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Enhanced biofiltration of O&G produced water comparing granular activated carbon and nutrients

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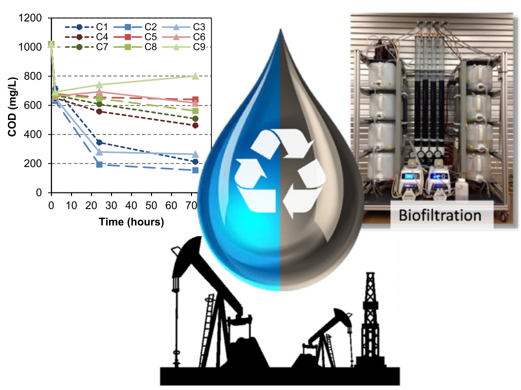
HIGHLIGHTS

- The biofiltration system removed over 92% DOC and 81% COD from PW in 24 h.
- GAC type and condition impact BAF performance and nutrient addition had minor impact.
- Bioactivity and biofilm development on GAC correlated to BAF performance.
- Microorganisms indigenous to PW consistently seeded the biofilters.
- Biologically active GAC removed more organic matter than new GAC.

GRAPHICAL ABSTRACT



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ABSTRACT

Large volumes of water are required for the development of unconventional oil and gas (O&G) wells. Water scarcity coupled with seismicity induced by deep-well disposal promote new O&G wastewater management strategies, specifically treatment and reuse. One technology that has been proven effective for removal of organic matter and solids is biologically active filtration (BAF) with granular active carbon (GAC); however, further optimization is needed to enhance BAF performance. This study evaluated three GAC media (one spent and two new) and two nutrient-mix supplements for enhanced removal of chemical oxygen demand (COD) and dissolved organic carbon (DOC). Biofilm development was also monitored and correlated to BAF performance. The spent GAC with extant biofilm quickly acclimated to PW and demonstrated up to 92% DOC removal (81% COD) in 24 h, while little impact by nutrient addition was observed. In addition, virgin GAC was slow to establish a biofilm, indicating that appropriate GAC selection and pre-developed biofilm is critical for efficient BAF performance. Furthermore, the production of high quality BAF effluent (less than 20 mg/L DOC) presents the opportunity to apply BAF as a pretreatment for subsequent desalination—expanding the potential for reuse applications of PW.

1. Introduction

Increased demands on fresh water resources due to drought conditions and continued population, industrial, and agricultural growth is stressing water supplies; yet, it is spurring innovative approaches and

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novel technologies to treat and reuse impaired waters. The seemingly high volumes of fresh water used and consumed during the development of unconventional oil and gas (O&G) wells has especially garnered public attention—an average of 2.5 million gallons (~10,000 m^3) of water is used to hydraulically fracture a single well (Freyman, 2014). 10–40% of the water used to fracture a well returns to the surface in the first ten days of production as fracturing flowback, before transitioning to produced water (PW) (Torres et al., 2016). Most O&G wastewater is disposed of in Class II injection wells (i.e., disposal wells) because it is typically the most effective and economical management option, with the lowest risk and energy requirement (Mantell, 2011; Puder & Veil, 2006). Other common management techniques include onsite evaporation ponds, treatment for reuse (e.g., hydraulic fracturing, dust suppression, road stabilization, crop irrigation), or surface discharge (Torres et al., 2016).

While management of PW varies with location, treatment for subsequent reuse applications has often been limited to basins with limited access to disposal wells (e.g., Marcellus region) or extensive pipeline infrastructure for water conveyance (e.g., Woodford) (Mantell, 2011; Strong et al., 2017). Although the economics and logistics of treatment and reuse are currently unfavorable for O&G operators in many regions, environmental concerns with deep-well injection (e.g., seismicity, groundwater contamination), legislation restricting PW disposal volumes, and growing interest of the O&G industry to improve best management practices and public relations is altering the O&G water paradigm to encourage reuse (Weingarten et al., 2015).

Many physical, chemical, and biological unit processes are viable for treatment of PW, but the high and variable concentrations of total dissolved solids (TDS) (can reach more than 100,000 mg/L), metals (e.g., bromide, calcium, magnesium, iron, manganese, silica, strontium), and complex organic constituents (e.g., BTEX, oil and grease, hydrocarbons, biopolymers, humic substances) present unique challenges to most technologies (Benko & Drewes, 2008; Rosenblum et al., 2017). For example, TDS is most often removed using physical methods (e.g., membranes or distillation/evaporation); however, organic matter must be removed first to minimize fouling of membranes or other surfaces. Accordingly, organic matter can be removed using chemical (e.g., coagulation, ozonation) or biological methods (e.g., biofiltration, activated sludge), but various organic and inorganic constituents can interfere with chemical treatment and reduce efficacy or form harmful byproducts (Luek & Gonsior, 2017; Maguire-Boyle & Barron, 2014). Furthermore, TDS concentrations greater than 1% are known to adversely impact biological activity and even inhibit biological growth, challenging the use of biological technologies for PW treatment (Lefebvre & Moletta, 2006; Lester et al., 2015). Thus, deliberate strategy must be implemented for efficient and cost-effective removal of targeted constituents from PW to enable broad water reuse.

One technology that has proven effective at the bench-, lab-, and pilot-scale for removal of organic matter from O&G waste streams is biologically active filtration (BAF) with granular active carbon (GAC). The first study in the series was conducted by Freedman et al. and utilized spent GAC with an existing biofilm (GAC 816; from surface water treatment) (2017). Batch experiments were conducted at the bench-scale (1.3 cm diameter BAF columns) and demonstrated increased DOC removal efficiencies with decreasing dilutions of PW feed over time, respectively. Over 93% DOC removal from undiluted PW was achieved with constantly aerated biofilters, and the roles of adsorption, biodegradation, and air stripping for DOC removal were isolated. Columns operated with PW pretreated by coagulation or operated without aeration showed reduced DOC removal, suggesting that aeration alone is adequate and chemical costs are minimal with BAF.

Following successful bench-scale demonstration, Freedman et al. scaled BAF up to the lab-scale with 5 cm diameter columns (2017). Similar experiments were performed, including a set of experiments evaluating DOC removal with temperature variation (14–25 °C). Lab-scale BAF results without an acclimation period (i.e., no PW dilutions) showed similar DOC removal trends to the bench-scale system—up to 95% DOC removal was demonstrated in 72 h, while increased operating temperature improved the rate of biodegradation (Freedman et al., 2017). This study confirmed the ability of BAF to effectively remove organic matter from raw PW and suggests the ability of microorganisms to quickly adapt to high salinity streams (from surface water).

Riley et al. also examined the flexibility of BAF to treat challenging O&G streams using the same lab-scale BAF system with three varying quality PW and fracturing flowback wastewaters (2016). The wastewaters tested varied from 12,600–31,200 mg/L TDS and 36–732 mg/L DOC, respectively. BAF experiments operated with spent GAC 816 media in batch and continuous configurations reached over 90% DOC removal (~85% chemical oxygen demand (COD)) in only 24 h, serving as a benchmark for future BAF and PW studies. Furthermore, this study showed the effectiveness of BAF as a pretreatment for desalination, as subsequent ultrafiltration and nanofiltration (NF) using the BAF-treated PW revealed minimal membrane fouling and high TDS and DOC rejection (e.g., NF permeate of 900 mg/L TDS and 1.4 mg/L DOC) (Riley et al., 2016).

Following the previous bench- and lab-scale BAF studies with PW and fracturing flowback (Freedman et al., 2017; Riley et al., 2016), Riley et al. evaluated scalability and long-term performance (i.e., organic matter removal, microorganism development) with a pilot-scale BAF system (2018b). Operation for ~600 days with new GAC achieved up to 85% COD removal in 100 h of batch treatment (PW batches replaced weekly). However, analysis of the GAC showed significant loss of adsorption capacity, which was reflected in decreased COD removal following ~320 days. This study revealed the importance of GAC selection and suggested further investigation of various media for optimal BAF performance.

Several design and operating parameters impacting the removal of organic matter by biofiltration have been identified and examined in previous studies, including the filter media, empty bed contact time (EBCT), organic loading rate, nutrient addition, backwashing frequency, disinfectant in backwash water, aeration, temperature, and microorganisms (Chaudhary et al., 2003; Chowdhury, 2013; Terry & Summers, 2018). Two parameters that may substantially impact the removal of organic matter from PW by BAF and that were not thoroughly investigated in previous studies by Freedman et al. and Riley et al. include filter media type and nutrient addition. The enhanced performance of biological activated carbon (BAC) filters compared to inert and non-adsorptive media like anthracite and sand is well documented in the literature, with GAC biofilters (e.g., BAC) consistently resulting in higher removal of organic matter (Basu et al., 2016; Reaume et al., 2015; Wang, 1995). This is attributed to its high porosity and surface area that is conducive to biological attachment. However, fewer studies have compared the effects of GAC properties and types for removal of organic matter. One study comparing steam activated and chemically activated GAC showed that the steam activated GAC exhibited the highest DOC biodegradation, potentially due to its higher adsorption capacity of DOC (leading to biodegradation of adsorbed compounds) than the chemically activated GAC (Yapsakli & Çeçen, 2010). Other BAC studies focused on nutrient availability within the biofiltration system, improving performance with nitrogen and phosphorus supplementation (Basu et al., 2016; Lauderdale et al., 2012; Liu et al., 2017). Because PW used in the previous and current BAF studies is nutrient limited, especially phosphorus, it should be further investigated as a method to improve BAF performance.

Therefore, the main objective of this research was to improve the removal of organic matter beyond the previous studies conducted by Freedman et al. and Riley et al., particularly bettering 90% DOC removal in 24 h. This was accomplished through comparison of two new GAC media types that have not been tested in this application (but are commonly used by municipalities/utilities for biofiltration) to the previously tested GAC 816 with an existing biofilm. Two nutrient solutions were also tested with each GAC type. The primary parameters used to assess