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BACTERIA REMOVAL FROM STORMWATER RUNOFF USING STEEL BYPRODUCT FILTERS

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Bacteria Removal from Stormwater Runoff Using Steel Byproduct Filters

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ABSTRACT

Escherichia coli (E. coli) is often used as an indicator organism to quantify the microbial safety of natural water resources. E. coli in stormwater runoff can cause serious health risks in natural water bodies. Conventional stormwater best management practices are generally not effective at removing E. coli from stormwater runoff. A new filtration technology using recycled steel byproducts has been developed to remove E. coli from stormwater. Steel chips and steel slag are two common steel byproducts that showed high capacities for E. coli removal. The main objective of this study was to evaluate the long-term performance of steel byproduct filters and provide recommendations for future field applications. Laboratory and field scale studies were conducted to determine the impact of material mixing ratios and operation time on the filter performance. The results showed that the steel byproduct filter exhibited consistent E. coli removal during three consecutive years of operation. Steel chips and steel slag are effective filter materials for long-term field applications. Higher ratios of steel chips to steel slag in the steel byproduct filter resulted in higher E. coli removal. However, large quantities of steel chips in the filter may cause material agglomeration during filter operation, which can negatively affect the hydraulic properties of the filter. We recommend that the steel chips to steel slag ratios should not exceed 30% for field scale applications based on the results of this study.

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EXECUTIVE SUMMARY

Introduction

Water pollution has become increasingly problematic as global populations and industrialization continue to rise. Nonpoint source pollution is a major source of pollution in stormwater. It is a challenge to track and control nonpoint source pollution because it comes from a seemingly infinite number of sources. All unregulated activities like fertilization, improperly managed land use, chemical spills, and others contribute to pollution sources. These activities contribute to nonpoint source pollution by combining with runoff generated during storm events. Stormwater runoff occurs when precipitation events cause an excess amount of water to flow over surfaces; the runoff often picks up pollutants and contaminants, which can pose a threat to water systems. Therefore, it is important to develop technologies to control stormwater contamination.

To manage stormwater and protect aquatic ecosystems, there are several stormwater management practices 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. Wetlands are another common stormwater management practice that uses a variety of plants to control stormwater quality. Although these conventional technologies can remove some common water contaminants such as suspended particles, they are not effective at removing microorganisms. Pathogenic microorganisms in water may cause serious harm to human and animal health depending on the type and their abundance. Among different types of microorganisms in natural waters, Escherichia coli (E. coli) is commonly used to indicate potential health risks for human purposes. E. coli is often used as an indicator organism to quantify the microbial safety of natural water resources.

A new filtration technology using steel byproducts has been developed to remove E. coli from stormwater. 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 studied in recent years. The results showed that steel chips and steel slag are effective filtration materials for E. coli removal. A field scale study in the City of Brookings demonstrates that a mixed media filter with steel chips and steel slag effectively removed E. coli from stormwater at a residential detention pond.

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 an 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.

Laboratory Study

Laboratory batch and column experiments were conducted to evaluate the E. coli removal capabilities of new and aged steel chips and steel slag under controlled conditions. The results of the adsorption kinetics experiments showed that steel chips removed more than 99% of E. coli after a 24-hour adsorption time, while steel slag removed between 46% and 73%. New steel ships showed faster adsorption kinetics than aged steel chips. However, aged slag exhibited better E. coli removal efficiencies than new steel slag. The high E. coli removal observed for aged slag can be attributed to the surface iron coating due to the interaction with steel chips. Similar results were also observed during the continuous flow column experiments. Steel chip column reactors removed an average of 83% and 45% of E. coli for new and aged material, respectively. Steel slag column reactors removed 25% and 18% of E. coli for aged and new material, respectively. Additional column experiments were also performed to evaluate the impact of varying ratios of steel slag to chips on E. coli removal. The results showed that ratios of 5%, 10%, 20%, and 50% steel chips removed an average of 35%, 50%, 57% and 62% of E. coli, respectively.

Field Scale Study

After the laboratory batch studies, field filtration studies were performed to determine the removal efficiencies of E. coli and other contaminants from stormwater. The field filtration studies were conducted using an existing steel byproduct filter installed at a residential stormwater detention pond in the City of Brookings. Filter influent and effluent samples were collected from this 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 an 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 30%, 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. All dissolved iron effluent concentrations during the field scale study were below the EPA's recommended limit in recreational water.

Recommendations

The results of the lab and field studies demonstrate that recycled steel byproducts are effective filter materials for E. coli removal. The steel byproduct filter is also capable of removing phosphate from stormwater. Steel chips showed higher E. coli removal capacity than steel slag. The ratio of steel chips to steel slag is a critical factor determining E. coli removal efficiencies. Higher ratios of steel chips in the steel byproduct filter resulted in higher E. coli removal. However, more quantities of steel chips in the filter may cause material agglomeration during filter operations. Therefore, the steel chips to steel slag ratio should be carefully selected to minimize the material agglomeration while maintaining effective E. coli removal. The field filter operation experience from this study indicates that substantial material agglomeration occurred when the steel chips ratios exceeded 50%. When the steel chips ratio was adjusted to 30%, the material agglomeration issue was significantly improved. Therefore, we recommend that the steel chips to steel slag ratios should not exceed 30% for field scale applications. More full-scale filter studies should be performed to determine the optimum steel chips to steel slag ratios for stormwater treatment.

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 the Clean Water Act (CWA) in 1972 (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, the CWA heavily reduced contamination levels in many water systems by monitoring the pollution sources. However, unregulated 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, pet walking, small chemical spills, and others 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 1987 CWA amendments tries to address NPSP by providing annual funding for states to reduce contamination levels in their water systems (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% of the continental U.S. (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% of the world's population lives in urban areas, and this is expected to grow to 68% 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 their 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 rain event specifics, 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. Their abundance promotes the 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 fertilizer use followed 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 during 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 of particles can affect plants and wildlife. Increased suspended solids may deposit in sensitive ecosystems, blocking out sunlight or creating sediment buildups. Reduced levels of sunlight and sediment buildup 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. 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 such as 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 risk. Stormwater often recharges aquifers, and microorganisms in stormwater could negatively impact groundwater quality. 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 also develop. Advancements in pharmaceuticals, personal care products, industrial products, and other chemicals have led to 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 that 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 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—a bioretention pond—is also frequently used. These operate similarly to normal retention ponds but have vegetated areas that help stormwater treatment.

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. Wetlands can remove contaminants through sedimentation, adsorption and retention, biological degradation, and plant uptake. After flowing through the wetland, water is often discharged into infiltration basins. In these basins, 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

1.2.1 E. Coli Occurrence

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. E. coli is often used as an indicator organism to quantify the microbial safety of natural water resources. To limit pollutants and substances of concern in water systems, the United States Environmental Protection Agency (EPA) passed the Clean Water Act (CWA). In addition to this document the 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. Table 1.1 presents the EPA recreational water recommendations for E. coli.

Contaminated food and water sources are common causes of 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% of its untreated wastewater into the local river, E. coli concentrations increased from 3.5x100 MPN/100mL to 2.8x1000 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 was found that E. coli and total coliforms exceeded drinking water 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 of 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 1996 study highlighted the E. coli problem in the food service industry by examining vegetables and salads. Approximately 14% 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. The results showed that more than 60% of poultry packages had meat juice on them, the majority contaminated with E. coli (Chen et al., 2018). To reduce the risk of E. coli infection from foods, proper washing and cooking is recommended.

Commonly perceived as dangerous to human health, many strains of E. coli are harmless. However, some strains are pathogenic. There are several strains of E. coli that may have adverse effects on human health such as 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 health centers. 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 studies showed that antibiotic-resistant E. coli strains are increasing, which may pose future risks to the public.

1.2.2 Conventional Treatment for E. Coli

(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 byproducts (DBPs). DBPs are formed from chlorine reacting with natural organic matter (NOM) and other compounds of interest like bromide and iodide. Studies have shown that DBPs can pose serious risks to human health. Some classes of DPBs are regulated, such as trihalomethanes (THM) and haloacetic acids (HAA). Other classes exist but are unregulated due to a lack of information on health impacts and their small quantities. Some DPBs have been associated with increased risks of different cancer types in humans. Chlorine itself is toxic to many aquatic organisms even at low concentrations and therefore cannot be discharged into natural water systems.

Ultraviolet (UV) light is another common disinfection technique, with comparable disinfection effectiveness as chlorine. UV radiation emitted by mercury arc lamps damages 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 lead to 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 operational 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.

(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 exceeds 90% when designed properly (Struck et al., 2006).

While many factors impact the overall removal, the driving mechanism is considered to be sunlightmediated disinfection (Passos, Dias, and Sperling, 2020). This process is similar to that 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 natural organic matter. Retention ponds only release water through evaporation and infiltration, therefore have an infinite detention time. Therefore, retention ponds are limited in their capacity and may overflow during large storm events.

(3) Detention Ponds

Detention ponds are like retention ponds in that they use the same mechanisms to treat introduced water. However, detention ponds have limited hydraulic detention time. The water released from detention ponds may have slightly higher quality than the influent, but treatment is limited compared with 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.

(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 (EPA, 1990). Constructed wetlands are artificially designed to replicate these ecosystems. It is difficult to determine the exact efficiency of wetlands in removing bacteria due to their complexity and biodiversity. For example, a twoyear 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 rates, exceeding 90% 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 reported only a 6.7% E. coli removal rate (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 Technologies for E. Coli

(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. A 2018 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 1,000 CFU/mL. The clams only contained 1% to 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 using zebra mussels. After 24 hours, E. coli was completely removed from sample volumes (Mezzanottea 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% E. coli removal; fast media like perlite, vermiculite, growstone, expended clay, or river sand showed up to 20% (Prodanovic et al., 2017). Used properly, biofilters would be an ecologically friendly approach to reduce E. coli concentrations in stormwater.

(2) Biochar

Biochar is pyrolyzed biomass, a charcoal-like material 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 during filtration processes, 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).

(3) Coated Sands

Slow sand filtration was one of the first technologies used to effectively remove pathogenic bacteria from surface water. Bacterial removal is highly dependent on the composition and grain size of the sand. Sand filters demonstrate a wide range of removal capabilities for various contaminants. 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 require either backwash or filter replacement. 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.

(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 studied in recent years. The results from the research done on steel byproducts have 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 two 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. It was found that the largest size range of chips (4–9 mm in diameter) removed 60% of E. coli and the smallest (0.5–1 mm 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 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 steels chips examined the real stormwater removal by steel byproducts. It was found that the filter could consistently remove 50% of E. coli over several different runoff events (Neville, 2019). Overall, steel byproducts appear to be a promising low-cost medium that can remove E. coli from water.

1.3 Project Objectives

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 showed that steel byproducts have potential in removing high percentages of E. coli from stormwater through lab scale and field scale testing. An evaluation of steel byproducts' long-term performance 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 studies of Steel Byproducts

2.1.1 Introduction

Laboratory batch and column studies were conducted to evaluate the E. coli removal efficiencies of steel chips and steel slag. 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 new and aged steel chips and steel slag that were 2–4 mm in diameter and 4–9 mm in diameter, respectively. Different E. coli levels were used to simulate bacterial concentrations in stormwater. The column studies were used to evaluate E. coli removal under continuous flow conditions.

2.1.2 E. Coli Preparation

Cultured E. coli was used to prepare solutions for lab studies. 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 NaHCO₃ (0.84 g/L), 0.1 mM KCL (0.7455 g/L) and 1.2 mM H₂SO₄ (0.49 g/L).

The cultured E. coli was then evenly separated into three 50 mL centrifuge containers and rotated at 4,000 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 solution. The centrifuge containers were then reinserted into the centrifuge and this process was repeated a total of three times to clean the culture solutions. After the third time, E. coli in each container was resuspended with the buffer solution and poured into a 100 mL glass flask. To estimate the approximate number of E. coli cells per mL, a Hach DR/4000 spectrophotometer was used. The measured E. coli concentrations using this process fell within a range of $1.0-1.6x10^9$ cells/mL on the spectrophotometer. The 100 mL container of E. coli stock was stored at room temperature for up to five 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 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. These included 2– 4 mm new, 2–4 mm aged, 4–9 mm new, and 4–9 mm aged. Figure 2.1 and Figure 2.2 show the new and aged steel byproducts after sorting them according to their size and material, respectively. 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.

Aged materials were gathered from an existing field filter created by SDSU after two years in use. The existing filter was a mixture of 50% steel slag and 50% steel chips. Approximately 10 gallons of material were removed from the filter. Sieves were used to separate the mixed materials into different size groups. After sieving 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 2.1 New and aged steel slag materials (top: 2–4 mm new, 2–4 mm aged; bottom: 4–9 mm aged, 4–9 mm new)

Figure 2.2 New and aged steel chip materials (top: 2–4 mm new, 2–4 mm aged; bottom: 4–9 mm aged, 4–9 mm new)

2.1.4 Batch Studies

One gram of each of the eight tested materials was placed in 250 mL Erlenmeyer flasks. 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 107 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 ScientificTM MaxQTM 4000 benchtop orbital shaker set to 20℃ at 100 rpm for 24 hours. This setup can be seen in Figure 2.3. One mL of the sample was taken from each of the 10 flasks at times of one-half hour, one hour, two hours, three hours, 12 hours, and 24 hours to determine E. coli concentrations using the IDEXX method outlined in Section 2.3 Analytical Methods.

Figure 2.3 Benchtop orbital shaker used for batch studies

2.1.5 Column Studies

E. coli removal capabilities of steel byproducts were examined by pumping 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. Tables 2.1 through 2.3 show the characteristics of steel slag and steel chips used for the column studies.

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

Table 2.2 Characteristics of steel chips in the column study

Table 2.3 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

All column experiments used a flow rate of 1.18 mL/min to achieve an empty bed contact time (EBCT) of 15 minutes. The EBCT is the ratio of bed volume to flow rate as shown:

$$
EBCT = \frac{Bed \, Volume \, (mL)}{Flow \, Q \, (\frac{mL}{\min})}
$$

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. Based on this, flow rates were then adjusted by tightening or loosening the tubing.

A five-gallon glass container was used to prepare the influent for column studies. The container was filled with 18 liters of nanopure water and then made into a buffer solution using 1.0 mM NaHCO₃ (0.84g/L), 0.1 mM KCL (0.7455 g/L) and 1.2 mM H_2SO_4 (0.49 g/L). E. coli stock was added until the desired concentration was achieved after the buffer chemicals were added. To sample the influent and effluent of the reactors, 20 mL was taken in small glass vials starting at each sampling period. Influent was poured directly from the influent container and effluent was taken from the ends of the plastic tubing after the columns. Figure 2.4 shows how the column experiment was set up for each of the column studies performed.

Figure 2.4 Reactor setup for column experiments

2.2 Field Stormwater Filtration Studies

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, NE. Chips refer to the excess material from steel manufacturing, formed from physical processes like cutting and shaping steel into finished products. The steels chips were obtained from Alter Metal Recycling in Marshall, MN, and came from carbon steel.

2.2.2 2018—**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, SD, at the intersection of Camelot and Breckenridge Drive. Figure 2.5 shows the installed field stormwater filter. 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. A wider view of the stormwater detention pond for the field filter can be seen in Figure 2.6. 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 (4–9 mm), 12.5% small steel slag (2–4 mm), 50% large steel chips (4–9 mm), and 12.5% small steel chips (2–4 mm). While operating the filter, it was observed that the steel chips gradually agglomerated due to rusting and formed large chunks of rusted materials over time, which affected the filter's hydraulic properties (Figure 2.7). It was necessary to break down the agglomerated materials monthly to maintain the filter's performance.

Figure 2.5 Field stormwater filter site location

Figure 2.6 Stormwater detention pond for the stormwater filtration study

Figure 2.7 Filter material agglomeration observed during the field study

To address the agglomeration issues and test different filter material ratios, the field filters were changed to 50% steel chips and 50% steel slag in 2019 and 2020. 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, and no materials were taken or added during this time frame. 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.

2.2.3 2021 Field Study

To evaluate the removal potential of aged materials used in the field filter and to reduce filter agglomeration, small quantities of filter materials were taken in August of 2020 to perform lab studies. Based on the result of the lab studies, the new ratios for the field filter were chosen to be 30% steel chips and 70% steel slag for the 2021 field study. A higher ratio of slag to chips was desired to counteract the agglomeration observed in previous studies. To create the new filter, all aged materials were removed from the original filter using pickaxes and shovels. New slag and steel chips were added to the filter to reduce the existing steel chips to slag ratio. The filters materials were added in layers and then mixed in the filter to achieve the desired ratio. Figures 2.8, 2.9, and 2.10 show the different steps of the filter redesign in May 2021. Water samples were taken in the same manner as 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% of initial results.

Figure 2.8 Removal of aged materials from the field filter

Figure 2.9 Adjustment of steel byproduct ratio in field filter

Figure 2.10 Pilot scale steel byproduct filter after ratio adjustment

Four stormwater events were sampled for the 2021 field filtration study. The characteristics of the four events are shown in Table 2.4.

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

Table 2.4 Characteristics of stormwater events for 2021 field study

2.3 Analytical Methods

2.3.1 E. Coli Analysis

Accurate E. coli concentrations were determined using the Coliert 18 method, giving MPN (most probable number)/mL. Samples being tested were diluted enough to fall within an expected range of 100 to 2,000 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℃ for 24 hours. Trays were read out using blacklight in a dark room and using calculation instructions provided by IDEXX.

2.3.2 Other Water Quality Parameters

Five water quality parameters were tested to examine the quality of influent and effluent stormwater from the field filter. 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 the persulfate acid 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 to prepare E. coli stock were cleaned using an autoclave. All used glassware was submerged in a 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 $18M\Omega$ nanopure water, and set out to dry. Plastic containers did not receive an acid bath and were instead rinsed with $18M\Omega$ nanopure water after the soap bath and dried.

3. RESULTS AND DISCUSSION

3.1 Batch Study of E. Coli Removal Kinetics

The results from the batch study are shown in Figures 3.1 and 3.2. Duplicate samples were taken for influent and effluent samples. The duplicate sample results are generally within 15% variations. The steel chips removed higher percentages of E. coli with an influent concentration several orders higher than that of the steel slag. New steel chips (2–4 mm and 4–9 mm) achieved 100% removal of the initial E. coli after 24 hours. At the same time, both sizes of aged chips removed 99% of E. coli. As shown in Figure 3.1, the new chips reached a high removal percentage more rapidly, both reaching 99% removal within three hours. This is due to the rapid oxidation of the new chips. Figure 3.3 presents the Erlenmeyer flasks containing different materials. The solutions with new chips turn a brown and orange color. This suggests that new steel chips reacted with dissolved oxygen and produced iron oxides through rusting. The produced iron oxides effectively removed E. coli. Studies examining the bactericidal properties of ferric oxide nanoparticles have concluded that they are able to deactivate many strains of bacteria, including E. coli (Li et al., 2018; Gabrielyan et al., 2019; Diao and Yao, 2009). The aged steel chips also removed high percentages of the E. Coli through adsorption. The negatively charged E. coli attached themselves to the positively charged surface of the steel chips where they were 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 slags removed 73.2%, 71.6%, 58.7%, and 46.3%, respectively. The result for the steel slag batch study is shown in Figure 3.2. Unlike the steel chips, the aged material performed better than the new material. This can be attributed to the interaction of ferric oxides with E. coli. The aged material has ferric oxide attached to its surface, 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 3.1 Steel chips batch study results

Figure 3.2 Steel slag batch study results

Figure 3.3 Erlenmeyer flasks with 1 gram of material after 24-hour adsorption time (from left to right 2–4 mm new chips, 4–9 mm new chips, 2–4 mm old chips, 4–9 mm old chips).

3.2 E. Coli Removal by Steel Chips Column

New steel chips removed a higher percentage of E. coli than the aged steel chips during the column study. As presented in Figure 3.4, the 2–4 mm new, 4-9 mm new, 2–4 mm old, and 4–9 mm old chips removed an average of 80%, 50%, 30%, and 10% of E. coli, respectively. Higher removals were observed during the first two days, and the removal efficiencies stabilized after two days. 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 $(1-2 \text{ mm diameter})$ can removal more than 90% 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).

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 rapid oxidation of the new chips forming large quantities of iron oxides.

Figure 3.4 E. coli removal by steel chips in column study. Experiment conditions: flow velocity= 1.18 mL/min, EBCT=15 minutes

3.3 E. Coli Removal by Steel Slag Column

The steel slag demonstrated the opposite of the findings for the steel chips in that the aged material performed better than the new. Figure 3.5 shows that the 2–4 mm new, 4–9 mm new, 2–4 mm old, and 4–9 mm old slags removed an average of 17%, 13%, 25%, and 21% of E. coli, respectively. Slag shows lower removal rates than that of chips, likely because slag has a lower iron content.

Previous studies have shown that adsorption is the major removal mechanism used by slag in removing E. coli (Hooshyari, 2017; Dai, 2019; Neville, 2019). The aged slag removes higher percentages of E. coli because it was coated with iron oxides through interactions with steel chips in field studies. As explained previously, iron oxide particles have demonstrated bactericidal properties and bacterial adsorption capability in previous studies. The column studies indicate that as slag ages with steel chips its E. coli removal capacity increases.

Figure 3.5 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 3.6 shows the mixed materials prior to being placed in the columns to evaluate the impact of mixing ratios of steel chips and steel slag on E. coli removal. Figure 3.7 shows the E. coli removal of different mixtures of steel slag and chips in a column study. The 5%, 10%, 20%, and 50% steel chips removed an average of 35%, 49%, 57%, and 62% of E. coli over a span of eight days. The E. coli removal efficiencies increased with increasing steel chip ratios. This is consistent with the high E. coli removal potentials of steel chips observed in batch studies. Even low quantities of chips present in the mixture can significantly improve E. coli removal.

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 field filtration studies, the media would form large masses, resulting in reduced flow rates through the filter. Filter remediation was effective but required regular filter maintenance. Overall, the results from the mixture column study showed that even low steel chip percentages present can improve E. coli removal. Therefore, the steel chip percentages can be adjusted to balance the filter performance and required maintenance in field applications.

Figure 3.6 The mixed materials prior to being placed in the columns

Figure 3.7 E. coli removal by steel slag and steel chip mixtures in column study. Experiment conditions: flow velocity=1.18 mL/min, EBCT=15 minutes

3.5 SEM analysis

The surface structure of the new and aged steel chips and slag were observed using scanning electron microscope (SEM) analysis. Figures 3.8, 3.9, and 3.10 show these SEM images at various settings. Each figure illustrates the difference between the new and aged materials. The steel chip's surface is originally smooth with few deformations but after two years in use in the field, the surface is rusted. The new slag shows large amounts of surface pores and cavities. After two years interaction with steel chips, the surface of slag is coated with iron oxides and becomes smoother with fewer surface pores.

Figure 3.8 SEM images of steel byproducts at 10 um. (Top left new chips, top right old chips, bottom left new slag, bottom right old slag)

Figure 3.9 SEM images of steel byproducts at 20 um. (Top left new chips, top right old chips, bottom left new slag, bottom right old slag)

3.6 Field Stormwater Filtration Studies Results

3.6.1 2019 Field Filtration Study

The 2019 field filter was designed with 50% steel slag and 50% steel chips. The field filter was able to remove, on average, 53% of E. coli. Figure 3.11 shows the influent and effluent E. coli concentrations from the 2019 storm events. Influent concentrations in the stormwater ranged from 1,101 MPN/100ml to 15,531 MPN/100ml. The results indicate that higher influent concentrations led to better removals than low concentrations. For example, storm event A removed an average of 74% of E. coli while storm event D removed an average of 39%. Storm event A had a significantly higher average of influent E. coli concentration of 11,302 MPN/100mL; storm event D had an average of 4,644 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 70% chips and 30% slag and removed, on average, 50% of E. coli (Neville, 2019).

Figure 3.11 E. coli removal in 2019 field stormwater filtration study. (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 3.12 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 (EPA, 1986). This shows that the field filter did not release 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 3.12 Iron concentrations in 2019 field stormwater filtration study. (A) 06/22/2019, (B) 07/20/2019, (C) 08/12/2021, (D) 08/31/2019

Figures 3.13 and 3.14 show the total phosphate and orthophoto removals of the field filter in 2019. The field filter removed 48% of total phosphates and 49% of orthophosphates from the influent stormwater. The influent total phosphate concentrations ranged from 0.36 to 3.65 mg/L, and influent orthophosphate concentrations ranged from 0.08 to 2.12 mg/L for different storm events in 2019. The steel byproduct filter demonstrated consistent removal efficiencies of total phosphate and orthophosphate despite large variations in influent concentrations at different storm events. These results show that steel chips and steel slag are efficient materials for phosphate removal from stormwater. Previous studies showed approximately 50%– 75% phosphorus removal in full scale filters and as high as 99% in lab scale batch studies using steel slag (Lana et al., 2006; Bowden et al., 2009; Bratt and Shilton, 2010; Westholm, 2010).

Figure 3.13 Total phosphate removal in 2019 field stormwater filtration study. (A) 06/22/2019, (B) 07/20/2019, (C) 08/12/2021, (D) 08/31/2019

Figure 3.14 Orthophosphate removal in 2019 field stormwater filtration study. (A) 06/22/2019, (B) 07/20/2019, (C) 08/12/2021, (D) 08/31/2019

3.6.2 2020 Field Filtration Study

The field filter materials in the 2020 study were kept the same from 2019 to evaluate the performance of filter materials for two consecutive years. Figure 3.15 shows the E. coli removal in the 2020 field filtration study. The influent E. coli concentrations ranged from 325 MPN/100ml to 9,804 MPN/100ml. 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 typically led to high removal percentages. These results suggest that the filter materials maintained similar E. coli removal efficiencies in two consecutive years of filter operation.

Figure 3.15 E. coli removal in 2020 field stormwater filtration study. (A) 07/21/2020, (B) 08/09/2020, (C) 08/30/2020, (D) 09/07/2020

Figures 3.16 and 3.17 show the total phosphate and orthophoto removals of the field filter in 2020. Filter influent total phosphate concentrations ranged from 0.74 to 2.70 mg/L and influent orthophosphate concentrations ranged from 0.35 to 2.42 mg/L. The 2020 field filter removed 54% of total phosphorus and 45% of orthophosphates from the influent stormwater. Storm events with higher total phosphorus and orthophosphate influent concentrations led to greater removal than those with low concentrations. The July 21 event had the highest average removal of 58% for total phosphate, and all influent concentrations exceeded 2 mg/L. Like total phosphorus, the July 21 event had the highest removal of orthophosphate due to the highest influent concentration. 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 3.16 Total phosphate removal in 2020 field stormwater filtration study. (A) 07/21/2020, (B) 08/09/2020, (C) 08/30/2020, (D) 09/07/2020

Figure 3.17 Orthophosphate removal in 2020 field stormwater filtration study. (A) 07/21/2020, (B) 08/09/2020, (C) 08/30/2020, (D) 09/07/2020

Figure 3.18 shows the influent and effluent concentrations of dissolved iron for the sampled storm events in 2020. As expected, there was an increase in the dissolved iron concentrations in the filter effluent. Effluent iron concentrations in 2020 ranged from 0.184 to 0.836 mg/L, which were higher than the 2019 data. This may be due to the oxidation process of the steel chip surface. Despite this, all effluent concentrations measured do not exceed the EPA recommendation for recreational water of 1.0 mg/L (EPA, 1986).

Figure 3.18 Iron concentrations in 2020 field stormwater filtration study. (A) 07/21/2020, (B) 08/09/2020, (C) 08/30/2020, (D) 09/07/2020

Figure 3.19 presents the nitrate concentration in the filter influent and effluent samples. An average of 8.0% of nitrate was removed over four storm events during the 2020 field filtration study. The nitrate data suggest that only limited effluent samples showed reductions in nitrate, while others did not show apparent nitrate removal. This shows that the steel byproduct filter used in this study is not effective at nitrate removal. A batch study showed that modified steel slag could remove 20% 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. A column study on steel chips 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 have nitrate removal potential, their experimental conditions are very different from the stormwater field filter. Their studies used much higher nitrate concentrations and reaction times. Therefore, the nitrate removal by steel products would likely vary based on concentrations and reaction times.

Figure 3.19 Nitrate removal in 2020 field stormwater filtration study. (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. As shown in Figure 3.20, an average of 46% of total nitrogen was removed over four storm events sampled. It is possible that the steel byproduct filter more effectively removed organic nitrogen compared with nitrate. Detailed analysis of the removal of different nitrogen species would be necessary to elucidate the removal of different components of total nitrogen.

Figure 3.20 Total nitrogen removal in 2020 field stormwater filtration study. (A) 07/21/2020, (B) 08/09/2020, (C) 08/30/2020, (D) 09/07/2020

3.6.3 2021 Field Filtration Study

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. It was observed that the material agglomeration issue was significantly improved at this ratio. Figure 3.21 shows influent and effluent E. coli concentrations during the four storm events. Influent concentrations ranged from 1,230 MPN/100 mL to 10,460 MPN/100 mL. An average of 30% of E. coli was removed. The average removal of E. coli declined from 2019 and 2020, which may be attributed to the lower amount of steel chips in the filter.

Figure 3.21 E. coli removal in 2021 field stormwater filtration study. (A) 05/25/2021, (B) 07/10/2021, (C) 07/11/2021, (D) 07/25/2021

In 2021, a reduction in the percentage of chips in the filter also led to a substantially lower average dissolved iron concentrations in the filter effluent. As shown in Figure 3.22, effluent iron concentrations ranged from 0.013 to 0.080 mg/L, and the average net increase in dissolved iron was 0.029 mg/L. The decrease in effluent iron concentrations is due to the modification of the filter to 30% chips and 70% slag. Slag is comprised primarily of limestone and silica, whereas steel chips are predominately iron (Nippon Slag Association, 2003). All dissolved iron effluent concentrations in 2021 were significantly below the EPA's recommended limit of 1.0 mg/L (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 3.22 Iron concentrations in 2021 field stormwater filtration study. (A) 05/25/2021, (B) 07/10/2021, (C) 07/11/2021, (D) 07/25/2021

Figure 3.23 shows the filter influent and effluent orthophosphate concentrations. During four storm events, an average of 58% of orthophosphate was removed. The highest average removal of the individual events was during the first rainfall event on May 25, where an average of 77% was removed. This may be due to the addition of new materials to change the steel byproduct ratio of the filter. After the first event, the field filter still consistently removed orthophosphates but at lower percentages.

Figure 3.23 Orthophosphate removal in 2021 field stormwater filtration study. (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% removal. This indicates other species of phosphorus like organic phosphate and polyphosphate are not as readily removed. Figure 3.24 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 25, similarly to the orthophosphates.

Figure 3.24 Total phosphorus removal in 2021 field stormwater filtration study. (A) 05/25/2021, (B) 07/10/2021, (C) 07/11/2021, (D) 07/25/2021

On average, 7.6% of nitrate was removed by the 2021 filter over two stormwater samples tested. Figure 3.25 shows the influent and effluent concentrations of nitrate during these events. The stormwater had relatively low nitrate, not exceeding 0.5 mg/L as N. The nitrate removal efficiencies in 2021 were consistent with previous years.

Figure 3.25 Nitrate removal in 2021 field stormwater filtration study. (A) 05/25/2021, (B) 07/11/2021

Figure 3.26 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 other nitrogen species such as organic nitrogen are better removed by the steel byproduct filter than nitrate. The May 25 event had significantly high total influent total nitrogen concentrations, ranging from 2.3 to 2.6 mg/L as N, while the July 10 event had concentrations ranging from 0.3 to 1.0 mg/L as N. Despite this, both events removed similar amounts of total nitrogen from the water. The May 25 event removed concentrations ranging from 0.4 to 0.8 mg/L as N, while the July 10 event removed 0.2 to 0.5 mg/L.

Figure 3.26 Total nitrogen removal in 2021 field stormwater filtration study. (A) 05/25/2021 (B) 07/11/2021

3.6.4 Comparison of 2019—**2021 Field Data**

Figure 3.27 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 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 two-year period the performance of the field filter remains the same. This indicates that the steel byproduct filter can be used for long-term applications.

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 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 3.27 Pilot scale steel byproduct filter performance from 2019-2021. (A) Average E. coli removal, (B) average total phosphorus removal, (C) average orthophosphate removal, (D) average net iron increase

4. CONLUSIONS AND RECOMMENDATIONS

This study was performed to examine the long-term capability of steel byproducts in field filters to remove E. coli and other common contaminants from stormwater. Laboratory batch and column experiments were conducted to evaluate the E. coli removal capabilities of new and aged steel chips and steel slag under controlled conditions. The results of the adsorption kinetics experiments showed that steel chips removed more than 99% of E. coli after 24-hour adsorption time, while steel slag removed between 46% and 73%. New steel ships showed faster adsorption kinetics than aged steel chips. However, aged slag exhibited better E. coli removal efficiencies than new steel slag. Similar results were also observed during the continuous flow column experiments. Steel chip column reactors removed an average of 83% and 45% of E. coli for new and aged material, respectively. Steel slag column reactors removed 25% and 18% of E. coli for aged and new material, respectively. Additional column experiments were also performed to evaluate the impact of varying ratios of steel slag to chips in a mixture on E. coli removal. The results showed that ratios of 5%, 10%, 20%, and 50% steel chips removed an average of 35%, 50%, 57%, and 62% of E. coli, respectively.

After the laboratory batch studies, field filtration studies were performed to determine the removal efficiencies of E. coli and other contaminants from stormwater. The field filtration studies were conducted using an existing steel byproduct filter installed at a residential stormwater detention pond in the City of Brookings. Filter influent and effluent samples were collected from this 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 an 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 30%, 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 of the lab and field studies demonstrate that recycled steel byproducts are effective filter materials for long-term field application for E. coli removal. The steel byproduct filter is also capable of removing phosphate from stormwater. Steel chips showed higher E. coli removal capacity than steel slag. The ratio of steel chips to steel slag is a critical factor determining the E. coli removal efficiencies. Higher ratios of steel chips in the steel byproduct filter resulted in higher E. coli removal. However, more quantities of steel chips in the filter may cause material agglomeration during filter operation. Therefore, the steel chips to steel slag ratio should be carefully selected to minimize the material agglomeration while maintaining effective E. coli removal. The field filter operation experience from this study indicates that substantial material agglomeration occurred when the steel chips ratios exceeded 50%. When the steel chips ratio was adjusted to 30%, the material agglomeration issue was significantly improved. Therefore, we recommend that the steel chips to steel slag ratios should not exceed 30% for field scale applications. In addition, the filter detention time can be increased to improve the E. coli removal efficiencies. More fullscale filter studies should be performed to determine the optimum steel chips to steel slag ratios for stormwater treatment.

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