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Research article

# Exploratory study on modification of sludge-based activated carbon for nutrient removal from stormwater runoff

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ABSTRACT

Nutrients (P, N) in stormwater runoff are a major cause of eutrophication and algal blooms. A promising solution to this problem is to amend the rain garden growing medium (RGGM) with sewage sludge-based activated carbon (SBAC). To optimize the SBAC production process, different metals, pyrolysis conditions (temperature, heating time, carrier gas), and post-treatments were explored. When pyrolyzed at 400 °C for two hours, Zn-activated SBAC removed up to 41% of $PO_4$-P (initial concentration of 1 mg/L) and 72% of $NO_3$-N (initial concentration of 2 mg/L), at a dose of 1 g sorbent/L of nutrient-spiked distilled water. When the same dosage was applied to stormwater leachate made from RGGM and spiked with nutrients, the removal efficiencies were reduced to 20% for $PO_4$-P and 38% for $NO_3$-N. These reductions were probably caused by competition from other leachate components. Increasing the dosage to 3 g/L leachate improved $PO_4$-P removal to 31% and $NO_3$-N to 72%, while also resulting in the removal of 46% of total organic carbon. The major energy cost of producing such sorbents is estimated to be ~$0.76 CAD/kg SBAC.

## 1. Introduction

Pollution of urban runoff and its effect on receiving waters is a global concern (Paul and Meyer, 2001; Walsh, 2000). Pollutants of concern in stormwater include total suspended solids (TSS), nutrients (P, N), metals (copper, lead, and zinc), and organic compounds (polycyclic aromatic hydrocarbons, PAHs), which originate from landscaping, transportation, construction and other human activities. Metals and organic pollutants show both acute and long-term toxic effects on aquatic life, whereas increased nutrient concentrations can potentially lead to eutrophication and algal blooms. Nutrients in stormwater from urban areas mainly originate from application of fertilisers and deposition of vehicle emissions (Khwanboonbumpen, 2006), trees and streets (Janke et al., 2017; Ray, 1997). Without treatment, urban stormwater, as a non-point source, can have a significant negative impact on our receiving environment. In addition, urban stormwater runoff represents one of the most important and complicated challenges because urban stormwater is chemically complex (Amiard-Triquet et al., 2015).

Grover (1989) and Correll (1998, 1981) reported that nutrients such as total phosphate (TP) concentrations as low as 0.02 mg/L are sufficient to cause eutrophication. Average concentrations of nutrients in urban stormwater were reported to range from TP 0.263 mg/L, NOx-N ($NO_3$-N + $NO_2$-N) 0.558 mg/L (USEPA, 1983) high enough to trigger eutrophication. Other researchers (Lee and Bang, 2000; Reddy et al., 2014) have reported $PO_4$-P concentrations up to 0.5 mg/L and $NO_3$-N up to 1 mg/L in stormwater.

Currently in North America, there are no consistent regulations on nutrient discharge from either municipal wastewater treatment plants (WWTPs) (point-sources) or urban/agricultural runoff (non-point sources). Nutrient control is mainly based on designated uses of individual water bodies and required water quality standards (USEPA, 2010). To meet these standards, the U.S.Clean Water Act requires states to develop total maximum daily loads (TMDLs) for impaired water bodies, and then allocates the TMDLs to different sources. TMDL application is required by the Clean Water Act in the U.S. (USEPA, 2010). The National Pollutant Discharge Elimination System (NPDES) regulates the discharge from municipal WWTPs, but it does not always include effluent limits for "non-conventional" pollutants such as nitrogen and phosphorus. When the NPDES permits do include nutrient limits, they often set maximum values of 10 mg/L for TN and 1.0 mg/L for TP, with lower values (TN of 5.0 or 3.0 mg/L and TP of 0.5, or even 0.1 mg/L) becoming more common (USEPA, 2010). For municipal WWTPs (i.e., point sources), the United States regulates nutrient through the National Pollutant Discharge Elimination System (NPDES), whereas nutrient regulations are site-specific and may apply to sensitive

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and/or impaired water bodies. For these water bodies, the total maximum daily loads (TMDLs) are used to control the total nutrient discharge from both point sources (i.e., WWTPs) and non-point sources (i.e., runoffs). In Canada, regulations on nutrient discharge from WWTPs are regional and site-specific. However, for stormwater treatment practices, it might be more practical to either apply the total maximum daily loads (TMDLs) program (TP effluent $< 0.1$ mg/L), or utilize a guideline that captures at least 80% TSS and 40% TP on average annually (MOE, 2003).

Nutrient removal from stormwater is challenging because of the relatively low nutrient concentrations in, and the non-point-source nature of, urban runoff. Common facilities used to manage stormwater runoff include wet detention ponds, constructed wetlands and infiltration systems (Field et al., 1994). Designed mainly for hydraulic control of runoff (and sometimes also removal of TSS), they are not very effective in removing other pollutants. Rain gardens are increasingly being adopted in urban areas to mitigate urban stormwater impacts to the aquatic environment. A rain garden infiltration system may better remove various pollutants, including nutrients, through filtration, sorption, and ion exchange, especially when the rain garden growing medium (RGGM) is amended with effective sorbents. In addition, rain gardens can be flexibly installed at roadsides and parking lots. However, rain gardens to date have not been able to remove nutrients from stormwater effectively. This study focuses on waste-to-resource use for nutrient removal from rain gardens in urban areas before drainage enters the receiving environment.

Various sorbents have been synthesized to remove nutrients from wastewater. Sorbents can be roughly categorized as conventional (e.g., activated carbon, zeolite) and non-conventional (e.g., biosorbents or biowastes) (Chen et al., 2013; Ge et al., 2013; Gray et al., 2015; Penn et al., 2011). The latter group is attractive because their raw materials are inexpensive and readily available. For example, Zhang et al. (2012) synthesized five Mg-activated biosorbents (based on sugar beet tailings, sugarcane bagasse, cottonwoods, pine woods, and peanut shells) at 600 °C for one hour, then crushed and sieved them to retain the 0.5–1 mm fraction, and washed them with deionized water as a post-treatment to remove impurities. They reported removal efficiencies of phosphate from 0.5% to 66.7% and those of nitrate from 3.6% to 11.7%. Both nutrients were spiked to an initial concentration of 20 mg/L, while the sorbent dose was 1:500 (0.1 g sorbent: 50 mL solution).

Sewage sludge from WWTPs is a bio-waste that can be used to produce sludge-based activated carbon (SBAC). Because of its many contaminants (Hamid and Li, 2016; Kim et al., 2017; McBride, 2003) and despite its considerable nutrient content (Martinez et al., 2003), the sludge is of major environmental concern. The concept of using sludge to produce activated carbon is particularly attractive as it would assist in disposing of an inevitable by-product from the wastewater treatment process that requires continual disposal. The process to convert municipal sewage sludge to SBAC eliminates the negative environmental impacts from sludge disposal. Currently sludge disposal accounts for 50–60% of the total cost of operating a WWTP (Davis and Hall, 1997). The practice of spreading sludge on agricultural land has recently been discontinued in many jurisdictions, making the disposal problem more urgent. In addition, the savings of being able to convert the sludge into a useful product could significantly offset the energy costs of dewatering and drying sludge.

Several papers (Hadi et al., 2015; Smith et al., 2009; Xu et al., 2015) have reviewed the production, adsorption mechanism, and applications of sewage-based sorbents. Based on these reviews, chemical activation temperatures varied between 300 °C and 1000 °C, and time from 2 to 180 min. The reviews also showed that it is common practice to apply acid (HCl) washing post-treatment after chemical activation when $ZnCl_2$ is the activator. The treated pollutants included mainly organics (e.g., dyes, phenolic compounds) and heavy metals. Only one paper (Yu and Zhong, 2006), mentioned by Smith et al. (2009), reported the removal of phosphate from urban wastewater (19.7 mg P/L) by sewage sludge-based adsorbents. Removal of 96.6% was reported with adsorbent dosage ranging from 0.1 to 0.9 g per 100 mL of solution.

With the focus on waste-to-resource by converting sewage sludge to activated carbon (sludge-based activated carbon), the nutrients and other contaminants in the sewage sludge can be stabilized (Gong, 2013), while the SBAC acts as a sorbent. Previous studies by our research group focused on removal of heavy metals (Gong, 2013) and hydrophobic organic compounds (HOCs), frequently detected in stormwater using SBACs (Björklund and Li, 2017). The present study builds on this earlier work. The goal is to enhance nutrient removal by incorporating SBAC as a sorbent in rain gardens for urban areas to mitigate urban stormwater impacts to the aquatic environment. We know of no published research that has studied nutrient removal from stormwater using SBACs alone. The general objective of this exploratory study was to modify SBACs, based on the findings of Gong (2013) and explore the feasibility of using these modified SBACs as an additive in the growing medium of rain gardens to improve stormwater quality. Specific objectives included: (1) Different metals (Zn, Mg, and Ca) as chemical activators are explored. (2) Pyrolysis conditions, including pyrolysis temperature, heating duration, and carrier gas, are optimized. In an attempt to reduce the energy consumption, based on literature reviews and preliminary studies, we started from the lowest temperature (i.e., 300 °C) reported in the literature, then increased it to 400 °C in an attempt to produce an effective sorbent and reduce energy consumption in achieving waste-to-resource. Pyrolysis steady state temperature was then held at 400 °C for 1–2 h, and carrier gases were $CO_2$ and $N_2$. (3) Adsorption capacity is determined for single-nutrient and mixed-nutrients, and competition effects are analyzed. (4) Kinetics are studied to provide practical information for the design of rain gardens. The selection of the time for the kinetic adsorption tests is based on the field configuration of rain gardens in consultation with practical engineers. (5) pH effects are investigated, with adsorption assessed using simulated RGGM leachate. (6) SBAC production costs are also estimated.

## 2. Materials and method

### 2.1. Sources of materials

Municipal sewage sludge was obtained from a local full-scale WWTP in British Columbia, whose location cannot be named. The sludge used was waste activated sludge (WAS) from a trickling bed filter. This sludge contained 3.9% (w/w) TS, 3.4% VS, ammonia 617 mg/L, and nitrate 8.33 mg/L, with a pH of 5.7. The detailed characteristics of the studied sludge were given by Ahmad et al. (2016).

RGGM is the soil commonly used for rain gardens. The soil is marketed under the name Cascade Ecomedia and was obtained from Yardworks (Veratec, Chilliwack, BC, Canada) developed by Cascade Envirotech (Aldergrove, BC, Canada). The RGGM obtained for this study was from a 1.5-year-old rain garden that receives runoff from a 400 $m^2$ parking lot. It was sieved to particle size $< 2$ µm.

The stormwater was collected in December 2017 during the rainy season from the fore bay of a wetland (Lost Lagoon) built to treat contaminated runoff from the Stanley Park Causeway, a major commuter route through Stanley Park, Vancouver.

### 2.2. Reagents

The nutrients ($PO_4^{3-}$, $NO_3^-$ or $NH_4^+$) spiked in (a) distilled water (simple solute) and (b) stormwater (complexed background) are $KH_2PO_4$ from Fisher Scientific Canada at purity grade $> 99.5\%$; $NH_4Cl$ from Fisher Scientific Canada at purity grade $> 99.5\%$ and $NaNO_3$ from Sigma Aldrich with purity grade $> 99.0\%$.