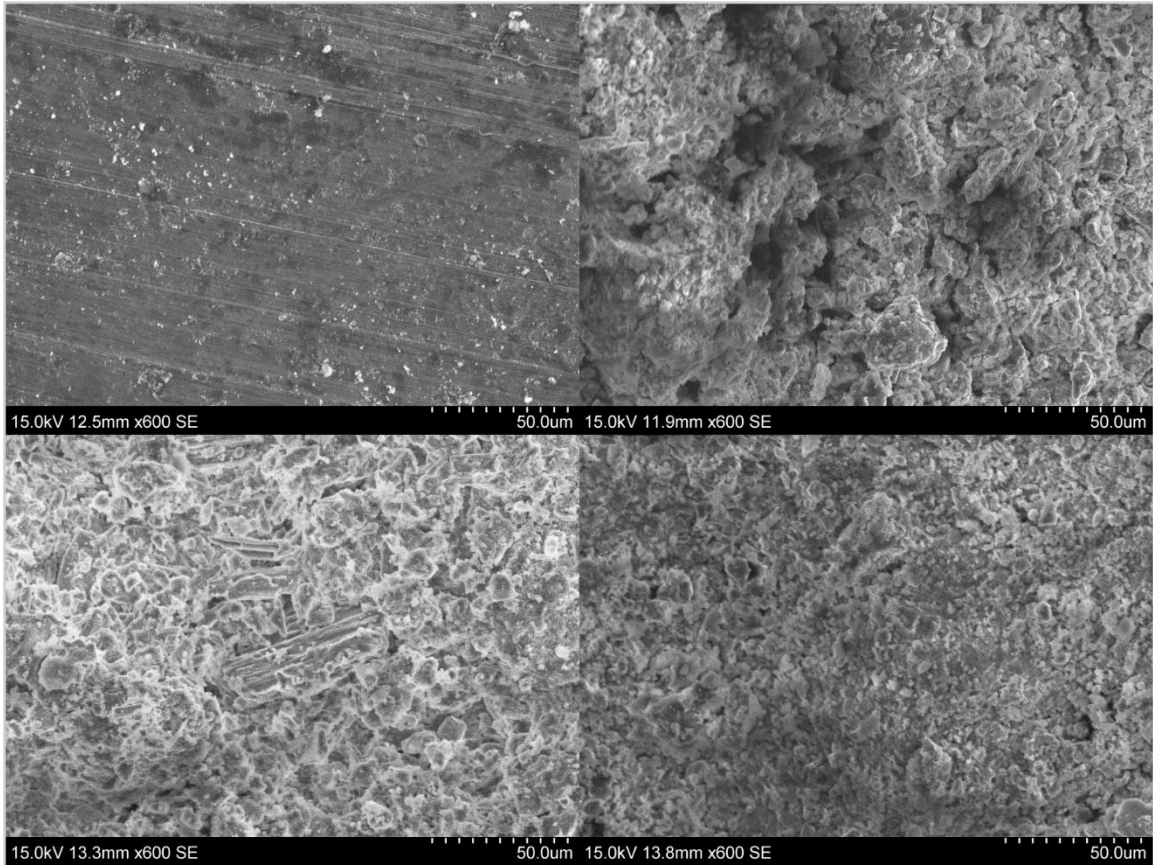


Figure 21. Images of steel byproduct surfaces using a scanning electron microscope at 50um. (Top left new chips, top right old chips, bottom left new slag, bottom right old slag)

3.6 Pilot study

3.6.1 2019 Field Filter

The 2019 field filter was designed with 50% steel slag and 50% steel chips. The field filter was able to remove an average of 53% of E. Coli. Figure 22 shows the influent and effluent concentrations from the 2019 storm events. Influent concentrations in the stormwater ranged from 1101 MPN/100ml to 15531 MPN/100ml. The results indicate that higher influent concentrations see better removal rates than low concentrations. For example, storm event 1 removed an average of 74% of E. Coli while storm event 4 removed an average of 39%. Storm event 1 had a significantly higher average influent E. Coli concentration of 11302 MPN/100mL while storm event 4 had an average of 4644 MPN/100mL. While high influent concentrations experienced higher removal rates, their effluent concentrations were still larger than that of low influent concentrations. Similar trends and results were found on a 2018 pilot scale study on steel byproducts. The filter was comprised of 65% chips and 35% slag and removed an average of 50% of E. Coli (Neville 2019).

The column studies in the laboratory indicate that removal should have been higher than 53%. Lower removal percentages in the field can be attributed to several factors including NOM, E. Coli concentrations, and temperature. Previous studies have examined how these impact E. Coli removal when using steel byproducts. High concentrations of NOM co-loaded with E. Coli have been shown to significantly reduce removal rates by competing for adsorption surfaces (Dai 2019). High temperature conditions have shown to traditionally remove higher amounts of E. Coli in lab scale testing (Dai 2019). As water temperature increases, the viscosity of water increases. The

activity of *E. Coli* also increases at higher temperatures. These factors in combination result in more movement of the *E. Coli.*, attaching more *E. Coli* to the steel byproduct surfaces. The air temperature during most rain events was close to 60 degrees Fahrenheit, whereas room temperature in laboratory is close to 70 degrees Fahrenheit.

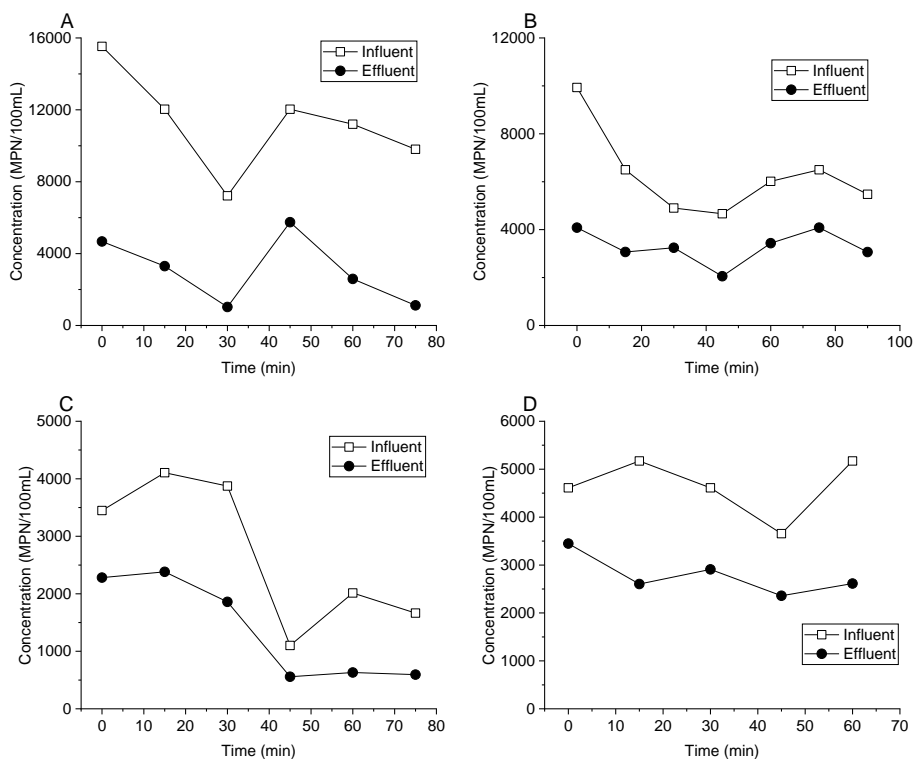


Figure 22. Escherichia Coli concentrations in influent and effluent stormwater versus duration of runoff event. (A) 06/22/2019 (B) 07/20/2019 (C) 08/12/2021 (D) 08/31/2019

There was an average increase of 0.076 mg/L of dissolved iron in the effluent from the field filter. Figure 23 shows the influent and effluent concentrations of dissolved iron for the sampled storm events. The highest effluent concentration was 0.312 mg/L which is well below the EPA recommendation of 1.0 mg/L for recreational water usage

(U.S. EPA 1986). This shows that during 2019 the field filter did not leech a significant amount of dissolved iron. Effluent concentrations using the field filter could therefore be safely discharged into recreational waters with no impact on public health or wildlife.

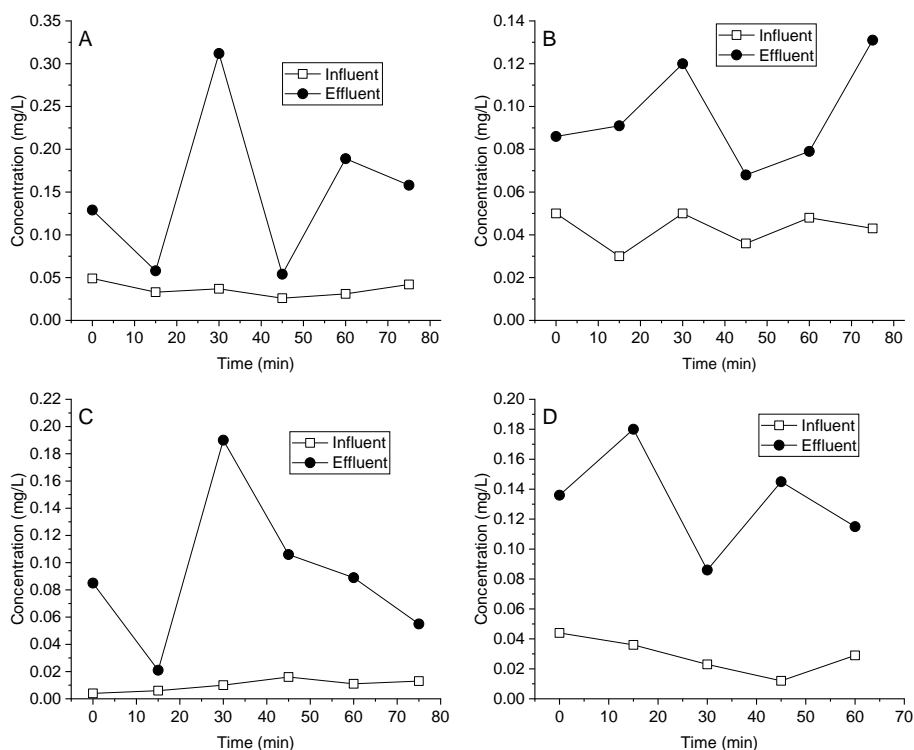


Figure 23. Iron concentrations in influent and effluent stormwater versus duration of runoff event. (A) 06/22/2019 (B) 07/20/2019 (C) 08/12/2021 (D) 08/31/2019

The 2019 field filter removed 48% of total phosphates and 49% of orthophosphates from the influent stormwater. The influent and effluent concentrations for the storm events sampled for total phosphate removal can be found on Figure 24. Influent concentrations ranged from 0.36 to 3.65 mg/L as PO_4^{-3} . Figure 29 shows the influent and effluent orthophosphate concentrations for four storm events. The influent concentrations ranged from 0.08 to 2.12 mg/L as PO_4^{-3} , indicating they comprised a

lower percentage of total phosphates than traditionally seen in stormwater. Despite low concentrations, orthophosphates were consistently removed even with largely varying concentrations seen on Figure 25. High removal percentages for phosphate and orthophosphate were expected. Steel slag has consistently shown the ability to remove phosphates in lab scale and field studies, therefore the results found are within expectations. Previous studies have shown approximately 50-75 percent phosphorus removal in full scale filters and as high as 99 percent in lab scale batch studies using steel slag (Lana et al. 2006, Bowden et al. 2009, Bratt and Shilton 2010, Westholm 2010).

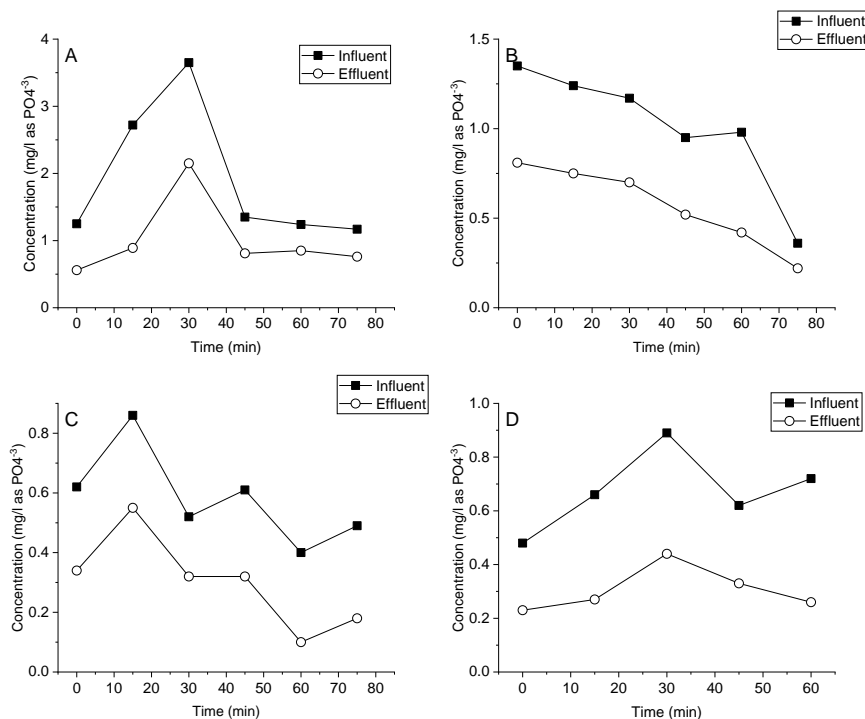


Figure 24. Total Phosphate concentrations in influent and effluent stormwater versus duration of runoff event. (A) 06/22/2019 (B) 07/20/2019 (C) 08/12/2021 (D) 08/31/2019

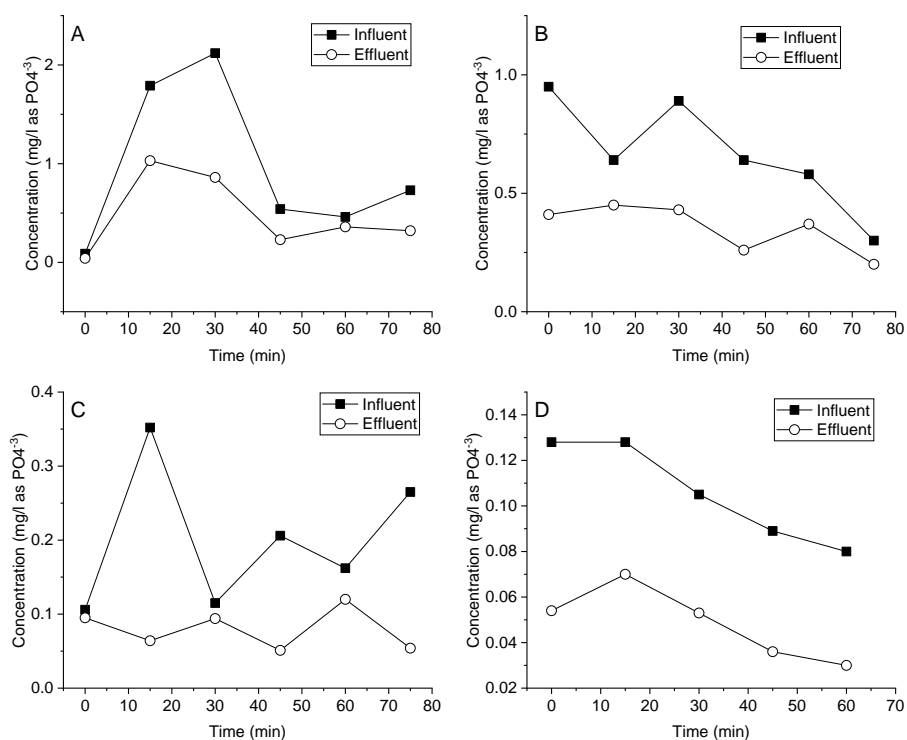


Figure 25. Orthophosphate concentrations in influent and effluent stormwater versus duration of runoff event. (A) 06/22/2019 (B) 07/20/2019 (C) 08/12/2021 (D) 08/31/2019

3.6.2 2020 Field Filter

The field filter and material from 2019 was left to age to examine the performance of steel byproducts over time. Aside from filter remediation to fix agglomeration of the media, the field filter was untouched. An average of 54% of E. Coli was removed for the four storm events sampled in 2020. Like the trend seen in the 2019 data, influent samples containing higher E. Coli concentrations removed higher percentages. This can be seen on Figure 26, which represents the influent and effluent concentrations during the four storm events. The influent concentrations ranged from 325MPN/100ml to

9804MPN/100ml. These all fall within the expected range of stormwater concentrations but are overall lower than the those seen in the 2019 storm events.

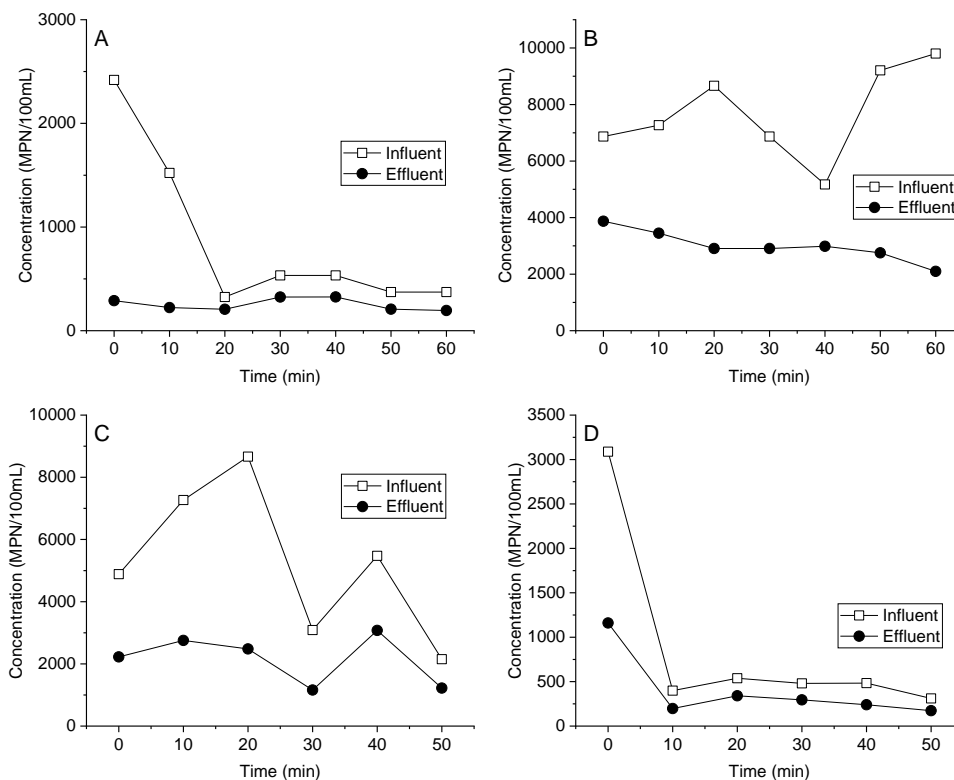


Figure 26. E. Coli concentrations in influent and effluent stormwater versus duration of runoff event. (A) 07/21/2020 (B) 08/09/2020 (C) 08/30/2020 (D) 09/07/2020

The 2020 field filter removed 54% of total phosphorus and 45% of orthophosphates from the influent stormwater. Influent concentrations for total phosphorus ranged from 0.74 to 2.70 mg/l as PO_4^{3-} . Data points for the influent and effluent total phosphorus concentrations can be found on Figure 27. Storm events with higher total phosphorus concentrations saw greater removal than those with low concentrations. The July 21st event had the highest average removal of 58% and all

influent concentrations exceeded 2 mg/l as PO_4^{-3} . None of the remaining three storm events had influent concentrations exceeding 2 mg/l as PO_4^{-3} and did not see removals exceeding 50%. Influent concentrations for orthophosphates ranged from 0.35 to 2.42 mg/l as PO_4^{-3} . Data points for the influent and effluent orthophosphate concentrations can be found on Figure 28. Higher influent orthophosphate concentrations saw higher average removal. Like total phosphorus, the July 21st event had the highest removal due to the highest influent concentration of orthophosphates. Orthophosphates made up a significant portion of the total phosphates in 2020. This indicates that there was a large amount of reactive phosphorus present in the stormwater influent during the storm events.

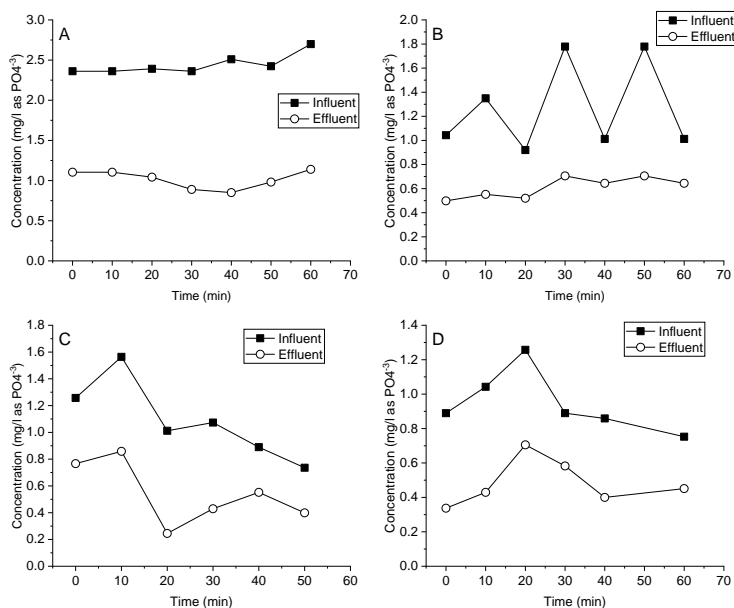


Figure 27. Total Phosphate concentrations in influent and effluent stormwater versus duration of runoff event. (A) 07/21/2020 (B) 08/09/2020 (C) 08/30/2020 (D) 09/07/2020

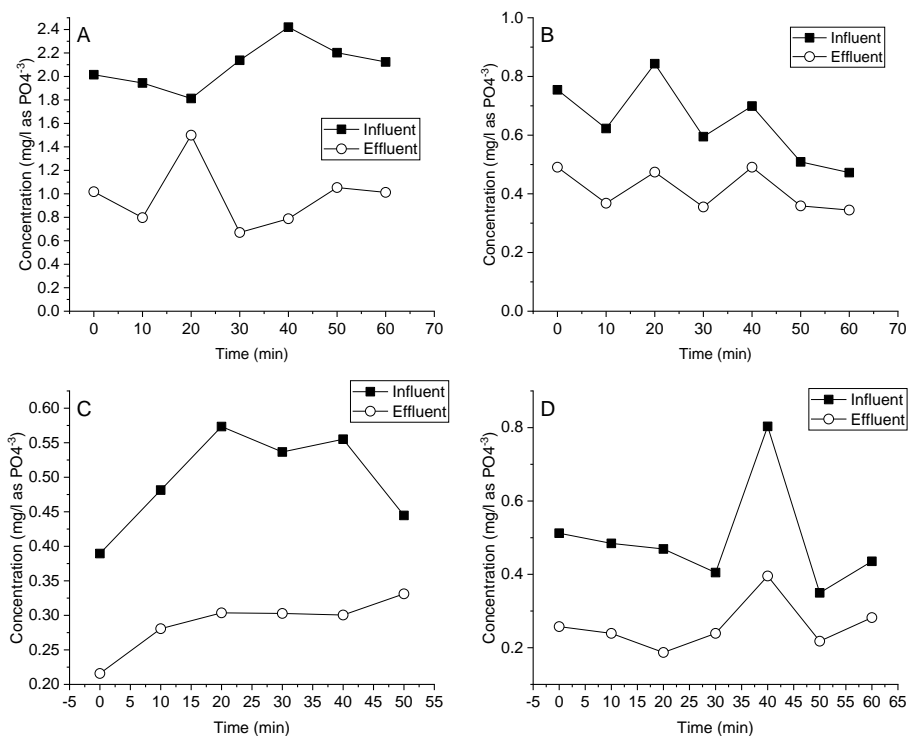


Figure 28. Orthophosphate concentrations in influent and effluent stormwater versus duration of runoff event. (A) 07/21/2020 (B) 08/09/2020 (C) 08/30/2020 (D) 09/07/2020

There was an increase in the average dissolved iron concentrations in the effluent in 2020. The average net dissolved iron in the effluent increased to 0.47 mg/L from 0.076 mg/L in 2019. The difference between 2019 and 2020 can be attributed to the oxidation of the steel byproducts as the filter aged. Material used for the 2019 filter was new and was not heavily oxidized during the sampling period. Influent and effluent concentrations of dissolved iron for the four storm events in 2020 can be seen on Figure 29. Effluent values in 2020 are much higher than those in 2019, ranging from 0.184 to 0.836 mg/l.

Despite this, all effluent concentrations measured do not exceed the EPA recommendation for recreational water of 1.0 mg/l (EPA 1986).

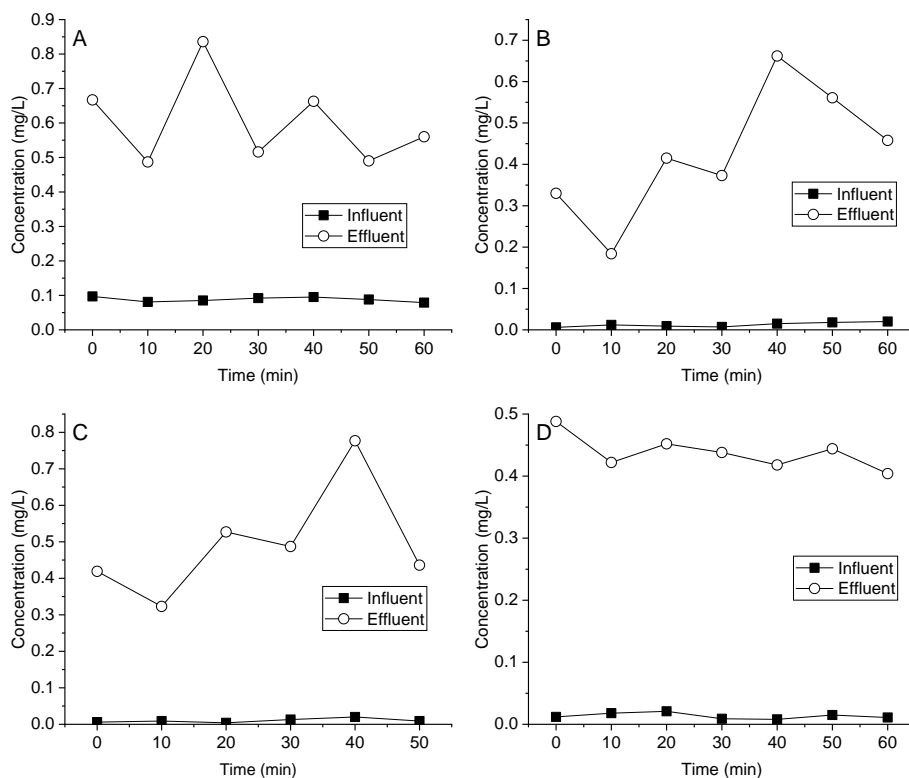


Figure 29. Iron concentrations in influent and effluent stormwater versus duration of runoff event. (A) 07/21/2020 (B) 08/09/2020 (C) 08/30/2020 (D) 09/07/2020

An average of 8.0 percent of nitrate was removed over 4 storm events sampled from the 2020 field filter. Influent and effluent nitrate concentrations for these events can be seen on Figure 30. Seen on the figure, high influent nitrate concentrations were more consistently removed by the filter. The July 21st storm event removed nitrate in 4 of the 5 data points, while the other events did not see removal in over half of their points. A batch study performed at the School of Metallurgical and Ecological Engineering in 2017 showed that modified steel slag could remove 20 percent of nitrate at an initial

concentration of 300 mg/L (Yang et al. 2017). This study demonstrates that slag may better remove high concentrations of nitrate, which is consistent with the findings of the field filter. A column study on steel chips by Morgan Salo showed 68.4% nitrate removal within the first 21 days with an influent concentration of 20 mg/L (Salo et al. 2016). While these studies indicate both steel byproducts being examined have nitrate removal potential, they are not done under similar conditions to the stormwater field filter. The studies have higher influent concentrations and detention times. The findings of this study more closely resemble those found by Jason Neville. While he found some minimal removal during his testing, he concluded that there was not sufficient evidence his filter could consistently remove nitrate (Neville 2019).

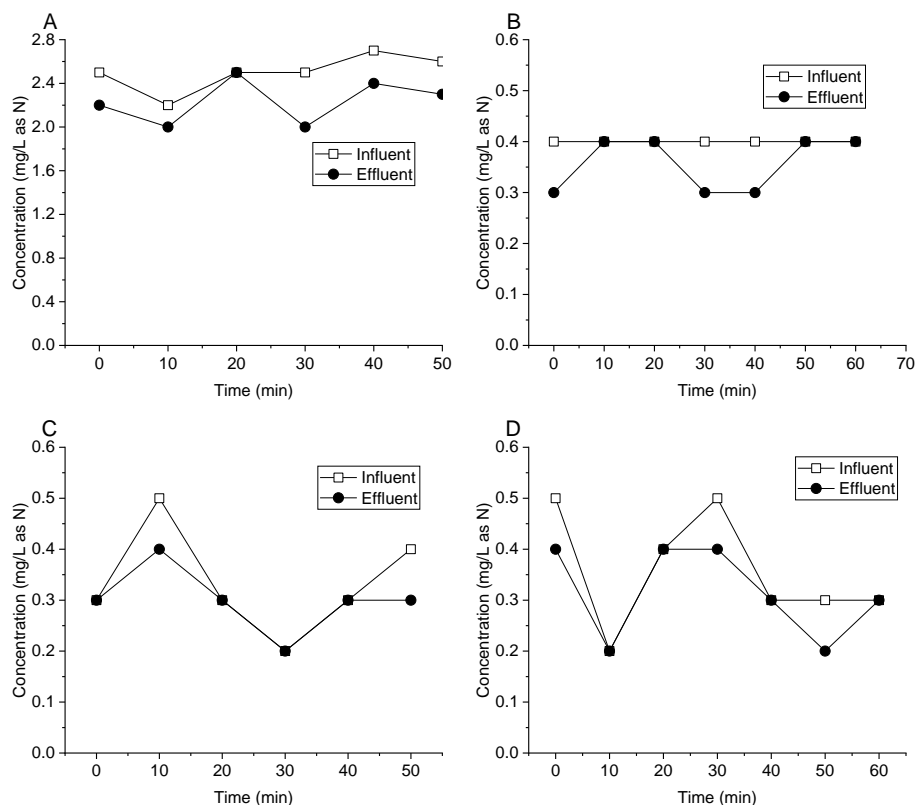


Figure 30. Nitrate concentrations in influent and effluent stormwater versus duration of runoff event. (A) 07/21/2020 (B) 08/09/2020 (C) 08/30/2020 (D) 09/07/2020

Total nitrogen was more readily removed than nitrates in the field filter. An average of 46 percent of total nitrogen was removed over 4 storm events sampled. This means the filter more effectively removed ammonia nitrogen, organic nitrogen, and nitrite compared to nitrate. Detailed analysis was not done on the individual removal of ammonia nitrogen, organic nitrogen, and nitrite so further analysis would be needed to draw conclusions on which is most effectively removed. Figure 31 shows the influent and effluent total nitrogen concentrations for the 4 storm events. As seen, high influent concentrations were removed similarly to lower concentrations. It should be noted that

the detection limit for nitrate and total nitrogen was low using the available equipment. Future studies examining the performance of steel byproducts in removing nitrate and total nitrogen should be conducted further to get obtain more accurate results.

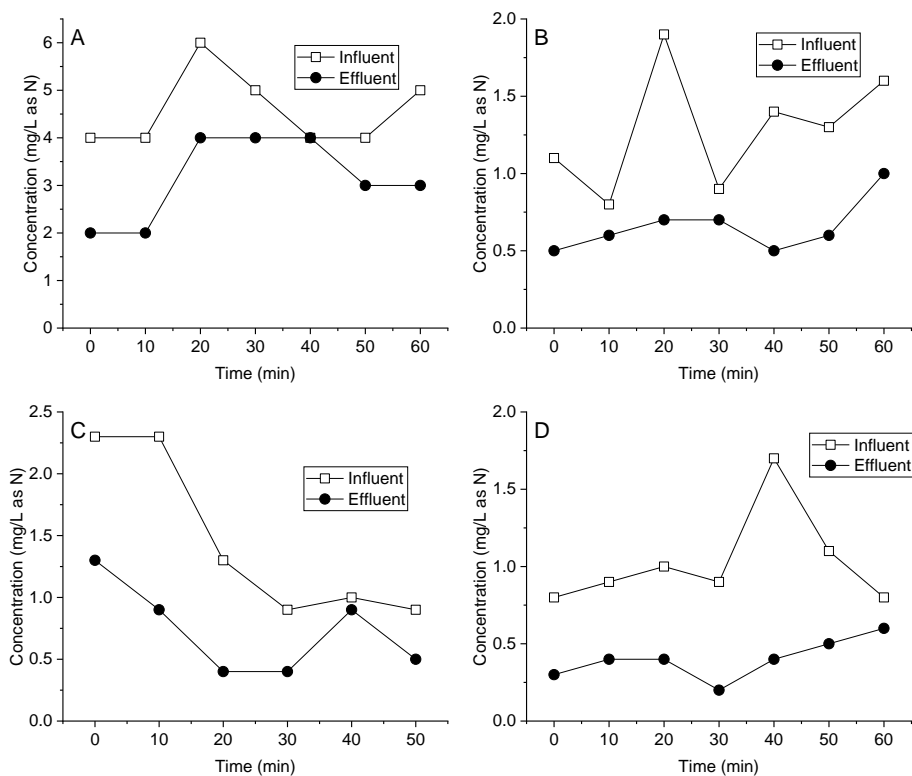


Figure 31. Total nitrogen concentrations in influent and effluent stormwater versus duration of runoff event. (A) 07/21/2020 (B) 08/09/2020 (C) 08/30/2020 (D)

09/07/2020

3.6.3 2021 Field Filter

To evaluate the impact of a reduced ratio of steel chips to steel slag, the 2021 field filter was altered to 30% chips and 70% slag. With this ratio, an average of 30% of E. Coli was removed. The average removal of E. Coli declined from 2019 and 2020, but the

new ratio still consistently removed E. Coli at similar concentrations to those seen in the 2019 storm events. Figure 32 shows influent and effluent E. Coli concentrations during the four storm events. Influent concentrations ranged from 1230 MPN/100ml to 10460 MPN/100ml. Overall higher percentages of E. Coli were removed at higher influent concentrations.

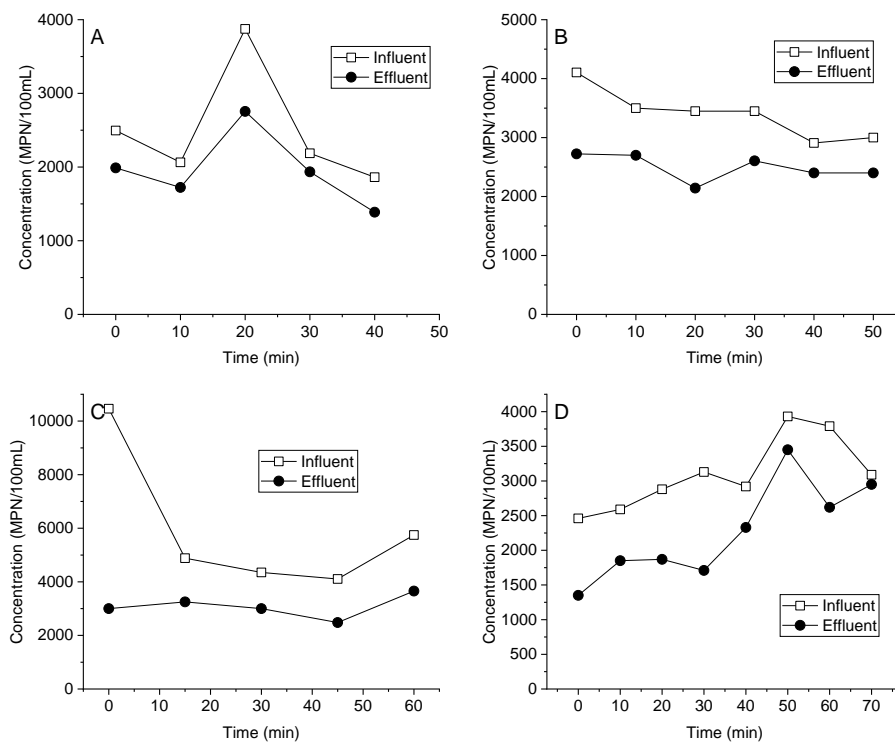


Figure 32. E. Coli concentrations in influent and effluent stormwater versus duration of runoff event. (A) 05/25/2021 (B) 07/10/2021 (C) 07/11/2021 (D) 07/25/2021

Reduction in the percentage of chips in the filter led to a substantially lower average dissolved iron concentrations in 2021. The average net increase in dissolved iron was 0.029 mg/l. This decrease is due to the modification of the filter to 30% chips and

70% slag and the aged material undergoing less oxidation. Slag is comprised primarily of limestone and silica whereas steel chips are predominately iron (Nippon Slag Association 2003). Figure 33 shows the influent and effluent concentrations from the four storm events, effluent concentrations ranged from 0.013 to 0.080 mg/l. All dissolved iron effluent concentrations in 2021 were significantly below the EPA's recommended limit of 1.0 mg/L (U.S. EPA 1986). This indicates that the dissolved iron concentrations leached from the field filter with the new chips to slag ratio would be safe in recreational water.

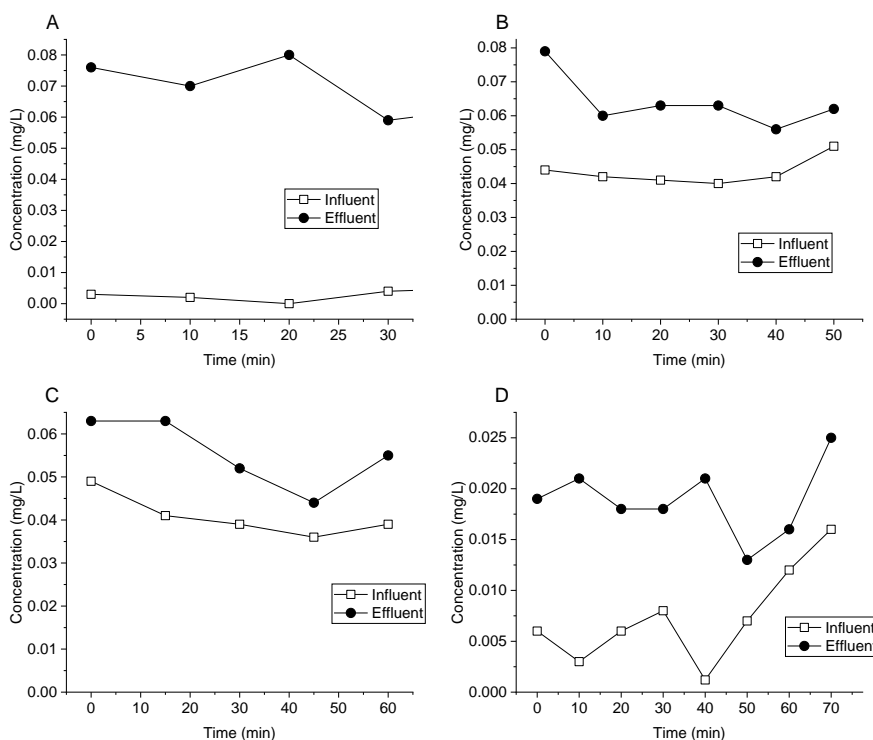


Figure 33. Iron concentrations in influent and effluent stormwater versus duration of runoff event. (A) 05/25/2021 (B) 07/10/2021 (C) 07/11/2021 (D) 07/25/2021

During four storm events an average of 58% of orthophosphate was removed. The influent and effluent orthophosphate concentrations can be found on Figure 34. The

highest average removal of the individual events was during the first rainfall event on May 25th, where an average of 77% was removed. This may be due to the addition of new slag to change the steel byproduct ratio of the filter. Prior to the first rain event, there was no water flow through the filter and the new slag remained uncoated by ferric oxide from the steel chips. It is possible that the orthophosphates were better removed by the new slag. After the first event the field filter still consistently removed orthophosphates, but at lower percentages.

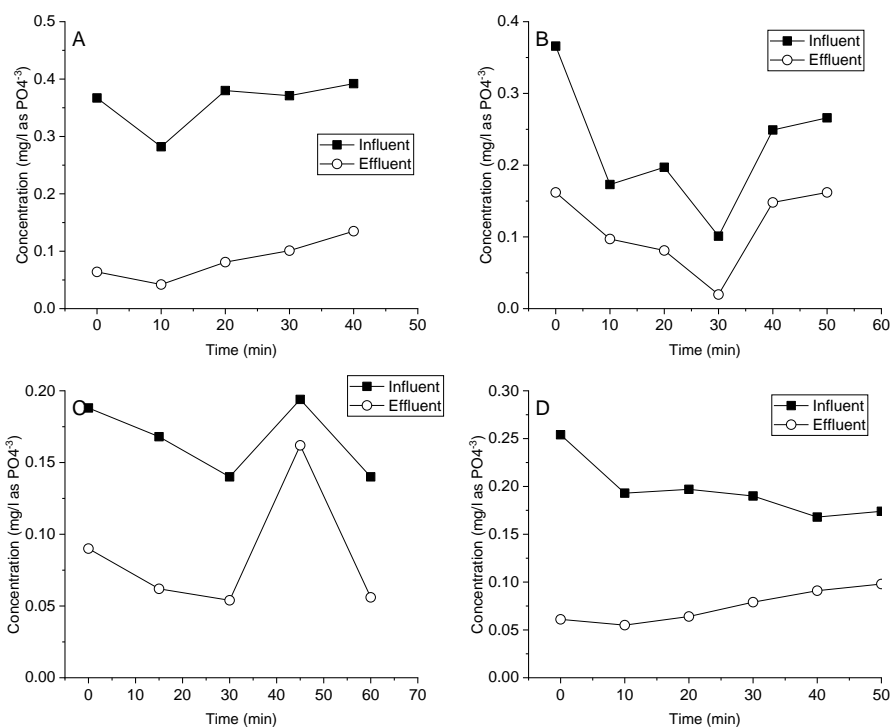


Figure 34. Orthophosphate concentrations in influent and effluent stormwater versus duration of runoff event. (A) 05/25/2021 (B) 07/10/2021 (C) 07/11/2021 (D) 07/25/2021

Total phosphorus was not as effectively removed as orthophosphates in the 2021 field filter, with an average of 41 percent removal. This indicates other species of

phosphorus like organic phosphate and polyphosphate are not as readily removed. Figure 35 shows the influent and effluent total phosphorus concentrations for the four storm events sampled. The highest average removal was during the first storm event on May 25th, similarly to the orthophosphates. This is likely due to the same reason explained in the previous paragraph on orthophosphate removal.

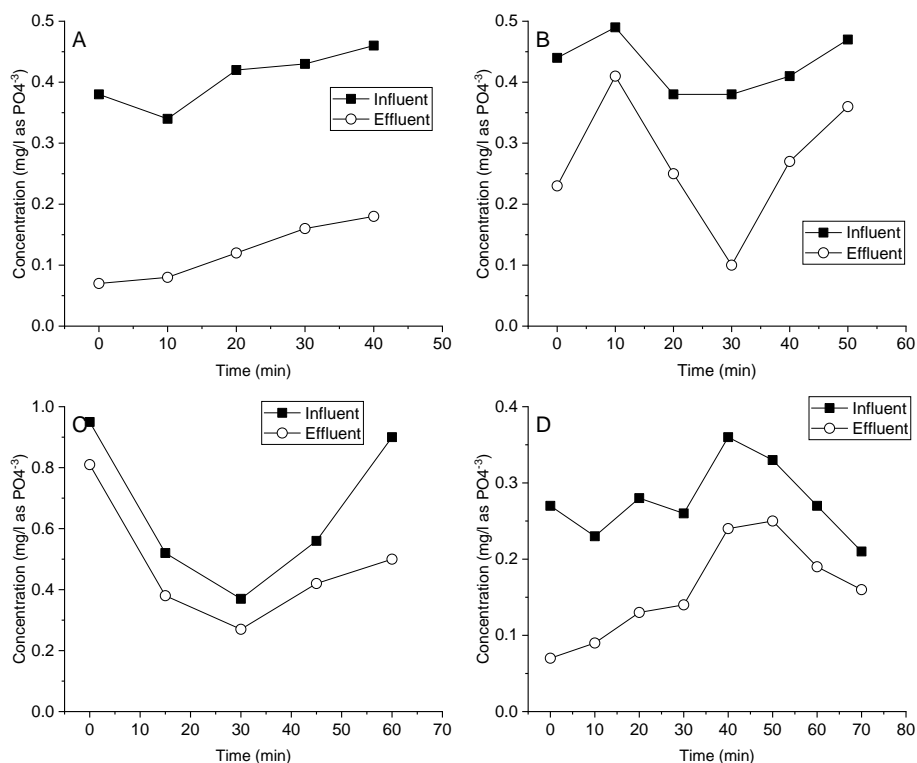


Figure 35. Total Phosphorus concentrations in influent and effluent stormwater versus duration of runoff event. (A) 05/25/2021 (B) 07/10/2021 (C) 07/11/2021 (D) 07/25/2021

An average of 7.6% of nitrate was removed by the 2021 filter over two stormwater samples tested. Figure 36 shows the influent and effluent concentrations of nitrate during these events. As seen on the figure, influent nitrate concentrations were

relatively low, not exceeding 0.5 mg/l as N. The low removal and low influent concentrations are like the results found in 2020. The change in steel byproduct ratio did not impact the removal rate of nitrate.

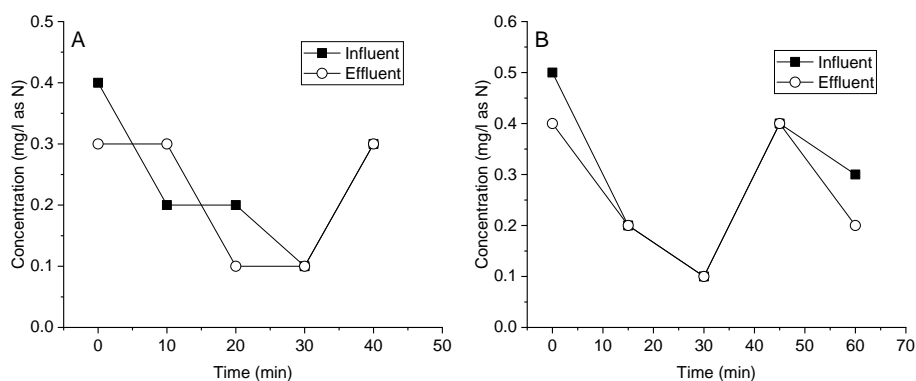


Figure 36. Total Nitrate concentrations in influent and effluent stormwater versus duration of runoff event. (A) 05/25/2021 (B) 07/11/2021

Figure 37 shows the influent and effluent total nitrogen concentrations over two storm events for the 2021 field filter. An average of 39% was removed over the two events. The results indicate that the field filter may be limited to the amount of total nitrogen it is able to adsorb at one time. The May 25th event has significantly higher influent concentrations ranging from 2.3 to 2.6 mg/l as N while the July 10th event has concentrations ranging from 0.3 to 1.0 mg/l as N. Despite this, both events remove similar amounts of total nitrogen from the water. The May 25th event removes concentrations ranging from 0.4 to 0.8 mg/l as N while the July 10th event removes 0.2 to 0.5 mg/l as N despite starting with much lower influent concentrations. It should be noted that the detection limit for nitrate and total nitrogen was low using the available equipment. Future studies examining the performance of steel byproducts in removing nitrate and total nitrogen should be conducted further to get obtain more accurate results.

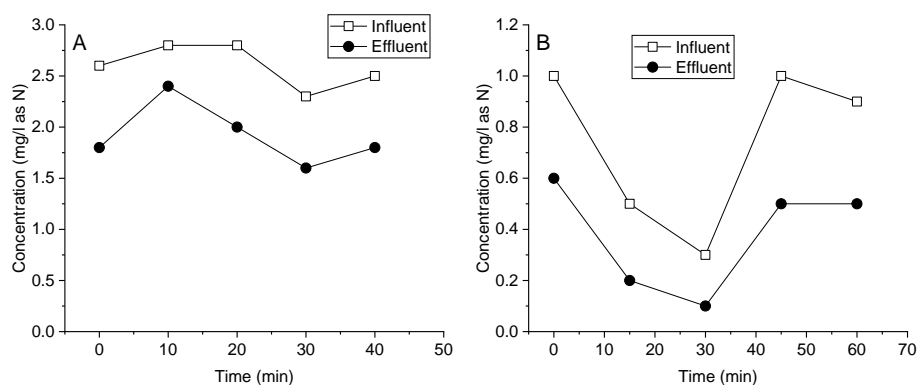


Figure 37. Total Nitrogen concentrations in influent and effluent stormwater versus duration of runoff event. (A) 05/25/2021 (B) 07/11/2021

3.6.4 Comparison of 2019-2021 Field Data

Figure 38 shows the average removal with standard error bars in 2019, 2020, and 2021 for E. Coli, total phosphate, orthophosphate, and dissolved iron. Statistical analysis using a two tailed t test at a 95% confidence interval with an α of 0.05 was used to compare the average removal percentages of E. Coli, total phosphate, and orthophosphate for each field study year. Statistical analysis of nitrogen and nitrate could not be done due to limitations on the detection limits by the equipment. The results indicated that there was no statistical difference in the average removals for E. Coli, total phosphate, and orthophosphate between 2019 and 2020. This shows that over a period of one year the performance of the field filter remains the same, indicating potential long-term use. Some studies have cited limitations on the adsorption capacity of steel byproducts during lab scale testing. The results indicate adsorption capacity may not be problematic in steel byproduct filters with intermittent flow events.

The t testing showed the average removal of E. Coli, total phosphorus, and orthophosphate in 2021 was statistically different from 2019 and 2020. Reduced E. Coli removal was expected after the filter ratio was modified to 30% chips and 70% slag. Lab scale column studies and batch studies have shown steel chips to be significantly more effective in removing E. Coli than steel slag. Slightly increased orthophosphate and decreased total phosphorus removal is likely due to the increase in slag in the mixture. Studies have shown that slag is more effective in removing reactive phosphate (orthophosphate) because positively charged elements and compounds within the slag attract the negatively charged orthophosphates (Ping et al. 2015).

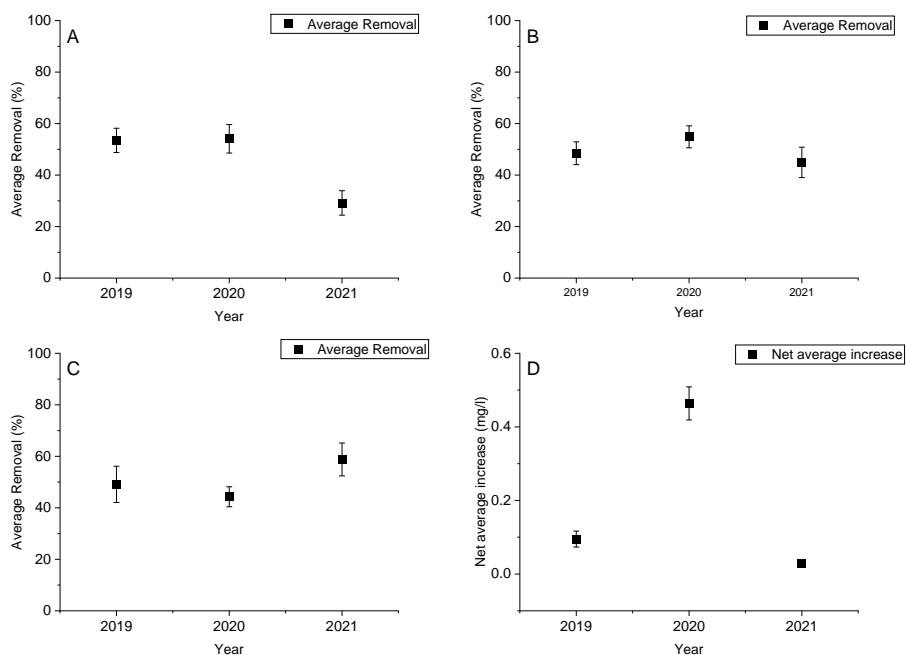


Figure 38. Pilot scale steel byproduct filter performance from 2019-2021 with standard error bars (A) Average E. Coli removal (B) Average Total Phosphorus removal (C) Average Orthophosphate removal (D) Average net iron increase

4. CONCLUSIONS AND RECOMMENDATIONS

This experiment was performed to examine the long-term capability of steel byproducts to remove E. Coli and other common contaminants from stormwater. Batch studies conducted over 24 hours showed that steel chips removed over 99% of initial E. Coli concentrations while steel slag removed between 46% and 73%. Column studies examining the impact of aging on material showed steel slag to remove 25% and 18% of E. Coli for aged and new material respectively. Similar column studies showed steel chips to remove an average of 83% and 45% of E. Coli for new and aged material respectively. Varying ratios of steel slag to chips were examined in column studies to modify the existing pilot filter. Ratios of 5%, 10%, 20%, and 50% steel chips removed an average of 35%, 50%, 57% and 62% of E. Coli respectively. Influent and effluent samples were collected from a field filter in 2019, 2020, and 2021 to examine its effect on E. Coli, phosphorus, orthophosphate, nitrogen, nitrate, and iron. In 2019 and 2020 the field filter was composed of 50% steel slag and 50% steel chips; this ratio was modified to 70% steel slag and 30% steel chips in 2021. In 2019 the field filter removed an average of 53%, 48%, and 49% of E. Coli, total phosphorus, and orthophosphate respectively. In 2020 the field filter removed average of 54%, 54%, 45%, 45%, and 8% of E. Coli, total phosphorus, orthophosphate, total nitrogen, and nitrate respectively. In 2021 the field filter removed an average of 29%, 41%, 58%, 39%, and 8% of E. Coli, total phosphorus, orthophosphate, total nitrogen, and nitrate respectively. The 2019 and 2020 field filters released an average of 0.27 mg/l of dissolved iron into the effluent while the 2021 filter released an average of 0.02 mg/l. The results from this experiment suggest that steel byproducts can potentially be used to remove E. Coli, total phosphorus, and

orthophosphate from stormwater while not releasing significant concentrations of dissolved iron into effluent water. Based on the study, steel byproducts appear to have limited potential in removing low concentrations of nitrate and nitrogen from stormwater. Due to limitations with the detection limits in the equipment being used, nitrate and nitrogen removal using steel byproducts should be reexamined using different testing methods. In the future, large scale field studies are necessary to examine the efficiency of the field filter under different stormwater conditions.

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