Organic Matter Removal via Biological Drinking Water Filters: Removal Efficiency based on Quantifiable System Factors

by

Leigh Gilmore Terry
B.S., University of Alabama, 2012
M.S., University of Colorado, Boulder, 2013

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This thesis entitled:

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written by Leigh Gilmore Terry
has been approved for the
Department of Civil, Environmental, and Architectural Engineering

_____
R. Scott Summers, University of Colorado, Boulder, Chair

_____
M. Robin Collins, University of New Hampshire

_____
Sherri M. Cook, University of Colorado, Boulder

_____
Mark Hernandez, University of Colorado, Boulder

_____
JoAnn Silverstein, University of Colorado, Boulder

Date_____

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ABSTRACT

Terry, Leigh Gilmore (Ph.D. Environmental Engineering)

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Thesis directed by R. Scott Summers, Professor

Biodegradable organic matter (BOM), found in all surface waters, is a challenge for drinking water utilities as it can lead to distribution system bio-regrowth, react to form disinfection by-products, or be a specific compound of concern. Drinking water utilities face the challenge of removing BOM to meet increasingly stringent regulations, often at higher costs and operational complexity. Biofiltration can be an efficient treatment technology to remove BOM from the influent water, but should be optimized to achieve maximum removal performance.

The objectives of this dissertation were to evaluate and model the impacts of biologically active filter (biofilter) design and operation on BOM removal as measured by dissolved organic carbon (DOC). Operational and water quality parameters, i.e. extended empty bed contact time (EBCT), temperature, biomass acclimation and distribution, and natural organic matter concentration and origin (microbial, terrestrial and wastewater effluent), were evaluated to determine impacts on biofilter performance. A novel bench scale methodology was developed in Chapter 2 that incorporated a batch reactor and a single-pass flow through reactor that allowed arduous pilot scale experiments to be replaced with streamlined bench scale testing, which could expedite biofilter implementation in drinking water utilities. In Chapter 3, a model derived from Monod kinetics was developed for biological filters based on EBCT and a single biomass measurement from the top of the filter. The model was developed for the control of DOC and successfully applied to predict DOC removal. Biomass activity, adenosine triphosphate (ATP),

measurements were a direct function of temperature, yet biomass concentration, phospholipid measurements, were not a function of temperature in the range of 5 °C to 22°C. Pilot scale work in Chapter 4 found acclimation of the 'fresh' media in terms of DOC removal and activity occurred over a two-month time frame. Chapter 4 and Chapter 5 found extended EBCT of a biofilter and higher temperatures improved the performance of biofilters for controlling DOC, yet influent DOC did not impact DOC removal directly. Biomass activity, ATP, was highest at the top of the filter and decreased with increasing filter depth. Chapter 5 bench scale work found biofilters were robust in removing DOC from microbial, terrestrial and wastewater effluent sources and reduced DBP precursors. In chapter 6, a life cycle assessment model was used to compare conventional filtration and biofiltration. Biofiltration had lower environmental impacts than conventional filtration for average U.S. source waters by about 25%. Chemicals, in particular alum and caustic soda, had the largest contributions to environmental impacts. The most effective way to substantially decrease negative environmental impacts of either filtration system is to optimize chemical doses. Higher temperatures can support greater DOC biodegradation, which increases the environmental benefits of biofiltration, and higher levels of biodegradation can also be achieved at lower temperatures when biofilter parameters are optimized.

DEDICATION

To my parents who have constantly instilled in me the importance of hard work, the love of learning, the pursuit of dreams, and joy through the process.

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Biodegradable Organic Matter and Rapid-Rate Biofilter Performance: A Review

1.1 Introduction

Dissolved organic matter (OM) found in drinking water sources includes a biodegradable fraction (BOM) and a non-biodegradable fraction. The difference between them is operationally defined, based on the BOM characterization method, including aerobic and anoxic methods. Drinking water utilities are concerned about controlling BOM as it: a) uniquely provides a carbon source for microbial regrowth in the distribution system, b) can react with disinfectants to yield disinfection by products (DBPs) and c) can be specific compounds that are considered contaminants (Volk et al., 1997a; LeChevallier, 2014). Regrowth of bacteria can accelerate corrosion, lower hydraulic capacity, and make it difficult to maintain a disinfection residual in the distribution system (Kaplan and Volk, 1994). BOM provides energy and carbon for the metabolism of heterotrophic bacteria (Volk et al., 1997c).

BOM, to varying levels, can be found in all drinking water sources, including groundwaters, independent of their origin, e.g., natural or anthropogenic OM. BOM is also created in a treatment plant after oxidant/disinfectant addition, especially by ozone and to lesser extent by chlorine. The BOM fraction includes identifiable specific biodegradable organic compounds, e.g., formaldehyde, geosmin, and as well compounds that are not identifiable, e.g., biodegradable humic substances. Again, those differences are operationally defined, based on the specific compound analysis methods utilized. Biodegradable organic carbon (BDOC) and assimilable organic carbon (AOC) are the most commonly used parameters to measure BOM

under aerobic conditions (Servais et al., 2005). Many specific compounds are not biodegradable under the conditions of aerobic drinking water biofilters, while others can be completely removed (Zearley and Summers, 2012; Hallé et al., 2015). An extensive evaluation of BOM occurrence in drinking water sources with and without ozonation has not been reported.

Filters which remove BOM via biological mechanisms, biofilters, can be operationally defined as filters in which there is no disinfectant residual in the effluent, and include bank filtration systems, slow sand filters and rapid-rate biofilters. This review is limited to rapid-rate biofilters. These filters may utilize a range of media including sand, anthracite, and granular activated carbon (GAC). Disinfectant residuals are readily reduced by GAC in the top portion of a filter bed, and as such all GAC filters are biologically active. While many biofilters are incidentally operated, additional performance may be achievable, both higher levels of removal and additional compounds removed, when the design and operation of biofilters are optimized (Kaplan et al., 1994; Basu et al., 2015; Selbes et al., 2017). While biofilter performance results have been reported (Zhu et al., 2010; Juhna and Melin, 2006), the impact of design and operational parameters examined (Basu et al., 2015; Brown et al., 2016), and models developed (Servais et al., 2005), a comprehensive overview and performance meta-analysis are not available.

The objectives of this paper are to review: a) the levels of BOM in drinking water sources with and without ozonation as assessed by AOC and BDOC and b) rapid-rate biofilter performance as assessed by the removal of BDOC. This review is limited to aerobic biological conditions focusing on surface waters and does not explicitly include specific biodegradable organic compounds. This review builds on earlier reviews by Urfer et al. (1997), Servais et al. (2005), Zhu et al. (2010), Wang et al. (2014), Basu et al. (2015) and Brown et al., (2016).

1.2 Results and Discussion

1.2.1 Biodegradable Organic Matter Occurrence Review

The suite of biodegradable compounds that comprise BOM included humic substances, amino acids, carbohydrates and the ozonated by-products (OBPs) aldehydes and ketoacids (Kaplan et al., 1980; Lytle and Perdue, 1981; Sweet and Perdue, 1982; Kaplan and Bott, 1983; Kaplan et al., 2005). Volk et al. (1997) found streamwater DOC was comprised of 72-78% humic substances, 9-16% carbohydrates (mostly polysaccharides), 2-3% amino acids and 15-22% OM with a molecular weight > 100 kDa. For this source, the BDOC represented 21–34% of the DOC and was comprised of 68-85% humic substances, 22–43% carbohydrates (mostly polysaccharides), 2-6% amino acids and 33-45% OM > 100 kDa. A portion of BOM is collectively measured by AOC, which measures growth of a microbial inoculum, and a larger BOM portion by BDOC measurement, which measures changes in organic carbon content of sample due to microbial metabolism (Huck, 1990; Kaplan et al., 2005). AOC is the portion of BDOC that is most readily used by bacteria and converted to cell mass, while BDOC is the portion of organic carbon that is biodegraded by heterotrophic microorganisms for 1) energy production and transformed into CO_2 or other organic substances or 2) for biomass growth (Huck, 1990; Escobar and Randall 2001b; Juhna and Melin, 2006; Wang et al., 2014). AOC and BDOC measurements provide complementary information and both are important indicators for water biostability (Volk et al., 1994; Charnock and Kjonno, 2000; Escobar and Randall 2001a; Wang et al., 2014). For instance, Huck (1990) suggested that AOC, bacterial biomass, should be measured when determining bacterial regrowth or growth of coliforms, and BDOC, change in organic carbon concentration, should be measured when determining decrease in chlorine demand or DBP formation potential.

Influent source water BOM varies over time, including seasonal variations, as a result of natural and anthropogenic events, i.e., floods, droughts, algal blooms, snowmelt, soil run-off, seasonal changes, wastewater, industrial and agricultural discharge (Servais et al., 2005; LeChevallier, 2014). Bradford et al. (1994) found the AOC to DOC ratios were between 0.5% to 31% in surface and groundwaters, suggesting that organic carbon can range from biologically stable to very assimilable. Studies have surveyed AOC and BDOC levels in source waters, distribution system, reclaimed water treatment, and desalination plants (Kaplan et al., 1989, 1993; Bradford et al., 1994; Najm et al., 2000; Volk and LeChevallier, 2000, 2002; Wang et al., 2014). Liu et al. (2002) found AOC concentrations in different distribution systems varied due to seasonal and drinking water treatment plant operating conditions. A summary of AOC and BDOC surveys is presented in Table 1.1 demonstrates BOM variability.

Table 1.1 AOC and BDOC occurrences reported in the literature.

AOC range (µg/L)	BDOC range (mg/L)	Sample	Reference
48 to 607	-	10 sites from 2 surface waters	Kaplan et al., 1989
52 to 99	-	1 drinking water utility source over a 11-months	McEnroe et al., 1992
34 to 247	-	13 North American drinking water utilities over 10 states	Kaplan and LeChevallier, 1993
94 to 275	-	2 surface water reservoirs	Bradford et al., 1994
75 to 731	-	16 locations along a river	Bradford et al., 1994
75 to 150	0.6 to 2	1 drinking water treatment plant effluent over 1 year	Kaplan et al., 1994
-	0.2 to 2.9	a river with higher BDOC concentrations associated with storms than with baseflow	Volk et al., 1997b
0.3 to 51	0.21 to 2.79	23 Norwegian drinking water utilities' sources over 18-months	Charnock and Kjonno, 2000
50 to 250	0.05 to 0.85	1 drinking water treatment plant effluent over 1 year	Najm et al., 2000
18 to 214	-	94 North American distribution systems	Volk and LeChevallier, 2000

43 to 937	0.10 to 1.58	6 conventional drinking water utilities' sources over 10 months (surface waters)	Volk and LeChevallier, 2002
-	0.03 to 1.03	30 North American distribution systems	Volk and LeChevallier, 2002

Oxidation of organic matter will increase the biodegradable portion by reducing the size of DOM molecules and increasing the number of oxygenated functional groups (Langlais et al., 1991; Krasner et al., 1992; Volk et al., 1993; Weinberg et al., 1993; Siddiqui et al., 1997; Hozalski et al., 1999; LeChevallier, 2014). Ozone is a strong oxidant and increases AOC levels in drinking water through the conversion of aromatic, unsaturated organic structures (humic and fulvic acids) to saturated polycarbonaceous compounds of low molecular weight (van der Kooij et al., 1989; Volk et al., 1997c). In addition, ozone fosters biological growth on media by providing large amounts of oxygen to the water, which is the electron acceptor for bacteria when during DOC oxidation (Juhna and Melin, 2006). Black and Bérubé (2014) found preozonation reduced high molecular weight compounds, but contrary to the majority of the literature, the authors found preozonation did not appear to increase the biodegradability of the source water as the oxidant reacted with both the biodegradable fraction and non-biodegradable fraction equally. Other oxidants, i.e., chlorine, potassium permanganate and chlorine dioxide, also react with OM to increase the level of AOC in treated supplies (van der Kooij, 1987; LeChevallier et al., 1991a). Bacteria can readily utilize the lower molecular weight compounds for growth and energy. LeChevallier et al. (1992) found a 2-fold increase in AOC after ozonation and filter effluent AOC levels were higher in preozonated waters compared to nonozonated waters. Ozonated byproducts include aldehydes, acetones, ketoacids, aldo-ketones, and carboxylic acids and they are highly biodegradable (Krasner et al., 1993; Weinberg et al., 1993). Carlson and Amy (1998) found the composition of BDOC removed in preozonation biofiltration was 3% of aldehydes, 12% ketoacids, 13 – 15% carboxylic acids, and 70-72% unknown.

1.2.1.1 BOM Methods

The AOC bioassay method correlates the growth of a test organism, either defined inoculum or indigenous microflora, with the concentration of BOM, i.e., biomass growth (Weinrich et al., 2009; Escobar and Randall, 2001a; Wang et al., 2014; Kaplan et al. 2005). Van der Kooij et al. (1982) first proposed the density of organisms that can grow in a water sample is proportional to the AOC concentration. Since, numerous AOC methods, thoroughly described by Kaplan et al. 2005, have been reported and are summarized in Table 1.2. AOC methods use various strains of organisms (i.e. known bacteria cultures, P. *fluorescens* P17, *Aquaspirillum* NOX, etc. or indigenous bacteria) to inoculate and grow bacteria over a period of several days and weeks as a result of biodegradable carbon assimilation. The growth is measured by colony-forming units (CFU)/mL (van der Kooij et al., 1982; van der Kooij & Hijnen, 1984; Kemmy et al., 1989; Reasoner & Rice, 1989; Kaplan et al., 1993a), adenosine triphosphate (ATP) (Stanfield & Jago, 1987; LeChevallier et al., 1993), turbidity (Werner & Hambsch, 1986; Hambsch & Werner, 1993), bioluminesce (Haddix et al., 2004; Weinrich et al., 2009), cell elongation (Bradford et al., 1994), or flow cytometry (Hammes & Egli, 2005; Elhadidy et al., 2016). The measured parameter is then converted to AOC in (µg/L) using a standard factor or calibration in known solutions of sodium acetate (ug C eq. acetate/l) or other organic compounds. Advantages of the AOC bioassay methods include sensitivity, precision, standardization, and high signal-to-noise ratio; however, disadvantages include sensitivity to contamination, procedure time requirements, selective inoculum and results reported in carbon equivalents (Kaplan et al., 2005). Huck (1990), Allgeier et al. (1996), and LeChevallier (2014) provide detailed descriptions of the AOC methods.

Several authors have compared AOC methods. Huck (1990) compared the AOC results by the van der Kooij et al. method to those from Werner et al. method for sampling points

throughout a pilot plant treatment train for two different dates and found a good correlation between the methods for one date, but poor correlation on the second date, potentially due to inhibition with the van der Kooij et al. method. Huck (1990) also summarized that Reasoner and Rice (1989) found similar values between their coliform growth response assay and van der Kooij et al. AOC assay. Huck (1990) and Wang et al. (2014) and Elhadidy et al. (2016) reviewed the evolution of bacterial growth measurements, AOC assays, based on the selection of bacterial species, optimization of inoculation method and incubation conditions.

The BDOC assay method measures the DOC consumption over time by indigenous bacteria OM oxidation, in batch reactors or continuous-flow bioreactors (Escobar & Randall, 2001a; Kaplan et al., 2005). Five basic BDOC methods have been developed, as shown in Table 1.3. BDOC methods use either fixed-bed reactors with attached media in both single pass (Ribas et al., 1991; Frías et al., 1992) and recirculation (Mogren et al., 1990) modes and batch reactors mixed with attached biomass (Joret & Levi, 1986; Allgeier et al., 1996) or suspended biomass (Servais et al., 1987) as inoculation methods. The BDOC value is calculated as the difference between DOC at the beginning and end of the designed time period, or in the case of the single pass method (Ribas et al., 1991; Frías et al., 1992), the column influent and effluent. Wangersky (1993) provides a detailed overview of DOC analysis. Frías et al. (1992) expanded on Ribas et al. (1991) method by measuring a discrete water sample similar to the other methods, while Ribas et al. (1991) measured continuous water samples. The DOC remaining is the nonbiodegradable or refractory DOC. Advantages of the BDOC assay are that the results are expressed in carbon units, shorter sampling period (Ribas et al., 1991; Frias et al., 1992); however, disadvantages include potentially longer colonization periods and overall long sampling periods (Kaplan et al.,

2005). Huck (1990), Allgeier et al. (1996), and LeChevallier (2014) provide detailed descriptions of the BDOC methods.

Several authors have compared results from different BDOC methods. Frias et al. (1995) compared three different BDOC methods and found statistically similar mean percentages for all tests (Frías et al., 1992; Joret & Levi, 1985; Servais et al., 1987,1989). Allgeier et al. (1996) compared the results of four different methods; the batch shaker method (Allgeier et al., 1996), the recirculating biofilter (Mogren et al., 1990), the aerated batch reactor (Joret et al., 1987), and a 7-day suspended biomass method (versus the 28-day standard method) adapted from Servais et al. (1987). They used four different waters: untreated Ohio River water, ozonated Ohio River Water, and an isolated groundwater NOM solution at TOC concentrations of 4.14 mg/L and 17.5 mg/L. For the untreated Ohio River, the Allgeier et al., Mogren et al., and Joret et al. methods yielded similar BDOC results after 7 days (BDOC = 0.55 ± 0.03 mg/L), but the time modified Servais et al. method yield lower BDOC, (0.22 mg/L) which was attributed to the shortened incubation period. For the ozonated Ohio River, larger variations were found across the methods with an average BDOC of 0.64 ± 0.18 mg/L, but Allgeier et al. and Mogren et al. methods produced similar results (BDOC = 0.75 m/L). Similar trends were seen with the low NOM concentration water as Allgeier et al. and Mogren et al. methods yielded similar results (0.53 mg/L and 0.57 mg/L, respectively), while Joret et al. and Servais et al. methods yielded lower BDOC values (0.32 mg/L). For the high NOM concentration water, the similar trends were found as Allgeier et al., Mogren et al., and Joret et al. methods yielded similar results (BDOC = 4.06 ± 0.24 mg/L), while the time modified Servais et al. (1987) method yielded lower results (BDOC = 1.81 ± 0.04 mg/L). An average of 5 to 28% agreement was seen over all the waters for Allgeier et al., Mogren et al., and Joret et al. Block et al. (1992) proposed the lower results for

suspended bacteria BDOC method, Servais et al. (1987), could be a result of a low number of heterotrophic cells in test samples due to smaller size biomass inoculum to avoid carbonaceous contamination and thus slower DOC kinetic bioremovals. Another study, Summers et al. (1993), found good reproducibility with Mogren et al., Allgeier et al., Servais et al. and Joret and Levi methods on the same river water (95% CI was 2.1, 1.6, 2.4 and 0.95, respectively). Mogren (1990) compared her biofilter BDOC method to the Joret et al. (1987) noncirculating batch reactor method using the same acclimated sands and humic substances for Ohio River and Delaware River and found no statistical difference in the results. Block et al. (1993) compared variations between BDOC analysis using mixed bacterial populations of inoculum and found laboratories comprised 57 – 71% of variation, inoculum origin comprised 12-26% of variation, while indigenous mixed bacterial populations (suspended or attached bacteria) did not introduce excessive variability.

Frias et al. (1995) compared van der Kooji et al. (1992) AOC method to four BDOC methods and as expected, found statistically lower values for the AOC test, in terms of percent chemical oxygen demand. Prévost et al. (1989) compared the van der Kooij et al. (1982) AOC method to the Servais et al. (1997) BDOC method and also found substantially lower AOC values. This is likely due to broader diversity of microorganisms in BDOC inoculum, while the AOC method is based on two species of bacteria, and the both synthesis and respiration are assessed with the BDOC method. The different AOC methods will also lead to different AOC results due to specific inoculum and procedures; however, BDOC methods should produce similar results if employing similar inoculum (attached or suspended). Kaplan et al. (2005) summarized the following: estimates of BDOC based on suspended inoculum are lower that estimates based on attached inoculum, and estimates of BOM from BDOC methods are greater

than estimates of BOM from AOC methods. Comparison of BDOC and AOC results are not appropriate, rather AOC and BDOC assays should be treated as complementary measurements. Unfortunately, Evans et al. (2010) survey of biological drinking water treatment perceptions and actual experiences in North America found that few utilities that employ biological filtration measure AOC or BDOC due to method complexity.

Table 1.2 AOC Methods adapted from Huck (1990) and LeChevallier (2014)

Author	Water Prep	Inoculation	Incubation conditions	Measured Parameter	Result Expression
Van Der Kooij et al. (1982), Van der Kooij and Hijnen (1984)	Pasteurization	Pure Strains: P. fluorescens P17 and Spirillum, NOX (500 CFU/ml)	20 days 15℃	CFU/ml	AOC
Kaplan et al. (1993a)	Pasteurization	P. fluorescens P17 + Spirillum, NOX (500-1000 CFU/ml)	9 days 20℃	CFU/ml	AOC
LeChevallier et al. (1993)	Pasteurization	P. fluorescens P17 + Spirillum, NOX (10^4 CFU/ml)	5 days 22℃	ATP	AOC
Bradford et al. (1994)	Filtration	P. fluorescens P17	12 h 20℃	Cell elongation; Epi. Counts	AOC
Kemmy et al. (1989)	Filtration	Mixture of 4 strains: Pseudomonas fl., Curtobacterium, Corynebacterium, 1 species of coryneform type	6 days 20℃	CFU/ml	AOC
Stanfield and Jago (1987)	Filtration	Bacteria from raw water and sand filtered water	Until Max Growth 20℃	ATP	AOC
Werner and Hambsch, (1986),	Filtration	Water sample bacteria retained on the filter (5	60 h 20°C	Turbidity	Growth Yield (μ) Growth

Hambsch and Werner (1993)		10^4 cell/ml)			Factor ($\log Y/Y_0$)
Reasoner and Rice (1989)	Filtration	Enterobacter clocacae, Escherichia coli, Klebsiella oxytoca	5 days 20℃	CFU/ml	log N5/N0
Haddix et al. (2004); Weinrich et al. (2009)	Pasteurization & Insertion of *lux*CDABE luminescence	P. *fluorescens* P-17 and *Spirillum,* NOX	3-5 days 20℃	Cell Growth via Bioluminescence	AOC
Hammes and Egli (2005)	Filtration	P. *fluorescens* P-17 and natural microbial consortium	14 days 30 ℃	Cell Count via Flow cytometry	AOC
Elhadidy et al. (2016)	Heat treatment and Filtration	Indigenous bacterial community of the sample	21 days 30 ℃	Cell Count via Flow cytometry	AOC

CFU-colony-forming units, ATP-adenosine triphosphate, AOC-assimilable organic carbon

Table 1.3 BDOC Methods adapted from Huck (1990) and LeChevallier (2014)

Author	Water Prep	Inoculation	Incubation conditions	Result Expression
Servais et al. (1987)	Filtration	Batch reactor with suspended bacteria from sample's natural environment	28 days 20℃	BDOC = DOC_{intial} - DOC_{final}
Joret and Levi (1986); Joret et al. (1989)	-	Aerated batch reactor with acclimated sand	7 days 20℃	BDOC = DOC_{intial}- DOC_{final}
Mogren et al. (1990)	-	Recirculating column with acclimated sand	5 days 20℃	BDOC = DOC_{intial}- DOC_{final}
Ribas et al. (1991); Frias et al. (1992)	Filtration	Single-pass column with bacteria fixed on porous glass particles	2.5 h 20℃	BDOC= DOC_{inflow} - $DOC_{outflow}$

Allgeier et al. (1996)	-	Shaken batch reactor with acclimated sand	5 days 20℃	BDOC = DOC_{intial}-DOC_{final}

BDOC-biodegradable dissolved organic carbon, DOC-dissolved organic carbon

1.2.1.2 Occurrence Analysis

An extensive review of AOC and BDOC values in surface waters with and without ozonation was performed and a statistical analysis of the data is summarized in Tables 2 and 3, respectively. Detailed data are provided in Appendix A (Tables A1, A2, A3 and A4). Paired data, as available, were collected from each AOC and BDOC study. Some studies reported only one parameter, therefore the number of values, n, are not equal for all parameters. The ozonated waters have more AOC values than TOC, because most studies did not record TOC directly after ozonation. All AOC/TOC and BDOC/TOC ratios are only provided for paired data. All studies reported here used P17, NOX or combined P17 and NOX for the AOC assay inoculum.

For the AOC nonozonated data set (n=89), the median TOC value was 3.5 mg/L, with a range of 0.5 mg/L to 16.3 mg/L. The median AOC value was 120 µg/L with a range of 6.3 µg/L to 482 µg/L. AOC/TOC ratio varied from 0.2% to 38.3% and had a median of 2.8%. While this review focuses on surface waters, a cursory review of groundwaters was done for the same variables. Overall, groundwater OM yielded lower values for AOC/TOC ratios (n=30) as seen in Appendix A Table A5, which is consistent with other literature studies (Bradford et al., 1994; Kaplan et al., 1994; Volk and LeChevallier, 2002)

For the BDOC nonozonated data set (n=100), the median TOC value was 3.6 mg/L, with a range of 0.2 mg/L to 16.3 mg/L. The median BDOC value was 1.0 mg/L with a range of 0.1 mg/L to 7.8 mg/L. BDOC/TOC ratio ranged from 1% to 72%, with the median at 20%. This suggests that 20% of the TOC is biodegradable and can be used utilized for energy and biomass

growth. Groundwater OM yielded lower BDOC/TOC ratios (n=7), as seen in Appendix A Table A6.

The ratio of AOC to BDOC for 25 paired data sets varied from 3% to 89% and with a median of 22%. This suggest that 22% of the BDOC (3% of the TOC) is highly biodegradable and can be used for biomass growth. Escobar and Randall (2001a) found raw water AOC/BDOC ratio equaled 6% in a Florida aquifer with significant surface influence and salt-water intrusion.

Table 1.4 Analysis of AOC and BDOC distribution in nonozonated water

	AOC			BDOC			AOC/BDOC
	TOC (mg/L)	AOC (µg/L)	AOC/ TOC (%)	TOC (mg/L)	BDOC (mg/L)	BDOC /TOC (%)	AOC/BDOC (%)
Median	3.5	120	2.8	3.6	1.0	20	22
Average	4.2	143	4.6	4.7	1.2	23	29
Maximum	16.3	482	38.3	16.3	7.8	72	89
Minimum	0.5	6.3	0.2	0.2	0.1	1	3
Standard Deviation	2.6	109	5.9	2.8	1.2	12	24
Coefficient of Variance	0.6	0.8	0.8	0.6	0.9	0.5	0.8
n	89	89	89	100	100	100	25

AOC-assimilable organic carbon, BDOC-biodegradable dissolved organic carbon, TOC-total organic carbon, n-number of values. References and dataset found in Appendix A Tables A1-A6.

A best practice approach is to follow ozonation with biofiltration to improve the water biostability in the distribution system. In addition, this coupled process will yield a decrease in the OM. Ozonation will increase the BOM fraction and biofiltration will decrease it, leading to an overall decrease in the OM fraction (Speitel et al., 1993; Cipparone et al., 1997; Hozalski et al., 1999; Carlson and Amy, 2001; Dowbiggin et al., 2001; Pharand et al., 2015). The literature BOM values for ozonated surface waters are shown in Table 1.5, albeit ozonated data were

nonpaired with nonozonated data (Table 1.4), i.e., the ozonated data in Table 1.5 cannot be directly compared to the nonozonated data in Table 1.4 to ascertain the direct impact of ozonation.

For the ozonated AOC data set, the median TOC value was 3.2 mg/L (n=56), slightly lower than the nonozonated median TOC. The median AOC value for ozonated waters was 230 μg/L (n=71). The number of AOC values is higher than for TOC values, as not all studies reported TOC values, and as expected the median AOC value of the ozonated waters was much higher than the nonozonated median AOC value of 120 μ/L. While the data were nonpaired, other studies have shown similar increases in AOC values after ozonation (Janssens et al., 1984; van der Kooij et al., 1989; Huck, 1990; LeChevallier et al., 1992). The ratio of AOC to TOC (n=55) increased by a factor of 3 for the ozonated waters compared to the nonozonated ratio values; from 2.9% to 8.8%.

For the ozonated BDOC data set (n=103), the median TOC value was 2.9 mg/L, which was lower than the nonozonated median TOC of 3.6 mg/L. The median BDOC value of the ozonated waters was slightly lower, 0.8 mg/L, compared to that, 1.0 mg/L, for the nonpaired nonozonated median value, but the median BDOC/TOC ratio increased from 20% for the nonozonated waters to 30% for the ozonation waters, with maximum and minimum ratio values of 62% and 8%, respectively. Other studies have shown similar increases in BDOC/TOC values after ozonation (Servais et al., 1987; Huck, 1990; Shukairy et al., 1992; Volk et al., 1993). Hence, after ozonation, 30% of the TOC is biodegradable and can be utilized by bacteria for energy and biomass growth.

The nonpaired AOC and BDOC data, with and without ozone, suggest that ozone increases AOC values more than BDOC values. Shukairy et al. (1992) and Escobar and Randall

(2001b) found ozonation increased AOC more than BDOC. This suggests that ozone preferentially generates the more easily assimilated compounds that are detected by AOC measurements. AOC/BDOC paired ratio values increased from 22% in nonozonated waters (n=24) to 30% in the ozonation waters (n=17), in nonpaired study comparison. Thus, after ozonation, 30% of the BDOC (9% of the total TOC) can be used by bacteria for biomass growth. Ozone dose/TOC was found to be significantly correlated to BDOC/TOC, AOC/TOC and AOC/BDOC ratios (p values <0.05 for all three scenarios), but a weak linear, or other type, correlations were found for all three relationships (R^2 <0.4) due to the scatter around the regression line.

Table 1.5 Analysis of AOC and BDOC distribution in ozonated waters

	AOC			BDOC			AOC/BDOC
	TOC (mg/L)	**AOC (µg/L)**	**AOC/ TOC (%)**	**TOC (mg/L)**	**BDOC (mg/L)**	**BDOC/ TOC (%)**	**AOC/BDOC (%)**
Median	3.5	230	8.8	2.9	0.8	30	30
Average	3.1	298	11.0	3.3	1.1	29	42
Maximum	7.1	1300	45.5	10.5	7.4	62	83
Minimum	0.7	27.5	1.7	1.0	0.1	8	11
Standard Deviation	1.1	245	9.4	1.9	1.2	11	26
Coefficient of Variance	0.4	0.8	0.9	0.6	1.1	0.4	0.6
n	56	72	56	103	103	103	18

AOC-assimilable organic carbon, BDOC-biodegradable dissolved organic carbon, TOC-total organic carbon, n-number of values

A distribution analysis of the AOC/TOC and BDOC/TOC ratios with and without ozone can be seen in Figure 1.1. The distribution of the data found 25th and 75th percentiles of source

waters without ozone were 1.3% and 6% AOC/TOC, and 14% and 27% BDOC/TOC, respectively. For waters with ozone, the data found 25th and 75th percentiles of source waters were 5% and 14% AOC/TOC, and 20% and 38% BDOC/TOC, respectively.

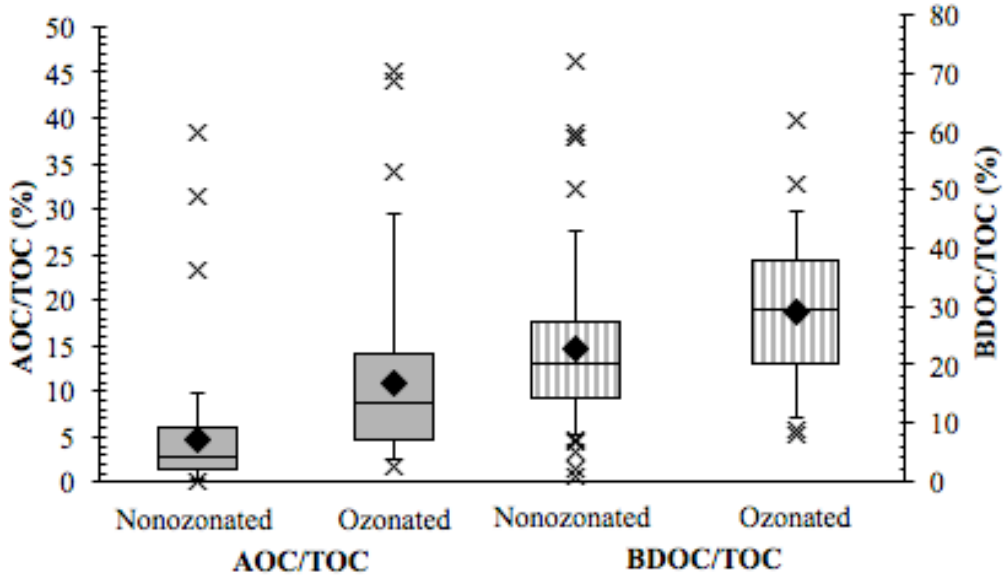


Figure 1.1 Distribution analysis of the ratio of BDOC/TOC and AOC/TOC with and without ozone. The boxes represent 25th, 50th (median) and 75th percentiles, the diamonds represent averages, the error bars represent 5th and 95th percentiles, and the "x" represent outliers.

1.2.2 Biofiltration Performance Review

Biofiltration is an attractive treatment option for BOM control due to its effective contaminant removal, low maintenance cost and ease of operation. For all surface water treatment plants with single stage filtration, the primary and paramount objective is the control of microbial pathogens. While the removal of BOM is a non-acute issue, both pathogen control and BOM removal can be both successfully accomplished with well operated biofilters (Goldgrabe et al., 1993). A survey of 38 North American utilities that employ biofiltration found the drivers for implementing biofiltration were as follows: multiple drivers (17 responses), taste and odor (15 responses), TOC removal (14 responses), improving filter performance (14 responses), DBP removal (11 responses), distribution system water quality stability (11 responses), sustainable

water treatment (5 responses), incidental (1 responses) and inorganics (1 responses) (Brown et al., 2016). Zhu et al. (2010) and Basu et al. (2015) have reviewed factors affecting biofilter performance in a semi-quantitative manner. Juhna and Melin (2006) reported a range of studies with ozone-biofilter removals of 15-30% TOC (n=19), 40–80% AOC (n=7) and 25-80% BDOC (n=9). As with any treatment option, design and operation should be optimized. Filter media, backwash disinfectants, empty bed contact time (EBCT), temperature, pre-oxidants and other parameters affect the removal efficacy of the biofilter and should be optimized, if possible, to achieve maximum removal (Hozalski et al., 1995; Urfer et al., 1997; Moll et al., 1999; Zhu et al., 2010, Basu et al. 2015). Again, this filter optimization needs to be carried out within the design and operation conditions for effective pathogen removal.

1.2.2.1 Impact of Design and Operation Parameters

Filter Media. Filter media, e.g., sand, anthracite and GAC, provides a surface for bacteria to attach and form communities and biomass (Servais et al., 1994). Media surface area is important, as well as media adsorption capacity. In many studies, the impact of residual adsorption capacity by GAC is not directly assessed, so caution needs to be used in over-interpreting results attributed to biological removal from short-term pilot studies run with GAC that has residual adsorption capacity. This is illustrated in Figure 1.2 where TOC removal is expressed as a function of time for two biofilters, one with inert media, such as anthracite and sand, and one with GAC media. With inert media, the acclimation time is normally 1 to 3 months for TOC removal at normal temperature ranges, 10 to 20 °C, after that the system will operate at steady state unless there are changes in the influent water quality, e.g., temperature, or in operating conditions, e.g., flow rate (Wang et al., 1995; Miltner et al., 1995). Also shown is the breakthrough history for GAC, which is impacted by the influent TOC concentration and the filter EBCT, with lower influent TOC and higher EBCTs leading to longer breakthrough, i.e.,

better removal at a given time. Initially the TOC removal by GAC is greater than that for a biofilter with inert media, but with time the GAC adsorption capacity for TOC becomes slowly exhausted and the removal becomes dominated by biological mechanisms. At this point the GAC biofilter is often referred to as a biological activated carbon (BAC) filter. In this study, a conservative approach was taken and TOC removal data from biofilters with GAC were only used when the GAC filter had been run for 18 months.

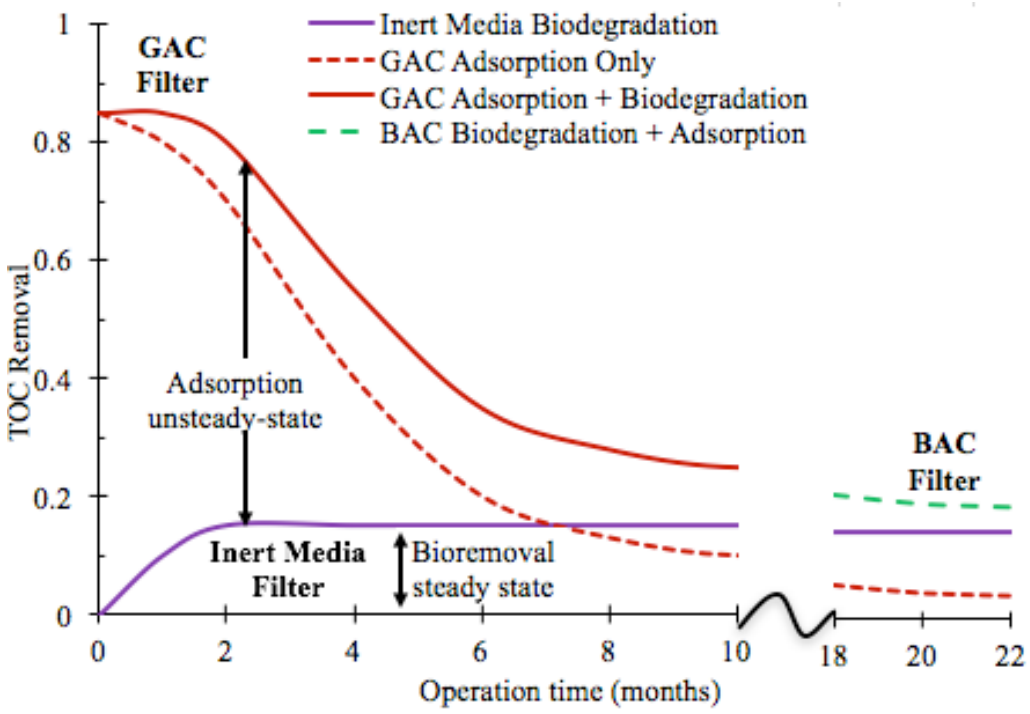


Figure 1.2 TOC removal acclimation for an inert media filter and a granular activated (GAC) filter, which becomes a biological activated carbon (BAC) filter once the adsoption capacity is exhausted.

A survey of 38 North American utilities found biofilter media configurations was predominantly GAC/sand (37%) and anthracite/sand (37%), while GAC alone (23%) and sand alone (3%) configurations were less abundant (Brown et al., 2016). Some studies have shown that at warm temperatures, GAC-sand and anthracite-sand performed similarly, but GAC-sand filters performed better at low temperatures, especially for aldehyde removal (LeChevallier et al.,

1992; Urfer et al., 1997; Liu et al., 2001; Emelko et al., 2006). Selbes et al. (2017) found GAC provided more removal than anthracite for DOC, DON and DBP precursors, while McKie et al. (2015) found GAC and anthracite performed similarly at two separate drinking water utility locations. Other studies reported that GAC is more adaptable to handle unfavorable conditions, such as lower temperatures, prechlorination, and disinfectant in the backwash, as well as provide a buffer for the biofilter ripening period through adsorption (Krasner et al., 1993; Carlson et al., 1994; Servais et al., 1994; Urfer et al., 1997; Liu et al., 2000, 2001). Wang et al. (1995) found at an EBCT of 9.2 min after 6 months of operation, the DOC removal for GAC, anthracite and sand biofilters was 29%, 16% and 20%, respectively. Basu et al. (2015) reviewed a study comparing GAC to anthracite and found at an EBCT of 8 minutes, GAC biofilter removed 11% to 14% of DOC and the anthracite biofilter removed 1% to 3% of DOC; additionally, at an EBCT of 16 minutes, GAC biofilter removed 15% to 20% of DOC and the anthracite biofilter removed 2% to 7% of DOC. Chowdhury et al. (2009) found GAC biofilter removed 9% of TOC, while anthracite biofilter removed 7% of TOC. Urfer et al. (1997) and Basu et al. (2015) suggested that better performance by GAC compared to inert media could be a result of GAC's ability to handle adverse operating conditions due to a higher surface area or porosity for biomass accumulation, additional adsorption capacity, continuous bioregeneration. The literature differs on biomass concentrations found on media type: some studies found GAC to have higher biomass concentrations (Wang et al., 1995; Huck et al., 2000), others found anthracite to have higher biomass concentrations (at high temperatures) (Emelko et al., 2006), while others found similar biomass concentrations for anthracite and GAC (Evans et al., 2013; Pharand et al., 2014), even at cold temperatures (Emelko et al., 2006). Therefore, media selection should be site specific based on influent water quality parameters (Urfer et al., 1997).

Backwashing procedures. Backwashing procedures can impact DOC removal efficacy. Basu et al. (2015) reviewed backwashing effects on biofilter performance through 'various mechanisms, including detachment and removal of biomass, redistribution of media and associated fixed biomass, adverse impacts of potential oxidants in the backwash water, and elimination of accumulated particles'. Hozalski et al. (1999) found periodic backwashing did not affect biofilter performance as long as 60-80% of the biomass was retained. Liao et al. (2015) found the biomass in a GAC filter could quickly recover to pre-backwash conditions (within 2 days) and DOC removal was highest directly after backwashing. Disinfectants in the backwash water can have adverse impacts on the performance of biofilters by damaging the biomass through oxidation. The detrimental effects of chlorine in the backwash water has been reported by several authors (Reckhow et al., 1992; Xie and Reckhow, 1992; Ahmad et al., 1994; Pharand et al., 2013; McKie et al., 2015). Miltner et al. (1996) compared biofilters with different backwash configurations and found the anthracite biofilter with no disinfectant in the backwash outperformed the biofilter backwashed with chloramine disinfectant, and both outperformed the biofilter backwashed with chlorine, in terms of control of NOM, ozone DBPs and DBP precursors. Liu et al. (2000) found the biofilter operated with a chlorinated backwash during low temperatures performed worse than the biofilter with no chlorine in backwash during higher temperatures. Liu et al. (2001) found that backwashing with chloramines did not significantly affect the BOM removal, but if chlorine could not be avoided in the backwash water, higher temperatures and GAC helped reduce the impairment from chlorine. Wang et al. (1995) reported better control of effluent concentrations with a biofilter with no disinfectant in the backwash, compared to a biofilter backwashed with chlorine. However, Chowdhury et al. (2009) found chlorine in the influent and the backwash had no effect on TOC removal with GAC biofilters.

Wang et al. (1995) found higher biomass concentrations on the anthracite filter (55 nmol lipid P/gmedia) with no exposure to chlorine compared to the anthracite filter that was backwashed with chlorinated water (6 nmol lipid P/g media). Miltner et al. (1995) suggested that biofilters perform better without disinfectants in the backwash due to a short-term weakening of the biomass rather than physical abrasion of the biomass when backwashing. Urfer et al. (1997) found chlorine in the backwash water lowered the amount of biomass in the anthracite-sand filter and the removal of chlorine demand, but did not affect the removal capacity of quickly biodegrading compounds, such as formaldehyde and AOC. Urfer et al. (1997) stated that backwashing biofilters using air scour does not lead to loss of attached biomass because bacteria attached to the media more strongly than nonbiological particle. Emelko et al. (2006) also found that backwashing with or without air scouring did not affect TOC removal for GAC/sand or anthracite/sand filters.

Overall, the reviewed performance data indicates the better BOM removal occurs when a disinfectant is not present in the backwash water. Fortunately, 76% of the 38 North American utilities surveyed in the did not use chlorinated backwash water (Brown et al., 2016). For many existing plants, the plant layout is such that producing backwash water without a disinfectant residual would be impractical, but the option of a disinfectant residual free backwash water should be provided in new and retro-fit plant designs when biofiltration is being considered. Therefore, treatment objectives must be defined before selecting backwash water conditions in order to get the best efficiency from the biofilter.

EBCT. Empty bed contact time is a measure of the water residence time in the biofilter and is a crucial design parameter for operating biofilters. The actual water residence time is about 50% lower as about 50% of the empty bed is occupied by the media. Hydraulic loading

rate (HLR) measures the flux (rate of volumetric flow per unit area) of water onto the biofilters and is inversely related to EBCT, i.e., lower HLRs increase EBCT, as seen in Equation 1.1.

EBCT = filter media depth/HLR = filter media volume / flow rate Eq. 1.1

A survey of 38 North American utilities that employ biological filtration found 33% of facilities operated at 2-5 min EBCT, 20% of facilities operated at 5-7 min EBCT, 27% of facilities operated at 7-10 min EBCT and 20% of facilities operated at >10 min EBCT (Brown et al., 2016). Longer EBCTs yield longer exposure and can enhance utilization of the substrate, which leads to more BOM and other contaminant removal (LeChevallier et al., 1992; Krasner et al., 1993; Carlson and Amy, 1995, 2001; Klevens et al., 1996; Miltner et al., 1996; Wu and Xie, 2005; Ko et al., 2007; Zhang et al., 2017). EBCT, not HLR, has an impact on DOC removal efficacy and should be the optimized parameter, as long as EBCT does not conflict with particle removal goals or hydraulic efficiency. It has been shown that external mass transfer in the HLR range of 1.5 to 15 m/hr (0.6 to 6 gpm/ft^2) does not constrain the removal of DOC, rather utilization of the substrate at the media surface limits DOC biodegradation (Wang and Summers, 1994). More than half of North American utilities surveyed operated in the loading rate range of 6.6 – 12.8 m/hr (2.6 – 5.0 gpm/ft^2) (n=38) (Brown et al., 2016). Wang and Summers (1994) found that HLRs in this range of 1.5 to 15 m/hr did not affect substrate removal when measured at the same EBCT, yet as EBCT increases from 3 to 33 minutes, DOC removal increased from 16% to 24%. Servais et al. (1994) found similar DOC removals from three biological activated carbon (BAC) filters operated at a constant EBCT of 10 minutes and three different HLRs (6, 12 and to 18 m/hr). Similarly, Carlson and Amy (1998) found similar DOC removals from two anthracite biofilters operated at different HLRs (5.0 and 9.7 m/hr), but the same EBCT (10 and 11 min). Chowdhury et al. (2009) found when EBCT increased from 7.1 to 9.3 minutes in GAC

filters, TOC removal increased from 9% to 11%. Urfer et al. (1997) summarized that studies have shown DOC removal increases with EBCT, but the relationship is not linear. LeChevallier et al. (1992) found DOC removal increases from 30% to 50% by extending the EBCT from 5 min to 20 min in a GAC filter. Therefore, longer EBCTs improve DOC removal efficiency. While increasing EBCT will increase BOM removal, but there may be a practical limit or threshold as increasing EBCT either yields longer filter bed depths at a set HLR, or more filter area (larger filters) at a set filter depth.

Temperature. Studies have shown that temperature affects biofiltration performance (Moll et al., 1999; Huck et al., 2000; Fonseca et al., 2001; Liu et al., 2001; Fonseca and Summers, 2003; Wu and Xie, 2005; Evans et al., 2013; Hallé et al., 2015; Basu et al., 2015; Selbes et al., 2016), as microbial growth and degradation rates are a function of temperature (Urfer et al., 1997). Selbes et al. (2016) found a large seasonal variability in biofilter performance over a 12-month period with DOC removal efficiency the lowest in the colder months (~5%) and the highest in warmer months (~24%). Moll et al. (1999) found NOM removal decreased at 5°C (15% DOC removal) compared to filters operated at 20 and 35°C (24% DOC removal). Emelko et al. (2006) showed water temperature significantly affected oxalate removal in GAC columns, with removals decreasing with decreasing temperature. Hallé et al. (2015) also found the temperature of the feedwater affected DOC removal in the biofilters. Wulfeck and Summers (1994) saw TOC removal decrease by 34% in full-scale biofilters operating at less than 14°C compared to operating at above 14°C. Hozalski et al. (1999) demonstrated that decreased water temperature required longer EBCTs to achieve the same steady state removal. Since DOC removal is dependent on temperature, conversion of a conventional filter to a biofilter should begin at warmer conditions to allow for a quick ripening

period (Urfer et al., 1997; Liu et al., 2000; Mofidi et al., 2005). Fonseca and Summers (1993) reported that temperature did not affect biomass concentration, but did impact biofilter activity. Persson et al. (2006) found the specific activity of biofilter biomass was a function of temperature, but BDOC removal was only slightly correlated to temperature in the range of 7°C to 20°C. Tradeoffs exists with temperature and EBCT, as both parameters effect DOC removal efficacy. Higher temperatures and longer EBCTs increase DOC removal, thus EBCT should be optimized for the required removal at specific influent temperatures, i.e. longer EBCTs are required at colder temperatures to maintain constant removal.

Biomass. Biomass concentration affects removal efficacy throughout the biofilter, as higher biomass allows for more oxidation of TOC (Carlson and Amy, 1998). However, Emelko et al. (2006) and Huck et al. (2000) found biomass levels, as measured by phospholipid methods, were not directly related to BOM removal. Biomass profile, ATP and phospholipid measurements, throughout the filter have shown to be the highest at the top of the filter and to decrease with increasing filter depth (Wang et al., 1995; Huck et al., 2000; Urfer and Huck, 2001; Persson et al., 2006; van der Aa et al., 2006; Velten et al., 2011; Rahman, 2013; Xiang et al., 2013; Pharand et al., 2014). Biofilters with residual disinfectants entering the filter have low biomass at the top of the filter, but once the residual is quenched, biomass quickly increases followed by the decreasing trend seen with filters without influent residuals (Urfer et al., 1997; Boon et al., 2011; Velten et al., 2011; Evans et al., 2013). Basu et al. (2015) reviewed 6 studies and found biomass ripening periods ranged between 20 days to > 16 months. The North American Biofiltration Knowledge Base Project 4459 survey found acclimation of biofilters treating surface waters, ground waters, and a blend ranged between 2 weeks to 6 months (Brown et al. 2016). Liu et al. (2001) found biofilter acclimation was a function of temperature, backwash conditions, type of

media, and BOM characteristics. DOC removal increases with increasing biomass until some threshold biomass concentration is reached (Urfer et al., 1997). In the steady-state biofilm, a mass balance of biomass growth and loss (due to detachment, abrasion and decay) is achieved (Siddiqui et al., 1997). Operating conditions that damage the biomass should be avoided, i.e., oxidants in the influent or backwash water, (Miltner et al., 1995; Wang et al., 1995).

Preozonation. Preozonation, as stated above, can increase the BOM fraction and thus enhance DOC removal in the biofilter (Huck, 1990; Siddiqui et al., 1997; Urfer et al., 1997; Carlson and Amy, 2001; Magic-Knezev and van der Kooij, 2004; Black and Bérubé, 2014; Selbes et al., 2017). A survey of 38 facilities found 63% of them utilize preozonation (Brown et al., 2016). Preozonation has been shown to increase biomass activity at the top of the biofilter compared to nonozonated biofilters (Goel et al., 1995; Fonseca et al., 2001; Magic-Knezev and van der Kooij, 2004; Pharand et al., 2014). Studies have shown that BDOC degradation rates increase linearly with ozone dose, but a threshold ozone dose exist where biodegradation rates plateau (Carlson and Amy, 1997, 2001; Yavich et al., 2004). Siddiqui et al. (1997) found increasing ozone/DOC ratio converted more DOC to BDOC but did not increase DOC removal after an ozone/DOC ratio of 1 mg/mg. Basu et al. (2015) summarized from 9 studies that the optimal ozone dose for enhancing DOC biodegradation in biofilters was 1 to 2 mgO_3/mg TOC. However, the residuals of oxidants into the biofilter should be minimized as they damage the biomass at the top of the biofilter (Evans et al. 2013). Urfer et al. (1997) reported ozone concentrations of 0.1 to 0.2 mg/L onto the filters inhibited biological activity of anthracite-sand filters. If oxidant residuals entering the biofilter cannot be minimized, GAC should be used at the top of the filter to reduce the disinfectant and allow the remaining filter bed to remain biologically active.

1.2.2.2 Biofilter Performance Analysis

An extensive review of biofiltration performance for TOC removal under a wide range of conditions, including temperature, preoxidation, EBCT, and media type, is summarized in Table 1.6 and the details provided in Appendix A Tables A7 and A8. The studies consisted of bench-, pilot- and full-scale data. TOC removal values are presented for all temperature ranges and oxidation conditions, then further divided into respective temperature range (≤10°C, 10 – 20°C, ≥20°C) and oxidation condition (nonozonated and ozonated). In addition, the EBCT range is reported at the bottom of the table for the respective temperature on ozonation conditions, as it has an impact on TOC removal efficiency. For all temperature and oxidation conditions, biofilters operated in an EBCT range of 2 to 38 minutes (average 12 minutes), removed 12% (median) of the influent TOC (n=117), with minimum and maximum removals of 2% and 47%.

Table 1.6 Biofilter performance for TOC removal for nonozonated and ozonated waters under different temperature ranges

	TOC REMOVAL (%)											
	All Data				Nonozonated				Ozonated			
Temperature (°C)	all	≤10	10 - 20	≥20	all	≤10	10 - 20	≥20	all	≤10	10 - 20	≥20
Median	12	10	12	17	10	7	10	15	15	11	13	20
Average	14	10	13	18	11	8	11	16	16	12	15	19
Maximum	47	24	47	45	22	14	22	22	47	24	47	45
Minimum	2	2	3	6	2	2	5	10	3	3	3	6
Standard Deviation	7.7	5.7	7.9	7.8	4.8	3.9	4.2	4.5	8.6	5.8	9.3	8.4
Coefficient of Variance	0.6	0.5	0.6	0.4	0.5	0.5	0.4	0.2	0.5	0.5	0.6	0.5
n	117	32	53	33	45	15	21	9	72	16	32	24
Average EBCT (min)	12	15	11	13	14	14	12	19	12	16	10	10

EBCT Range (min)	2 – 38	4 – 30	2 – 38	2 – 30	4 – 30	4 – 30	5 – 30	7 – 30	2 – 38	7 – 28	2 – 38	2 – 30

TOC-total organic carbon, n-number of samples, EBCT-empty bed contact time

Temperature affected the biofilter TOC removal efficacy. A histogram of temperatures throughout the studies (n = 117) ranging from 0.5°C to 35°C can be seen in Figure 1.3. About half of the studies, 51%, were performed in the temperature range between 10°C and 20°C. Comparatively, a survey of 38 full-scale utilities found 47% of utility source waters ranged between 15 °C and 20°C (Brown et al., 2016). In a nonpaired analysis of the removal data by temperature shows that as temperature increased from ≤10°C, 10 – 20°C, ≥20°C, TOC removal (median values) increased from 10% to 12% to 17% for all the data, 7% to 10% to 15% for the nonozonated data, and 11% to 13% to 20% for the ozonated data, respectively, as seen in Table 4. Thus, at higher temperatures, biofilters removal efficacy increases for both oxidation conditions, with at ≥20°C doubling that at ≤10°C, and 50% higher than that at 10 – 20°C.

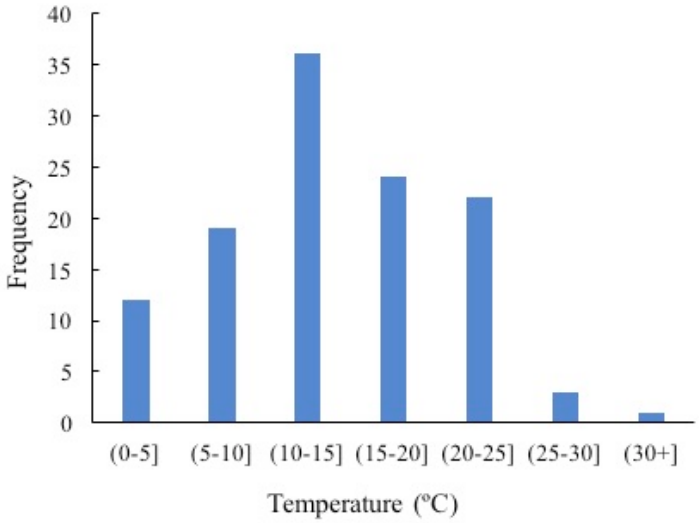


Figure 1.3 Temperature histogram (n=117). Ranges divided by ≤ 5°C, >5°C thru ≤ 10°C, >10°C thru ≤ 15°C, >15°C thru ≤ 20°C, >20°C thru ≤ 25°C, >25°C thru ≤ 30°C, and > 30°C.

When the data were separated by oxidation conditions the filter efficiency for nonozonated water was 10% median removed (n=45) and that for ozonated water was 15%

median removed (n=72). Further separating into temperature ranges, studies with high temperatures (≥20°C) and ozonated conditions demonstrated the highest TOC removal at a median of 20% (n = 24), with a range of 6% to 45%. The TOC removal efficiency at the high temperature range increased from 15% for the nonozonated waters to 20% for the ozonated waters (medians). However, lower differentials were found for median removals at the lower temperature range.

Filter media type had only slight impact on biofilter removal efficacy when analyzing the literature data. GAC, sand and anthracite media were included in the review. The studies with GAC were only included in our analysis if the GAC was in operation for over 1.5 years (about 75,000 bed volumes at a 10 min EBCT) to ensure the GAC adsorption capacity for TOC was operationally exhausted and removal was due to biodegradation. The median TOC removals for GAC, sand and anthracite at all temperatures and oxidant conditions were 16% (n= 22), 14% (n = 19) and 13% (n=76), respectively.

As discussed in the literature review, EBCT also affects the biofilter removal efficacy. Figure 1.4 shows a histogram of the EBCTs (n = 102) reported in the studies. The distribution of EBCT peaks at 7.5 minutes and drops off after 10 minutes, which then tails out to 38 minutes. A small peak is also seen for 25 to 30 minute EBCTs. Most of the studies, 65%, were run at EBCTs between 2 and 10 min. Longer EBCTs allow for more TOC removal and should be optimized for treatment objectives.

While a systematic impact of temperature and pre-oxidation condition can be seen in the Table 1.6 results, there are high levels of variability in any given category, as indicated by the high standard deviations and the resulting coefficient of variation (CV) values, 0.2 to 0.6. Part of this variation can be attributed to the range of EBCTs in a category, which are shown in Table

1.6. The TOC removal data only increased by a maximum of 3% when temperature increased from the ≤10°C range to the 10 – 20°C range in the all data, nonozonated and ozonated conditions. This small increase could be a result of longer EBCTs at the lower temperature range compared to the medium temperature range. For all data, EBCT averaged 15 minutes for ≤10°C range and averaged 11 minutes for 10 – 20°C range. For nonozonated data, EBCT averaged 14 minutes for ≤10°C range and averaged 12 minutes for 10 – 20°C range. For ozonated data, EBCT averaged 16 minutes for ≤10°C range and averaged 10 minutes for 10 – 20°C range.

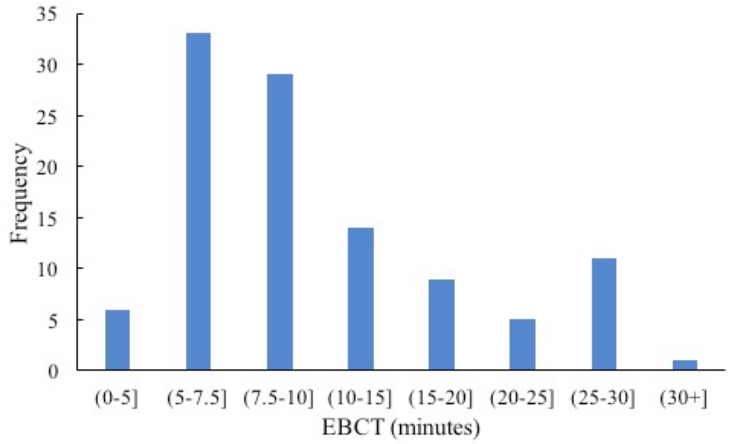


Figure 1.4 EBCT Histogram (n = 108). Ranges divided by ≤ 5 min, >5 min thru ≤ 7.5 min, >7.5 min thru ≤ 10 min, >10 min thru ≤ 15 min, >15 min thru ≤ 20 min, >20 min thru ≤ 25 min, >25 min thru ≤ 30 min and > 30 min.

This wide range of EBCTs in a given oxidant and temperature category complicates the analysis of the data. To overcome this a pseudo-first order model was applied to the TOC removal data to demonstrate the effects of EBCT (Equation 1.2) (Huck et al., 1994; Wang and Summers, 1994; Zhang and Huck, 1996; Chen et al., 2016). With this simplified model, BDOC removal is an exponential function of the observed rate constant, k', and EBCT as follows,

$$c/c_{inf} = \exp(-k'*EBCT) \qquad \text{Eq. 1. 2}$$

Where, c = the BDOC concentration of the effluent, c_{inf} is the BDOC concentration of the influent, k' = the observed rate constant (min^{-1}) and EBCT (min). One drawback of applying this

approach is that an average BDOC fraction, 20% for the nonozonated waters (Table 1.4) and 30% for the ozonated waters (Table 1.5), must be assumed to calculate the influent BDOC concentration from the influent TOC concentration values. The removal of BDOC is first calculated with Equation 1.2 and the TOC removal is then calculated based on the assumed BDOC fraction of the TOC, i.e., 20% for the nonozonated studies and 30% for the ozonated studies. More sophisticated models have been developed for BOM removal by biofilters and they are reviewed by Servais et al. (2005). However, the required additional parmeters, such as biomass and the distribution of easily to more recalcitrant biodegradable OM, are not available in the literature studies. The intent is to use this model to illustrate the impact of EBCT and temperature on BOM removal, not as a calibrated predictive model.

The k' values, shown in Table 1.7, were calculated from the literature TOC removal and associated EBCT. Table 1.7 is organized similar to TOC removal from Table 1.6, with the data presented all together and then divided into temperature range and oxidation condition. The n values for Table 1.7 may not equal to that in Table 1.6 because not all studies reported EBCT. For all data (n=107), the average k' value was 0.10 min^{-1}, with a range of 0.01 to 0.36 min^{-1}. As temperature increased, the average k' value increased. For the combined non- and ozonated data, the average k' increased from 0.05 min^{-1} at ≤10°C to 0.09 min^{-1} at 10 – 20°C, to 0.14 min^{-1} at ≥20°C. For all temperature ranges the average k' values increased from 0.08 min^{-1} for the nonozonated data (n= 44) to 0.10 min^{-1} for the ozonated data (n= 62). For the nonozonated data, the average k' values increased from 0.05 min^{-1} at ≤10°C to 0.09 min^{-1} at 10 – 20°C, to 0.11 min^{-1} at ≥20°C. The ozonated data showed the greatest increase as the k' value increased from 0.04 min^{-1} at ≤10°C to 0.09 min^{-1} at 10 – 20°C, to 0.15 min^{-1} at ≥20°C. The observed rate constant illustrates the rate at which TOC is biodegraded, at higher temperatures TOC biodegrades more

quickly, which is consistent with literature findings (Moll et al., 1999; Huck et al., 2000; Fonseca et al., 2001; Liu et al., 2001; Fonseca and Summers, 2003; Wu and Xie, 2005; Evans et al., 2013; Hallé et al., 2015; Selbes et al., 2016). The large CV values, 0.5 to 1.0, are partially an outcome of assuming a constant BDOC fraction of the influent water, as the CV values for the BDOC fraction are 0.5 (Table 1.4) and 0.4 (Table 1.5) for the nonozonated and ozonated waters, respectively.

Table 1.7 Observed first order constant, k' (min^{-1}), calculated from literature studies

	Observed first order constant, k' (min^{-1})											
	All Data				Nonozonated				Ozonated			
Temperature (°C)	all	≤10	10 - 20	≥20	all	≤10	10 - 20	≥20	all	≤10	10 - 20	≥20
Median	0.07	0.03	0.07	0.13	0.07	0.03	0.09	0.11	0.06	0.03	0.06	0.15
Average	0.10	0.05	0.09	0.14	0.08	0.05	0.09	0.11	0.10	0.04	0.09	0.15
Maximum	0.36	0.17	0.34	0.36	0.32	0.17	0.32	0.18	0.36	0.10	0.34	0.36
Minimum	0.01	0.01	0.01	0.04	0.01	0.01	0.02	0.05	0.01	0.01	0.01	0.04
Standard Deviation	0.07	0.04	0.07	0.08	0.06	0.05	0.06	0.06	0.08	0.03	0.08	0.08
Coefficient of Variance	0.8	0.8	0.8	0.5	0.7	1.0	0.7	0.6	0.9	0.7	0.9	0.5
n	107	29	50	28	44	14	21	9	62	15	28	19
Average EBCT (min)	12	15	11	13	14	14	12	19	12	16	10	10
EBCT Range (min)	2 – 38	4 – 30	2 – 38	2 – 30	4 – 30	4 – 30	5 – 30	7 – 30	2 – 38	7 – 28	2 – 38	2 – 30

TOC-total organic carbon, n-number of samples, EBCT-empty bed contact time

An analysis of the observed rate constant values as a function of EBCT for each temperature and oxidant category showed that the k' values are 69% higher for EBCTs less than 6 min (n=26) compared to EBCTs greater than 6 min (n=81). Above 6 min EBCT, there was not

a significant correlation between EBCT and k' values for each temperature and oxidant category (p values were all > 0.12). This is consistent with the approach of dividing the BOM into a fast reacting fraction, which is consumed in the top of the filter bed and a slower reacting fraction which is consumed throughout the filter (Servais et al., 2005).

A distribution analysis of the TOC percent removal and k' constants with and without ozone can be seen in Figure 1.5. For both parameters, the nonozonated water data should not be compared to the ozonated water data, as they are not from the same population, i.e., compounding variables (EBCT and temperature) affect TOC removal and they are not equally represented in the distribution of the data. The distribution of the data found 25th and 75th percentiles of biofiltration TOC removal without ozone were 8% and 13%, and with ozone were 10% and 20%, respectively. For k' constant, the distribution found 25th and 75th percentiles of k' values without ozone were 0.03 min^{-1} and 0.09 min^{-1} and with ozone were 0.04 min^{-1} and 0.13 min^{-1}, respectively.

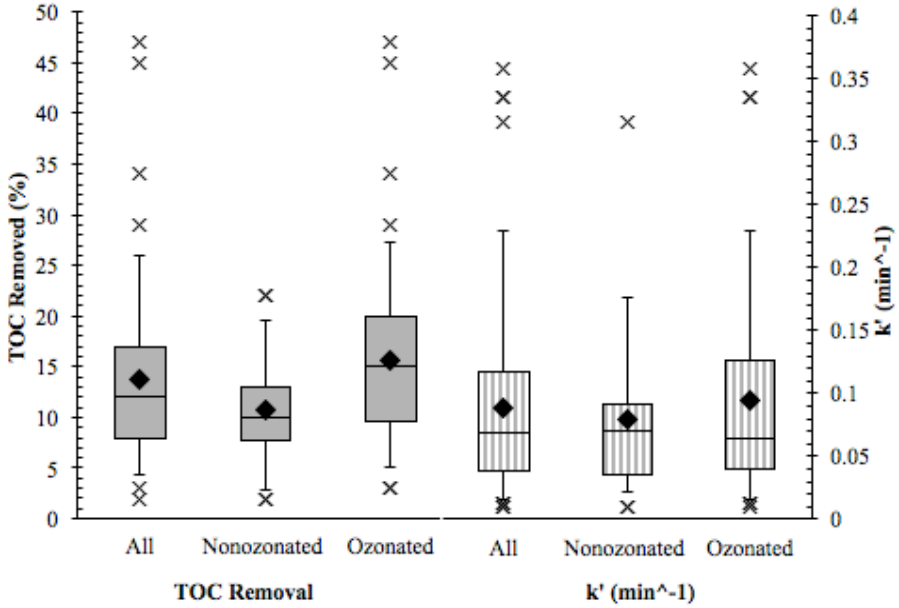


Figure 1.5 Distribution analysis of TOC removal and k' constant with and without ozone. The boxes represent 25th, 50th (median) and 75th percentiles, the diamonds represent averages, the error bars represent 5th and 95th percentiles, and the "x" represent outliers.

Understanding the impact of EBCT on performance allows for optimization of biofilter operations. Figure 1.6 demonstrates the effect of EBCT, using Equation 1.2 and the average literature observed k' values from Table 1.7. One k' value was used for each temperature range, calculated from the literature data (k' = 0.05 min^{-1} for ≤ 10°C, k' = 0.09 min^{-1} for 10-20°C, k' = 0.14 min^{-1} for ≥20°C). The TOC removals were capped at 20% for the nonozonated studies and 30% for the ozonated studies, based on the BDOC values of surface water found in Tables 1.4 and 1.5, respectively. Ozonated and nonozonated TOC removals were modeled with EBCT using Equation 1.2, as seen in Figure 1.6. As EBCT increases, TOC removal increases exponentially with EBCT and asymptotically approaches BDOC values of 20% or 30%, depending on oxidation conditions. The advantage of using a constant k' value for each temperature range allows utilities to determine the optimal EBCT for the desired TOC removal depending on site specific water temperature. In the EBCT range of 5 to 10 min (the range reported for 60% of the data) differences in removal at ≤10°C compared to ≥20°C for either the ozonated or nonozonated waters are greater than that due to ozonation at a given temperature range. This illustrates the importance of temperature, EBCT, and ozone on performance.

Ozone is expensive and utilities that do not employ ozone for disinfection can overcome the lack of preoxidant by utilizing longer EBCTs to achieve similar TOC removals. The ability of overcoming the TOC removal benefit of preozone through an extension of EBCT can be demonstrated with the results in Figure 6. If a utility wants a 15% TOC removal and is operating in the temperature range of 10 to 20°C, based on Figure 6, an EBCT of only 7 minutes is required if the water is preozonated. However, if pre-ozonation is not used, an EBCT of 15 minutes can be used to achieve the same 15% TOC removal. Again, this analysis is only

intended as an estimate of the trade-offs that utilities can consider when evaluating biofiltration. Long-term pilot filters are a better way of assessing the design trade-offs.

Colder climates with low water temperatures have lower removals at similar EBCTs compared to that at warmer climates. As shown in Figure 1.6, the biofilter treating ozonated water at ≥20°C will achieve 20% removal in 8 minutes. However, the biofilter treating ozonated water at ≤10°C will need 22 minutes of EBCT to achieve 20% removal. Thus, if the cold water biofilter were operated at longer EBCTs, it could achieve similar removal as the biofilter operated in a warmer climate. Seasonal water temperature changes require optimization of EBCT, with longer EBCTs needed to compensate for less DOC removal during colder months. Fortunately, water demand variation is often seasonal and coincides well with the biofilter, as during colder seasons, the water demand is lower, and EBCT can be increased without adverse effects. For cold temperatures, drinking water utilities can operate all of the filters, but at a lower HLR in order to extend the EBCT. During warmer months, shorter EBCTs will achieve similar TOC removals compared to colder months with longer EBCT. Therefore, drinking water utilities can increase the HLR, lowering EBCT, to meet their demand without reducing TOC removal.

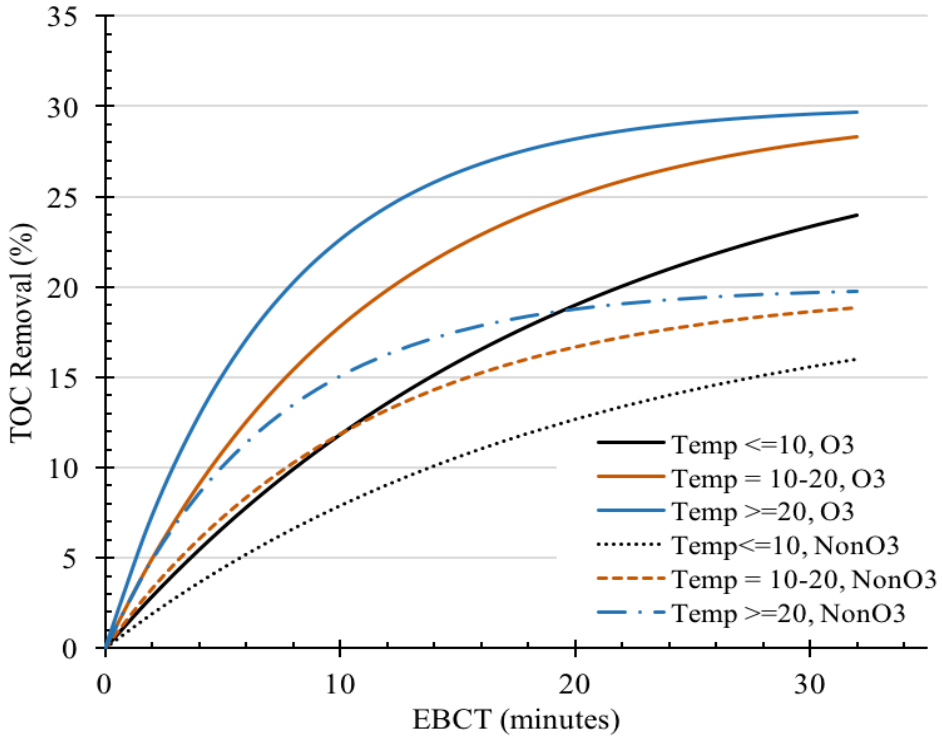


Figure 1.6 Simulated TOC removal as a function of EBCT at three temperature ranges for ozonated and nonozonated waters with associated k' values (k' = 0.05 min^{-1} for ≤10°C, k' = 0.09 min^{-1} for 10-20°C, k' = 0.14 min^{-1} for ≥20°C).

1.3 Conclusions

An extensive literature review found BDOC and AOC comprised 20% and 3%, respectively, of the TOC found in nonozonated water, with AOC comprising 22% of the BDOC in the paired data. The review of ozonated waters found BDOC and AOC comprised 30% and 9%, respectively, of the TOC with AOC comprising 30% of the BDOC in the paired data. For all temperature and oxidation conditions (n=117), biofilters operating in an EBCT range of 2 to 38 minutes removed 12% (median) of the influent TOC. The median filter efficiency for nonozonated water was 10% TOC removed (n=45) and that for ozonated water was 15% TOC removed (n=72). Temperature had a large impact on the TOC removal efficiency. As temperature increased from ≤10°C to 10 – 20°C to ≥20°C, TOC removal (median values) increased from 10% to 12% to 17% for all data. A pseudo first-order model is useful for

illustrating the impact of EBCT, pre-ozonation and temperature on biofilter performance. Biofiltration can be an efficient treatment technology to remove the biodegradable portion of organic matter from the influent water, but should be optimized to achieve maximum removal. EBCT, temperature, oxidant conditions, and backwash strategies, among others, can impact biofilter efficacy and should be carefully chosen or taken into consideration in the design and operation of biofilters.

1.4 Research Objectives and Hypotheses

The goal of this dissertation is to comprehensively study the removal of BOM by biological filters and to develop optimized operational strategies. The overall objectives of the proposed research are to evaluate the impacts of operational parameters (EBCT, the amount of biomass within the filter) and water quality (temperature, NOM matrix) on the removal efficiency for BOM and complete a comparative life cycle analysis of a biological filter and a conventional filter.

Hypothesis 1: Extended EBCT will lead to a higher removal efficacy of slowly degrading organic contaminants and organic matter.

Hypothesis 2: Larger accumulation of biomass will lead to more degradation and removal of contaminants.

Hypothesis 3: Increasing temperatures will yield increasing removal of contaminants in biological filtration.

Hypothesis 3a: Increasing temperatures result in increasing biomass activity within the filter.

Hypothesis 3b: Increasing temperatures lead to increasing degradation rate constants of contaminants.

Hypothesis 4: Biofiltration performance of BOM removal can be modeled using Monod kinetics when BDOC of the source water, total biomass of the filter and a contaminant utilization rate constant are known.

1.5 Scope

This research was performed with bench-scale and pilot-scale biofilters with media from a full-scale drinking water facility. The ranges of parameters tested were relevant for biofilters operating in a drinking water facility treating surface water. The feed waters were environmentally relevant source waters and chosen as a representative sample of source waters likely to be an intake for drinking water treatment facilities.

1.6 Thesis Organization

This thesis is divided into seven chapters to address the objectives and hypotheses. Chapter 1 is a literature review on organic matter occurrences in surface water and biofiltration performance. Chapter 2 outlines the materials and methods used throughout the research. Chapter 3 evaluates and models biological filtration performance for DOC removal. Chapter 4 investigates biofiltration performance based on extended EBCT, and biomass acclimation and distribution at the pilot scale. Chapter 5 investigates how different water quality parameters, temperature and NOM characterization, effect biofilter DOC removal and DBP formation. Chapter 6 evaluates environmentally sustainable scenarios for biological filters. Concluding, Chapter 7 summarizes the findings of this research.

Chapter 2
Materials and Methods

2.1 Introduction

In this chapter, a new method for assessing the effectiveness of bench scale biofiltration for drinking water treatment is developed and verified. This chapter also includes a detailed description of the material and methods employed throughout this dissertation. Each subsequent chapter has a smaller, abridged materials and methods section, but the current chapter should be the reference for duplicated experiments.

2.1.1 Development of a Bench Scale Biofiltration Method

Application of biological filtration for a drinking water treatment process typically involves bench scale proof-of-concept, on-site pilot scale demonstration, and full scale implementation. Researchers often need to scale-up or scale-down process designs for the purpose of transferring locations that require different sizes or for the purpose of simulation (Manem & Rittmann, 1990). The pilot scale demonstration can be costly and logistically difficult, but provides the benefit of ample influent primary substrate. If the pilot scale demonstration could be simulated at the bench scale, applying a new treatment process could be streamlined to reduce time to implementation, cost of materials and supplies, and operator time and energy.

Bench scale biofiltration experiments in replace of pilot scale biofiltration experiments can have a positive impact on implementation of the technology in drinking water treatment plants (DWTP). Bench scale experiments are more easily executed, thus utilizing a bench scale system in exchange of an on-site pilot experiment can reduce the time required to evaluate biofiltration for a specific water quality scenario by minimizing time lost to logistical setup with

an on-site experiment at a DWTP. Bench scale experiments utilize less mass of materials than the pilot scale due to the smaller size. Bench scale experiments are often executed in the laboratory, which reduces the time required for sample collection, transportation and analysis, and also eliminates additional pathways of contamination. Assessing biofilter contaminant removal in a laboratory setting allows for experimental control, i.e., control of operating parameters, that is often not controllable at the pilot scale. Experiment control allows for versatility in what parameters are tested and when, a commodity not available at the pilot scale. Bench scale experiments eliminate water quality variability present in flow through pilot operations that can impact overall results, such as variability in temperature, influent total organic carbon (TOC), pH, etc. In addition, bench scale experiments enable controlled assessment of variables in a condensed time period, regardless of current operating and water quality conditions. Spikes in contaminant concentration and corresponding acclimation time can be evaluated in a laboratory setting to mimic boundary conditions at the full scale (Halle et al., 2015).

Pilot scale systems are flow through systems with a constant supply of primary substrate for microorganisms, while bench scale systems are normally batch reactors with finite supply of primary substrate. Flow through experiments at the bench scale are often not performed due to the requirement of hauling multiple barrels of source water from the field to the laboratory. Bench scale batch experiments are commonly proof of concept and can have limited applicability for full scale treatment. There have been several bench scale methods proposed in the literature to measure biodegradable organic carbon (BDOC) in source waters, as reviewed in Chapter 1. BDOC is calculated as the change in DOC concentration over a specified time period. Methods employ batch reactors (Servais et al., 1987; Joret et al., 1989), recirculating columns

(Mogren et al., 1990), single-pass columns (Frias et al., 1992), and shaken batch reactors (Allgeier et al., 1996). However, most of these methods are limited in scaling up and must be done at the bench scale.

Based on the literature and to the best of the authors' knowledge, a methodology for scaling biofiltration pilot and bench scale experiments does not exist. While some studies included a bench scale and pilot scale component, a direct comparison was not made. Snyder et al. (2007) looked at the removal of organic micropollutants in drinking water and reuse water treatment processes at the bench scale and pilot scale; however, the researchers did not perform a direct comparison and saw different removals at each scale. For the bench scale, biodegradable dissolved organic carbon (BDOC) method was adapted from Allgeier et al. (1996) and at the pilot scale a flow through system was employed. Reasons for removal discrepancies could be due to adsorption, substrate availability, contact time, or differing hydraulics (Snyder et al., 2007). Manem and Rittmann (1990) developed a scaling procedure for continuous biofilm processes when the same type of microorganisms and reactors were used and when the following biofilm kinetics were equal in the prototype and the scaled process: substrate concentration at the surface of the biofilm, the biofilm shear loss-rate and the substrate mass balance. However, in drinking water treatment, influent primary substrate is low, thus discontinuous or patchy biofilms develop, not continuous biofilms like in wastewater treatment (Rittmann, 1993). Rittmann (1993) stated the discontinuous biofilms were important when modeling spatial distribution of attached biomass and permeability loss, but not critical when modeling substrate removal because diffusion resistance was not significant in fully penetrable discontinuous biofilms; however, the results were not verified experimentally. Researchers who want to scale up or scale down a process design need all aspects of the design to be scalable, not just substrate removal. Therefore,

the Manem and Rittmann (1990) scaling procedure is not applicable when discontinuous biofilm develops due to low incoming primary substrate.

The objective of this study was to develop a methodology that comparatively simulates pilot scale flow through biological filtration experiments at the bench scale. This innovative methodology built upon recirculating batch reactor methods (Mogren et al., 1990) combined with single pass flow through methods (Frias et al., 1992) to prevent limited substrate availability and maintain adequate contact time. The uniqueness of this method allowed for controlled variables that are often not controllable at the pilot level, i.e. influent primary and secondary substrate concentrations, temperature, and replication. In addition, this methodology minimized the volume of water required from the field to simulate flow through conditions. This methodology was tested for DOC removal and biomass activity in drinking water at the bench and pilot scale.

2.2 Experimental Design and Operations

2.2.1 Media Type and Origin

Biologically active anthracite media from a full scale filter, which was in operation for over seven years at the City of Longmont (CO) Nelson Flanders Drinking Water Treatment Plant, was sampled two times (October 2013 and January 2015) and transported to the University of Colorado, Boulder. The anthracite media was used exclusively in all experiments, except Chapter 4 when 'fresh' anthracite, along with acclimated anthracite, was used to evaluate acclimation. The anthracite media had an effective size of 1.0 mm and an approximate uniformity coefficient of 1.3. The plant source water was a combination of the St. Vrain Creek and Colorado-Big Thompson Project sources, which were not impacted by wastewater discharges. The raw water was treated by aluminum sulfate coagulation, flocculation and sedimentation. The biological filters were backwashed with unchlorinated water.

Once at CU Boulder labs, the biologically active media was placed in an upflow reactor and water was recirculated through the reactor at a flow rate of 2 mL/min prior to use. Three liters of dechlorinated tap water (DCT), spiked with natural organic matter, NOM, from Big Elk Meadows, CO (BEM) at a TOC concentration of 3 mg/L, were held in an amber glass reservoir and recirculated through the reactor. The reservoir was changed weekly. The reactor was constructed of three-inch Schedule 40 PVC pipe with threaded end caps. Stainless steel connectors were tapped into the caps to attach plastic tubing and stainless-steel mesh was placed at the bottom and top of the filter to prevent media loss.

2.2.2 Bench Scale Experiments

2.2.2.1 Biofilter Design

Glass chromatography columns with Teflon caps (ACE Glass 5820-12 and 5820-24, Vineland, NJ) and stainless-steel metal fittings (Swagelok Cleveland, OH) were used for constructing the biofilters. The biologically active anthracite media was packed into either three 15 mm or two 11mm inner diameter columns connected in series. In order to prevent media loss or clogging, the bottom of each column was packed with 2 inches (5 cm) of support media, 2 mm diameter glass beads, encased in a wire mesh. A needle valve after each column was used to control flow. Sampling ports were located immediately after each column to assess removal associated directly with the filter. The columns were covered to minimize the growth of photosynthesizing microorganisms in the filters.

2.2.2.2 Biofilter Operation

All columns were operated at a hydraulic loading rate of 1.0 gpm/ft2 (2.44 m/hr). Experiment 1 consisted of two 11 mm inner diameter columns in series. The flow rate for Experiment 1 was 3.48 mL/min and the EBCTs were 15 and 30 minutes, calculated as the ratio of the depth to the hydraulic loading rate or column cross-sectional area to volumetric flow rate. Experiment 2 consisted of three 15 mm inner diameter columns connected in series. The flow

rate for Experiment 2 was 7.2 mL/min and EBCTs were 5, 15 and 30 minutes. The support media was not included in the calculation of the EBCT. The columns were operated down-flow using peristaltic pumps (Masterflex Models 7553-30 and 7518-10) and Masterflex tubing. Biofilter design parameters can be seen in Table 2.1.

In order to emulate a flow-through pilot scale experiment at the bench scale, the bench scale biofilters were operated in recirculating batch mode (adapted from Joret & Levi 1986, Mogren et al. 1990, Frias et al. 1992) followed by flow through single pass mode then sampling. In order for microorganisms to adjust to each new influent condition, the biofilters were acclimated for five days through batch recirculation. The recirculation period reduced the amount of water needed for the experiment. Single pass mode was employed to simulate pilot scale flow through and allow for ample primary substrate to be consumed prior to sampling. Lastly, sampling occurred as would during a pilot flow through experiment. In order to execute this method, the feed water was split into two batches. The first half of the feed water was recirculated to acclimate the media to the specific water matrix for a period of 5 days and the second half was stored at 4 °C and brought to room temperature before use. Following the acclimation period, the second half of the feed water was run in a single pass mode for 4 hours, then sampling occurred. Referring to Figure 2.1 and 2.2 showing the biofilter apparatus at CU Boulder, valve 1 was open for recirculation and valve 2 was open for single pass. The acclimation period followed by single pass and sampling allowed the bench scale experiment to simulate a pilot scale flow through experiment and combined the benefit of each scale.

The flow varied slightly due to particle/biomass buildup within the biofilter apparatus and was measured every three days and adjusted as necessary. The biofilter flow rate was not affected by the change in hydraulic head due to water decreasing in the feed tank during single

pass sampling. The flow was monitored by collecting and measuring effluent water in a graduated cylinder over a one-minute time frame. The flow was adjusted accordingly by a needle valve immediately after the biofilter. The biofilters were not backwashed, as buildup never warranted the need for backwashing.

The biofilters in normal operation were operated at lab temperature (20 ± 2 ° C). Experiments were ran at varying temperatures to determine temperature effects on biofilter performance. In order to run experiments at cold temperatures, biofilters were set up in the walk-in refrigerator (Bally Refrigerator Boxes, Inc. Model 3678) set for either 6 °C or 28 °C. In other scenarios, apart from lab temperature, experiment temperatures were controlled by a water chiller with capacity for cooling and heating. The feed water was directly inserted into the chiller, acting as a temperature sink, and the biofilters were wrapped with temperature controlled tubing. Figure 2.1 displays Experiment 1 apparatus and Figure 2.2 displays Experiment 2 apparatus.

Table 2.1 Biofilter Design Parameters

Biofilter	Media Type	Experiment	Target EBCT (min)	Media Height (cm)	Inner Diameter (mm)	Support Media Height (cm)	Flow Rate (mL/min)
1a	Anthracite	Bench	15	55	11	5	3.48
1b	Anthracite	Bench	30	55 (+55 from 1a)	11	5	3.48
2a	Anthracite	Bench	5	20.4	15	5	7.20
2b	Anthracite	Bench	15	40.6 (+20.4 from 2a)	15	5	7.20
2c	Anthracite	Bench	30	61.7 (+61 from 2a and 2b)	15	5	7.20

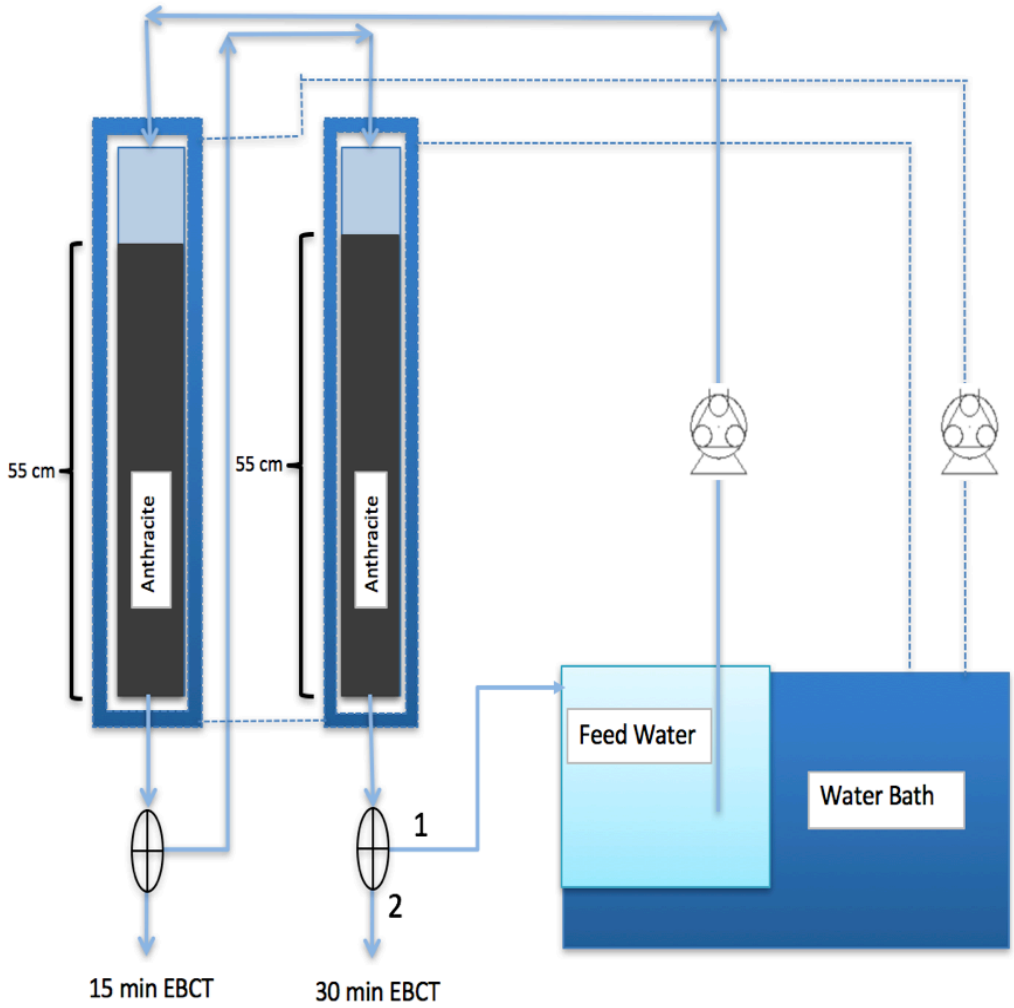


Figure 2.1 Biofilter apparatus 1 (1a and 1b) for CU-B bench scale biofilters, recirculation (valve 1 open) vs. single pass mode (valve 2 open)

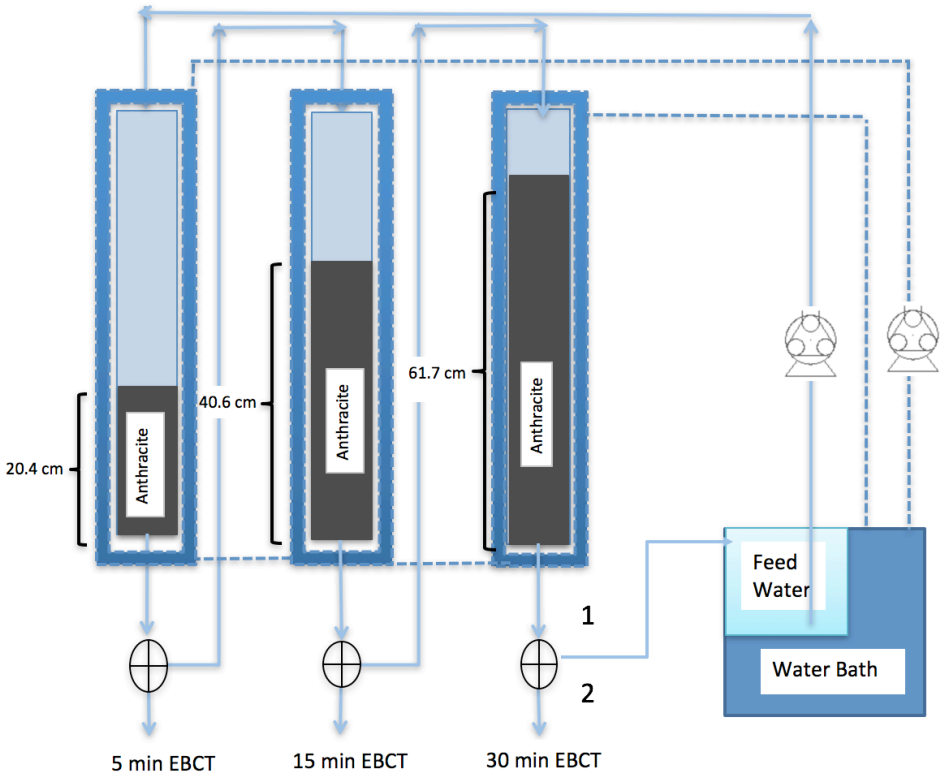


Figure 2.2 Biofilter apparatus 2 (2a, 2b, and 2c) for CU-B bench scale biofilters, recirculation (valve 1 open) vs. single pass mode (valve 2 open)

2.2.3 Pilot Scale Experiments

2.2.3.1 Pilot Biofilter Design

Pilot filters were set up at the City of Boulder's (CO) Betasso Water Treatment Plant (Betasso WTP). The Betasso WTP pilot was composed of a pilot treatment train operated at a flow rate of 2 gal/min. The train consists of rapid mix, three stage tapered flocculation, sedimentation, and filtration. The pilot system was modified and two new biofilter columns were fabricated with depth taps to achieve filter depth samples, as seen in Figure 2.3. The placement of the sampling ports allowed for measurement of only removal associated with the biological media and not the feed system. Filter 1 was packed with 'fresh' anthracite and Filter 2 was packed with bioacclimated anthracite from Longmont (CO) Nelson Flanders Water Treatment Plant. The anthracite is described in detail in Section 2.2.1 Media Origin above. The anthracite was uniformly placed into clear PVC columns with 76 mm inner diameter to a depth of 100 cm.

The sample taps were located at the top of the media to get the top of filter samples and 17 cm, 50 cm, and 100 cm below the top to represent EBCTs of 5, 15 and 30 minutes, as seen in Table 2.2.

2.2.3.2 Biofilter Operation

The filters were online for six months of the winter, spring and summer and captured surges in turbidity and NOM during spring runoff, with temperature changes from 5 to 20 °C. Source water, a combination of Barker Reservoir and Lakewood Reservoir (water quality data for duration of study seen in Table 2.3), was sent to the raw water tank, from which water was pumped to the treatment train. The biofilters were backwashed once per week with chlorinated water. Flow rates were monitored online via a flow analyzer. The analyzer installed was a Blue White F-400N Inline Rotameter with a range of 0.025 – 0.25 gpm. The flow was changed by adjusting the ball valve at the end of each filter. The flow rate was measured after each filter using in-line flow meters and averaged at 0.04 gpm, and the hydraulic loading rate averaged 2 m/hr.

Table 2.2 Pilot Design Parameters

Biofilter	*Media Type*	*Experiment*	*Target EBCTs (min)*	*Media Height (cm)*	*Inner Diameter (mm)*	*Support Media Height (cm)*	*Flow Rate (gpm)*
3 & 4	Anthracite	Pilot	5	17	76	8	0.04
		with taps	15	50			
			30	100			

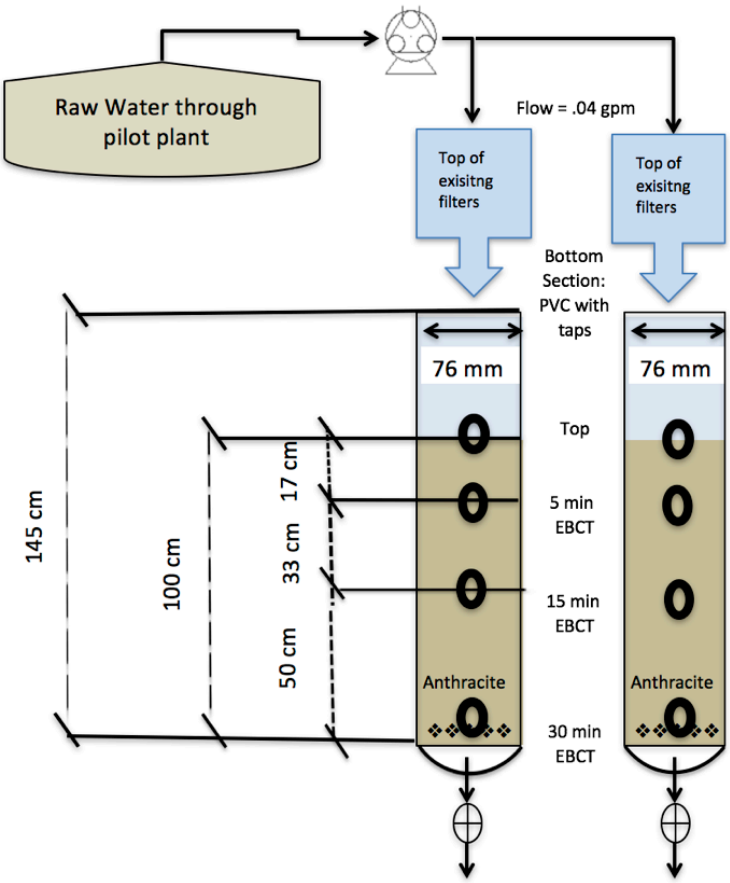


Figure 2.3 Pilot Scale Experiment Apparatus

2.2.4 Biofilter Feedwaters

2.2.4.1 Terrestrial Source Water

Barker Reservoir is located in Netherland, Colorado and receives snowmelt input yearly. Barker Reservoir served as the terrestrial source for the bench scale experiments. Water quality data at the time of sampling can be found in Table 2.3.

2.2.4.2 Microbial Source Water

Wonderland Lake is located in Boulder, Colorado and is not impacted by runoff or wastewater effluent. Wonderland Lake served as the microbial source for the bench scale experiments. Water quality data at the time of sampling can be found in Table 2.3.

2.2.4.3 Wastewater Effluent

Boulder Wastewater Treatment Plant is located in Boulder, Colorado. The wastewater tertiary effluent (Boulder WWEff) served as a wastewater effluent source water for the bench scale biofilters. Water quality data at the time of sampling can be found in Table 2.3.

2.2.4.4 Betasso Water Treatment Plant Influent

City of Boulder's Betasso Drinking Water Treatment Plant's raw water served as the influent for the pilot plant experiments. Betasso WTP's raw water was comprised of a combination of Barker Reservoir and Lakewood Reservoir. The source water characteristics for the duration of the pilot plant can be seen in Table 2.3

Table 2.3 Source Water Characteristics

Source	*DOC (mg/L)*	*UVA_{254} (cm^{-1})*	*SUVA (L/mg-C/m)*	*Alkalinity (mg-CaCO3/L)*	*Temperature (°C)*	*pH*
Betasso WTP	2.1-7.0	0.01-0.23	1.2-3.5	11-20	5-20	6.5-7.9
Barker Reservoir	3.5	0.103	2.8	20		7.2
Wonderland Lake	10.3	0.162	1.6	180		8.4
Boulder WWEff.	6.7	0.141	2.12	110		7.14

2.2.5 Biofilter Sampling

2.2.5.1 Laboratory Bench Scale Sampling

Liquid samples were collected from the influent feed and at sampling ports immediately after each biofilter column to represent respective EBCTs. Liquid samples were collected in amber glassware that had been previously cleaned with deionized water and muffled at 550 ˚C for 3 hours.

2.2.5.2 Pilot Scale Sampling

Influent feed water samples were collected at the tap directly between the sedimentation basin and the filters. Subsequent EBCT samples were taken at the corresponding EBCT tap. Liquid samples were collected in amber glassware that had been previously cleaned with deionized water and muffled at 550 ˚C for 3 hours. Samples were then transported to the University of Colorado, Boulder to be analyzed.

2.2.6 Water Quality Analysis

Water quality analysis was conducted routinely on biofilter influent and effluent samples. Table 2.4 outlines the water quality measurement, the lab instruments, and analysis method employed. Further descriptions of fundamental parameters can be seen below.

Table 2.4 Water Quality Analysis, Instruments and Methods

Analyte	*Measuring Units*	*Detection Limit*	*Equipment/Procedure*	*Reference method*
pH/Temp	N/A	N/A	Denver Instruments Model 220 pH and conductivity meter	SM 4500-H^+
TOC/DOC	ppb	4	Sievers 5310 C TOC	SM 5310 C
TN	mg/L N	1-16	Hach DR 4000 UV Spectrophotometer/Hach Method TNT826	
Ortho-phosphorus	mg/L – PO_4^{3-}	.045	Hach DR 4000 UV Spectrophotometer/Hach Method 8048	SM 4500-P E
UVA	cm^{-1}	0.001	Hach DR-4000 UV Spectrophotometer	SM 5910 B
Alkalinity	mg/L as $CaCO_3$	2	Hach Digital Titrator Model 16900-01	SM 2320 B
Free chlorine	mg/L as Cl_2	0.02	Hach Pocket Colorimeter/Hach Method 8021	SM 4500-Cl G
NH_3	mg/L NH_3-N	0.015	Hach DR 4000 UV Spectrophotometer/ Hach Method 10205	
NO_3	mg/L NO_3-N	0.23	Hach DR 4000 UV Spectrophotometer/ Hach Method 10206	
NO_2	mg/L NO_2-N	0.015	Hach DR 4000 UV Spectrophotometer/ Hach Method 10207	EPA Method 354.1
BDOC	mg/L	N/A	5-day batch test	Mogren et al. 1990
THM/HANs	µg/L	0.1-0.79	Gas Chromatography (6890 GC, Agilent Technologies, CA)	EPA Methods 551.1
HAAs	µg/L	0.5-1.2	Gas Chromatography (6890 GC, Agilent Technologies, CA)	EPA Methods 552.2

Polysaccharides (EPS)	µg/g	Hach DR-4000 UV Spectrophotometer	Taylor (1995)
Fluorescence spectroscopy	EEMs	FluoroMax-4 spectrofluorometer (John Yvon Horiba, NJ)	Korak et al. (2014)

2.2.6.1 Dissolved Organic Carbon

DOC concentrations were measured at the University of Colorado, Boulder on a Sievers M5310 C Laboratory Organic Carbon Analyzer using the ultraviolet irradiation/persulfate oxidation method (SM 5310C). The samples were collected and immediately filtered through a 0.45 mm membrane filter (Pall Life Sciences). Filters were first rinsed with 250 mL of reverse osmosis water to ensure that carbon leaching from the filters did not occur. After filtration, the samples were stored at 4°C until DOC analysis. All DOC analysis was performed within the hold time of 2 weeks of sample collection. Samples were taken in duplicate and analyzed in groups of four with a blank in between as a quality control measure to ensure stable operations and no organic carbon carryover from previous samples. The instrument was calibrated in accordance with the Operations and Maintenance manual. Quality assurance and quality control tests were performed monthly to ensure instrument accuracy.

For the pilot study, the samples were collected at Betasso WTP and immediately filtered on site. After filtration, the samples were transported to CU-Boulder and stored at 4°C until DOC analysis.

2.2.6.2 Biodegradable Dissolved Organic Carbon

BDOC was measured using the 5 day biofilter column test adapted from Mogren et al. (1990). BDOC was determined by subtracting the DOC remaining after 5 days of recirculating the feed water through an acclimated column at room temperature from the initial DOC of the feed water, as seen in Equation 2.1.

$$BDOC = DOC_i - DOC_{5days} \quad \text{Eq. 2.1}$$

2.2.6.3 Ultraviolet Absorbance

Ultraviolet absorbance at 254 nm (UVA_{254}) was measured using a HACH DR/4000 Spectrophotometer. Samples were analyzed using a 1-cm quartz cell and absorbance values were reported with units of cm^{-1} as spectral absorbance coefficients. Samples were taken directly out of the same vials that were run on the Sievers TOC Analyzer. UVA analysis was performed within two weeks of sample collection and stored at 4°C.

2.2.6.4 pH

pH was measured at Betasso WTP and CU labs using a HACH HQ40 Portable Meter and an IntelliCAL PHC725 pH electrode. The pH probe was calibrated weekly with pH values of 4, 7, and 14. This range of pH allows for the full range of values that are expected throughout the tests. Measurements were made by directly inserting the probe into each vial after sampling.

2.2.6.5 Turbidity

Turbidity was measured at Betasso WTP and CU labs in accordance with Method 2130 B (Standard Methods, 1998) using a HACH 2100N Turbidimeter. Turbidity measurements are reported as nephlometric turbidity units (NTU) and were measured on unfiltered water. Measurement vials were stored at room temperature and pre-rinsed with deionized water and dried before using to assure no dilution of samples.

2.2.6.6 Alkalinity

At the bench scale, alkalinity was measured with a HACH Digital Titrator Model 16900-01. At the pilot scale, influent alkalinity samples were taken biweekly and measured at Betasso WTP using a titration apparatus with 0.020N Sulfuric Acid Standard Solution. Alkalinity was measured to examine the buffer capacity of the influent water.

2.2.6.7 Disinfection Byproducts (DBP)

DBP formation was evaluated on all three raw and treated source waters: Barker Reservoir, Wonderland Lake and Boulder WWeff (Chapter 5). Bench scale chlorination was used to measure DBP formation following the uniform formation conditions (Summers et al.,

1996). A 24-hour chlorine residual of 1.0 mg/L (±0.4 mg/L) was determined by chlorine demand curves. The DPD (*N,N-diethyl-p*-phenylenediamine*)* colorimetric method (SM4500-Cl G) was used to measure the chlorine residuals and ammonium chloride was used to quench the samples, immediately following the 24-hour period. The samples were buffered with pH 8 borate buffer. Total trihalomethanes (TTHMs) and haloacetonitriles (HANs) were analyzed using EPA Method 551.1 (1995). Haloacetic acids (HAAs) were analyzed using EPA Method 552.2 An Agilent 6890 Gas Chromatography system with an electron capture detector (ECD) was used to analyze the samples.

2.2.7 Biomass Measurements

2.2.7.1 Media Extraction

Media samples were extracted from the top of the columns for the bench scale experiments and at each depth tap for the pilot scale experiments. The columns were drained, then a sterilized spatula was used to remove roughly 3g from the sample port and immediately placed in a 15-mL sterile conical tube.

At the bench scale, samples were taken before and after each temperature change to assess activity changes associated with each temperature. At the pilot scale, media samples were extracted weekly from each column in order to measure biomass activity throughout column depth. Samples were taken at the influent, 5 min, 15 min and 30 min sample ports.

2.2.7.2 Adenosine Triphosphate

Adenosine triphosphate (ATP) concentrations were assayed with a LuminUltra Deposit Surface Analysis kit (DSA-100C, Fredericton, NB) following the manufacturer's instructions. Activity was measured directly after each sampling period. For ATP analysis, each media sample was first drip-dried using a vacuum filter with a 0.45-micron filter. Approximately 1 gram of media was weighed into a test tube and extracted (The LuminUltra Deposit and Surface Analysis test). Samples were vortexed after each step to ensure sufficient mixing. A

luminometer (Kikkoman C-110) was used to read light output from samples and results were given in relative light units (RLUs). The RLUs were converted to pg cATP/g using the ratio of the RLU's of the sample to the blanks and the mass of the sample. The detection limit varied slightly by the age of the luciferase, but generally lies at about 10,000 RLU's. This was determined by running a blank with the luciferase enzyme and a blank solution. The dry:wet ratio for the media and the bed density were determined to normalize all results to biomass per mL of dry media. The dry:wet ratio was found by weighing a known mass of wet media into a vial of known mass, dying the sample at 100°C for 24 hours, desiccating the samples for 24 hours, then re-weighing the dry sample vial. The dry:wet ratio was calculated by subtracting the weight of the glassware from the media sample weights. Bed densities for the media samples were calculated by filling a 100-mL graduated cylinder of known mass with 100-mL of wet media, drying the cylinder with media at 105 ˚ C for 12 hours, and re-weighing. The weight of the glassware was subtracted from the total weight, which yielded the media dry weight and the density results were reported in dry grams per cubic centimeter. Results were then converted to ngATP/cm^3 media using a dry:wet ratio of 57% and an anthracite bed density of 0.8 g dry weight/cm^3, as following: [pgATP/g(wet)]/[.57 g(dry)/g(wet)]*[.8 g(dry)/cm^3)*(1ng/1000 pg)= ngATP/cm^3. For consistency in ATP data, the authors of this paper and other researchers suggest biomass activity reported on a per unit of media volume basis (Urfer et al. 1997, Pharand et al. 2014).

2.2.7.3 Phospholipid Biomass

The phospholipids method used for the biofilter media samples was developed by Wang et al. (1995) after being adapted from a soil assay method developed by Findlay et al. (1989). In the Lipid Phosphate Analysis for Viable Microbial Biomass Determination method, phospholipids are extracted from 0.2 grams of filter media into chloroform. After a drying

process, the phospholipids are mixed into a solution that is dyed with malachite green. The solution absorbance is read using a spectrophotometer (HACH DR 4000). A standard curve is created using a phosphate solution to correlate absorbance to phospholipid molar concentrations. Three blanks are run with every test and samples are ran in triplicates. The detection limit of this method is the absorbance of blanks, which averages approximately 0.080 cm^{-1}. Samples were extracted and stored at 4°C until all samples have been extracted from the filter. Phospholipid biomass concentrations are reported on a volumetric basis (nmol PO_4/mL) using a packed bed density of 0.8 kg L^{-1} for anthracite and the dry:wet ratio of the media. The biofilter packed bed density was determined by packing a 100-mL graduated cylinder with wet media and measuring the mass before and after drying at 105 ˚C for 12 hours.

2.2.7.4 Polysaccharides

First polysaccharides were extracted from the media using a centrifuge (BECKMAN Model J-21C), phosphate buffered saline (PBS), sonicator, and vortexer. Approximately 2g of media were removed from the biofilter and added to 15 mL centrifuge tube along with 10 mL PBS. In order to dislodge the biofilm from the anthracite, the tube was submerged in a sonicator bath for 1 minute and vortexed for 5 seconds and repeated 5 times. The supernatant (8 mL) was transferred to a clean 50 mL centrifuge tube and centrifuged for 15 minutes at 10,000 rpm at 4°C to separate the free and bound extracellular polymeric substances (EPS). The supernatant was the free EPS and the pellet was the bond EPS. 5.5 mL of the supernatant was transferred to a clean 15 mL centrifuge tube to measure polysaccharides.

The Phenol-Sulfuric acid method (Taylor, 1995) was used to measure polysaccharides. Seven dextrose (D-glucose) standard solutions were used as the calibration curve and a 5% phenol solution and sulfuric acid were used to measure polysaccharides. The samples were

measured on a UV-Vis HACH spectrophotometer at wavelength of 488nm and converted to µg/g using the standard curve.

2.2.8 Data Analysis

All statistical analysis was performed with RStudio version 1.0.153.

2.3 Method Verification

In order verify the scale-up methodology, DOC removal from a drinking water source was tested at the bench scale using the batch/single pass method and a pilot scale flow through system. Betasso WTP influent was used as the raw water in both cases. Temperature varied seasonally at the pilot scale and was a systematic change at the bench scale. At each temperature and EBCT evaluated, the bench and pilot biofilters DOC removals were statistically similar. Temperature impacted DOC removal directly as DOC removal increased with higher temperatures at both scales. The results of the experiments with DOC removals occurring in the same range for each EBCT and temperature at the respective scales with error bars representing standard deviation of duplicates can be seen in Figure 2.4. Removals at all temperatures for the 15 min EBCT at the bench- and pilot- scale are not statistically different based on standard deviation of samples. At 30 minute EBCT, bench- and pilot-scale DOC removals are statistically similar at all temperatures except for 14°C, when removals have a difference of 3% across scales and a low standard deviation. At 5 °C and 30 minute EBCT, DOC removal averaged to 12% removal at the pilot scale and 10% at the bench scale, which is comparable to literature data (LeChevallier et al. 1992, Urfer et al. 1997, Emelko et al. 2006). At 22 °C and 30 min EBCT, DOC removal averaged to 22% removal at the bench-scale, but the pilot scale did not experience temperatures higher than 18°C. Experimental results indicate the batch/single pass bench-scale methodology compares to pilot-scale flow through systems, yet more experimental results are needed.

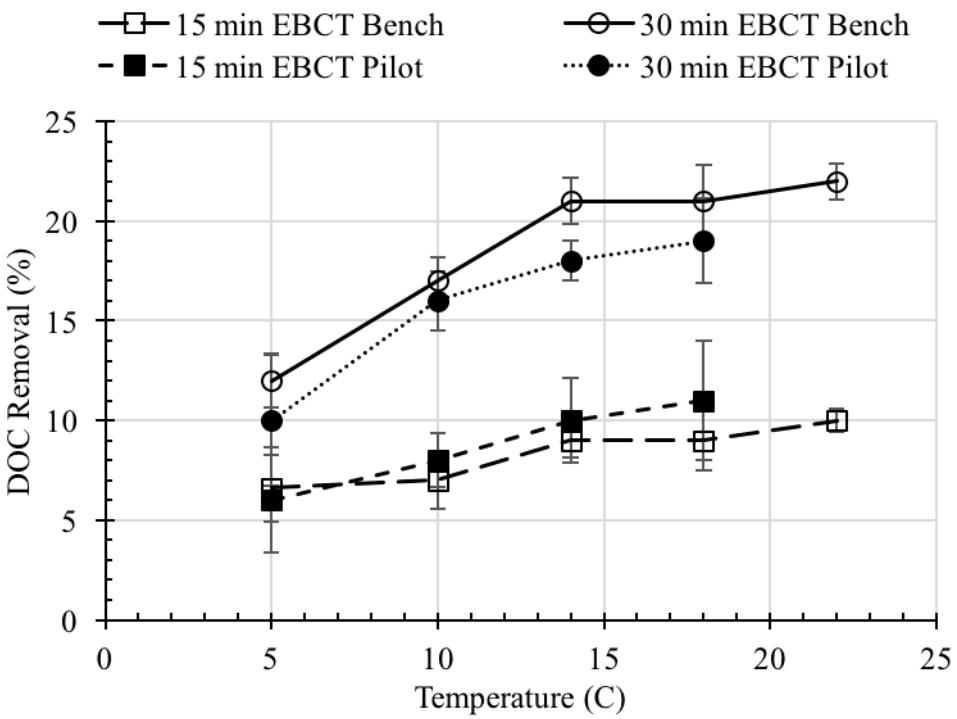


Figure 2.4 Bench scale (batch/single pass) compared to pilot scale (single pass) for DOC removal

Activity levels showed similar trends with increasing temperature at both the pilot and bench scales, but were not statistically similar at each temperature and depth. The activity levels were higher at the top and middle of the biofilters for the bench scale experiment compared to the pilot scale experiment. For all scales and depths, activity increased as temperature increased from 5 to 18 °C. At the top of the filter for the bench scale biofilters activity ranged from 446 to 965 ng ATP/cm^3, while at the pilot scale, the top of the filter activity ranged from 225 to 594 ng ATP/cm^3. At the middle of the filter for the bench scale biofilters activity ranged from 348 to 617 ng ATP/cm^3, while at the pilot scale, the middle of the filter activity ranged from 62 to 422 ng ATP/cm^3. Therefore, activity levels were not similar for the bench and pilot scales as seen in Figure 2.5. A possible explanation for the difference in activity levels could be due to a constant level of influent DOC at the bench scale, yet varied levels of influent DOC at the pilot scale.

Pilot scale activity levels were impacted by low levels of influent DOC with an intermittent surge in DOC during spring runoff. Another possible explanation is the time of sampling. The pilot scale experiments were performed first, then the media was transferred to the bench scale experiments. The additional exposure time for the bench scale experiments could have impacted the activity levels. Therefore, discrepancies in activity levels could be a result of seasonal variables at the pilot scale compared to constant variables at the bench scale, and time of experiments.

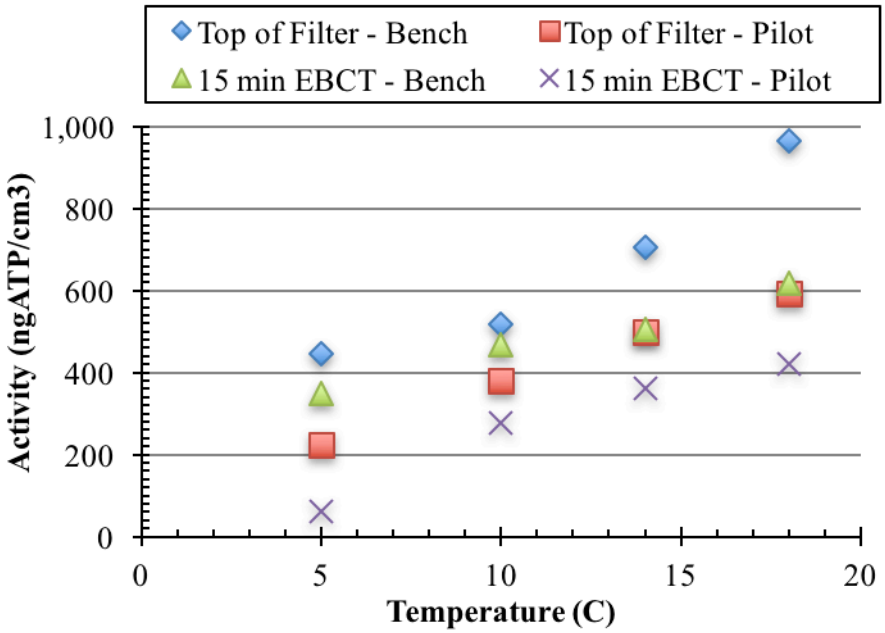


Figure 2.5 Bench scale (recirculation/single pass) compared to pilot scale (single pass) for activity (ATP) measurements

The activity values at each temperature and depth normalized to the activity at 18 °C for that specific depth showed similar trends, as seen in Figure 2.6. Temperature had an impact on activity and the impact was similar for pilot and bench scale biofilters. At 5 °C, normalized activity levels were between 0.38-0.56 of the activity at 18 °C in the same location. However, the 15-minute sample point in the pilot biofilter had a much lower normalized activity because the 5 °C sampling point occurred during the first week of operation. The acclimated media had been in

full scale operation for years, yet the media was not fully acclimated to the influent Betasso WTP DOM matrix at that time. At 10 °C, normalized activity levels to 18 °C activity levels were in the range of 0.54 and 0.76, and at 14 °C, they were in the range of 0.67 and 0.84. This trend suggests temperature impacts activity levels and further validates our bench scale batch/single pass method to a single pass flow through pilot scale method.

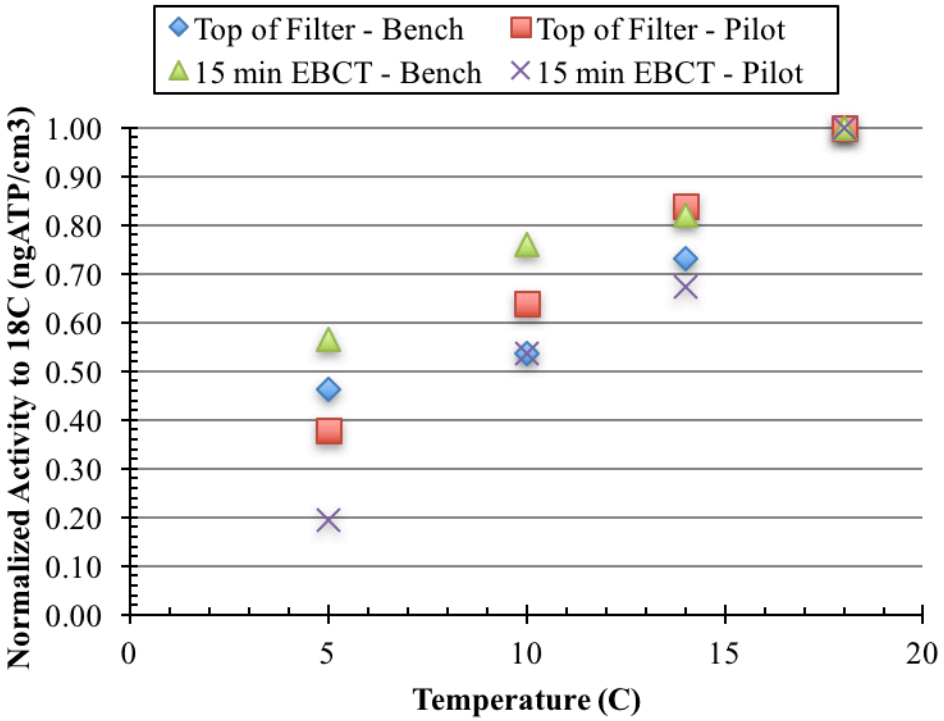


Figure 2.6 Bench scale (recirculation/single pass) compared to pilot scale (single pass) for activity (ATP) measurements normalized to activity measurements at 18 °C

2.4 Conclusions

The experimental results demonstrated that a batch/single pass bench scale biofilter can produce similar organics removal to a flow through pilot scale biofilter and potentially serve as an alternative to pilot-scale testing. In addition, the activity levels showed similar trends in both pilot and bench scale biofilter experiments. This novel bench scale methodology that incorporates a batch reactor and a single-pass flow through reactor allows arduous pilot scale

experiments to be replaced with streamlined bench scale testing, which would expedite biofilter implementation in DWTPs.

Chapter 3
Modeling Biological Filtration Performance for Organic Matter Removal

3.1 Introduction

Dissolved organic matter (DOM) is found in all surface waters and is comprised of a biodegradable organic matter (BOM) and non-biodegradable organic matter. Drinking water utilities face the task of controlling BOM as it can lead to biological regrowth in the distribution system, form disinfection byproducts (DBPs) (Miltner & Summers, 1992) and ozonation byproducts (OBPs) (Carlson & Amy, 1997), and can be specific contaminants, i.e., taste and odor (T&O) compounds (Westerhoff et al., 2005; Zearley & Summers, 2012). For existing and planned surface water treatment plants, biologically active filters (biofilters) have the potential for economically controlling these compounds. Biofilters utilize the indigenous microbial population attached to the filter media to oxidize the BOM. Complete control of some specific compounds can be achieved while complete removal of other compounds and BOM fractions may be limited by the biofilter design and operation (Zearley & Summers, 2012) or recalcitrant nature of the compound. The effectiveness of a biofilter for a given compound is impacted by the concentration and activity of the biomass in a filter and the residence time or empty bed contact time (EBCT) of the filter. For example, longer EBCT have been shown to increase DOC removal (Peldszus et al., 2012; Terry & Summers, 2017; Zhang et al., 2017). Other water quality factors, such as water temperature and BOM concentration can also have an impact, as reviewed in Chapter 1.

Modeling biofilter performance is advantageous in order to optimize treatment efficiency. In the low BOM concentration range typically found in source waters used by drinking water utilities, the microbial community utilizes an ill-defined subset of biodegradable organic

compounds to provide energy and biomass growth. There is likely not a single compound that serves as a primary substrate, but a conglomerate of compounds with similar structure and utilization rates that collectively provide the energy for biomass growth and energy. Several predictive models, which relate back to Monod kinetics, have been proposed, but most require site specific values for the multiple model parameters (Billen et al., 1992; Chen et al., 2016; Hozalski & Bouwer, 2001a; Huck et al., 1994; Rittmann & McCarty, 1980; Rittmann & Stilwell, 2002; Sáez & Rittmann, 1988; Wang & Summers, 1994; Zhang & Huck, 1996).

3.1.1 Biomass Measurements

Numerous methods are available to measure biomass concentration and biomass activity in biofilters (Pharand et al. 2014). This study focuses on phospholipids (PL) as a biomass concentration measure and adenosine triphosphate (ATP) as biomass activity measure. Phospholipids, located in the cell membrane, are molecules comprised of long lipid tails and phosphate heads and have been used to estimate microbial biomass (Findlay et al. 1989). The PL method of assessing biomass, while sensitive, is labor and time intensive and does not differentiate between live and dead cells (Pharand et al., 2014). ATP is a molecule used for energy transport in cells and is the primary energy carrier for all living organisms, therefore it can indicate bacterial metabolic activity (Prescott et al., 2005; Velten et al., 2007). Historically, the ATP biomass activity method was encumbered by the need to remove the biomass from the filter media. However, recent advances have overcome that problem (Evans et al., 2013). Recent data have supported ATP biomass activity measurements as a monitoring tool in drinking water (Hammes et al., 2010). The development of a direct method of predicting the control of BOM by biofilters has been evasive.

3.1.2 Effects of Temperature

Water temperature seasonal variations occur for most water treatment plants and can be quite significant, varying up to 20 or 30 degrees Celsius (LeChevallier et al., 1992; Moll et al., 1999). Research has shown that as temperatures decrease, DOC removal through biofiltration decreases (Evans et al., 2013; Fonseca et al., 2001; Hallé et al., 2009; Huck et al., 2000; Moll et al., 1999; Pharand et al., 2015; Stoddart et al., 2016; Terry & Summers, 2017). At lower temperatures, the microbial community structure can change and the rate of substrate metabolism decreases (Moll et al., 1999), and bacterial growth rates and the kinetics of attachment also decrease (Huck et al., 2000). However, literature expectations of this trend show conflicting results. Seger and Rothman (1996) reported that at cold temperatures (<5 °C), slow sand filter ATP biomass activity concentrations decreased. Hallé et al. (2015) found ATP biomass activity concentration in pilot-scale biofilters changed as a function of water temperature. Comparably, Huck (2000) reported PL biomass concentration decreased at the top of the biofilters when the filters were operated at 1°C - 3 °C compared to 21°C - 25° C. However, some studies found no correlation between ATP biomass activity and temperature in biofilters in temperature ranges of 3 to 28 °C (Pharand et al., 2014), 10 to 24 °C (Rahman, 2013), and 1 to 23° C (Stoddart et al., 2016). Evans et al. (2013) found no correlation with ATP biomass activity and temperature for four full scale biofilters experiencing seasonal water temperature changes. Other studies have shown no correlation between PL biomass concentrations and seasonal temperature variations (Fonseca et al., 2001; Persson et al., 2006). These inconsistencies in literature could be due to different upstream water treatment plant operations, methods of analysis, water quality characteristics, or scale.

3.1.3 Biomass Development and Impact of Operation

The development of biomass in a filter begins immediately upon startup if there is no disinfectant residual in filter effluent. Given the variation in source water temperature, pretreatment conditions, and BOM type and concentration a set time for biomass development cannot be expected. Biomass development of three demonstration scale filters and two pilot scale filters is shown in Figure 3.1 for a nonozonated water. The biomass increased over the first 6 months of operation, April to September, and biomass development was likely impacted by the increase in the influent TOC concentration, from 1.5 to 2.5 mg/L and increase in the temperature, more than 10 °C during this time (Chowdhury et al., 2009). In this study, filter media type and backwash conditions did not seem to impact the biomass development time. Wang et al. (1995) also showed a similar phospholipid biomass development time for an ozonated water and a lack of impact of media type. They also showed that the biomass decreased by about 50% when preozonation was terminated.

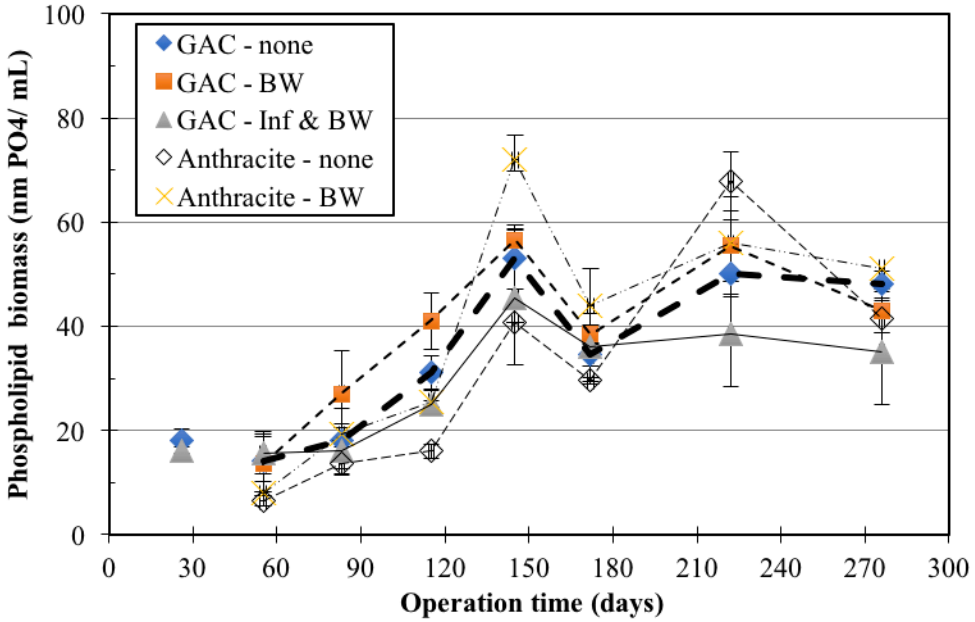


Figure 3.1 The impact of media type, chlorinated backwash water and influent chlorine residual on the development of PL biomass concentration for three GAC filters and two anthracite filters (EBCT of 7.1 min) (Chowdhury et al., 2009).

The impact of operating conditions on biomass was observed in the results of three studies (Chowdhury et al., 2009; Dugan, 1998; Wang et al., 1995), which are reported in Appendix B Table B1. The impact of media type indicated that on a volume filter basis that more biomass was associated with GAC, followed by sand and with the lowest amount associated with the anthracite media (Chowdhury et al., 2009; Wang et al., 1995). Preozonation yielded high amounts of biomass (Wang et al., 1995; Dugan, 1998). A chlorine residual in the influent all but eliminated biomass in the biofilter with anthracite as the media but not with the GAC, as the GAC reacted with the chlorine in the top part of the filter, it allowed the biomass to grow in the remaining part of the filter (Wang et al., 1995; Chowdhury et al., 2009). Chlorine in the backwash water decreased the biomass in the biofilters with anthracite, but had less impact on the biofilter with GAC media (Chowdhury et al., 2009; Wang et al., 1995;). Again, the GAC reacted with the chlorine such that it does not penetrate far into the filter depth during backwashing. Backwashing with a chloramine residual had less of a negative impact on the biomass (Dugan, 1998).

3.1.4 Objectives

The objectives of this study were to: 1) evaluate the relationship between ATP biomass activity and PL biomass concentration and their response to temperature variations, 2) evaluate and model the biomass depth and distribution profile, and 3) model the BOM removal by biofilters.

The approach taken included an extensive literature search to acquire past biofiltration performance and biomass data. A model that predicted biomass distribution in a biofilter, using data from the literature, was developed. PL biomass concentration data was converted to ATP biomass activity using Dowdell (2012) conversion. Temperature correction factors determined

from bench scale experiments were used to correct ATP biomass activity data measured at lab temperature to that of the field temperatures. Next, a model was developed to predict TOC removal throughout the biofilter at various temperatures and oxidation conditions, using data from the literature. Finally, the data from the literature was used to predict ATP biomass activity at the top of the filter using influent water quality parameters.

3.2 Materials and Methods

3.2.1 Bench Scale Experiments

Glass chromatography columns with an 11-mm inner diameter, Teflon caps (5820-12 and 5820-24, Vineland, NJ), Masterflex tubing, and stainless-steel metal fittings (Swagelok Cleveland, OH) were used for constructing the biofilters. Acclimated biologically active anthracite media, with an effective size of 1.0 mm, was taken from pilot plant biofilters (City of Boulder Betasso Water Treatment Plant) and packed into two columns connected in series with a media volume of 104 mL. To prevent media loss or clogging, the bottom of each column was packed with 5 cm of support media, i.e., 2 mm diameter glass beads encased in a wire mesh. A needle valve was used after each column to control flow. Sampling ports were located immediately before and after each column to assess removal associated directly with the specific filter depth. The columns were covered to minimize growth of photosynthesizing microorganisms in the filters. All columns were run at a hydraulic loading rate of 2.44 m/hr. The flow rate was 3.48 mL/min and the EBCTs were 15 and 30 minutes. The support media depth was not included in the calculation of the EBCT. The columns were operated in down-flow orientation using peristaltic pumps (Masterflex Models 7553-30 and 7518-10).

3.2.2 Phospholipid Biomass Concentration Analysis

The PL biomass concentration method used for the biofilter media samples was developed by Wang et al. (1995) after being adapted from a soil assay method developed by Findlay et al. (1989). In the Lipid Phosphate Analysis for Viable Microbial Biomass Determination method, phospholipids were extracted from 0.2 grams of filter media into chloroform. After a drying process, the phospholipids were mixed into a solution that was dyed with malachite green. The solution absorbance was read using a spectrophotometer (HACH DR 4000). A standard curve was created using a phosphate solution to correlate absorbance to phospholipid molar concentrations. The detection limit of this method was the absorbance of blanks, which averaged approximately 0.080 cm^{-1}, representing 0.0 nmol PO_4. Samples were extracted and stored at 4°C until all samples had been extracted from the filter. Samples were extracted from the top of each column with a sterile spatula before and after each temperature change.

3.2.3 ATP Biomass Activity Analysis

ATP biomass activity were assayed with a LuminUltra Deposit Surface Analysis kit (DSA-100C, Fredericton, NB) following the manufacturer's instructions. Activity was measured on-site, directly after each sampling period, which eliminated temperature changes in the media after extraction.

The dry: wet ratio for the media and the bed density were determined to normalize all results to biomass ngATP (or nmolPO4) per mL of dry media. The dry: wet ratio was found by weighing a known mass of wet media into a vial of known mass, dying the sample at 100°C for 24 hours, desiccating the samples for 24 hours, then re-weighing the dry sample vial. The dry:wet ratio was calculated by subtracting the weight of the glassware from the media sample

weights. Bed densities for the media samples were calculated by filling a 100-mL graduated cylinder of known mass with 100-mL of wet media, drying the cylinder with media at 105 ˚ C for 12 hours, and re-weighing. The weight of the glassware was subtracted from the total weight, which yielded the media dry weight and the density results were reported grams per liter. The average bed density for GAC, anthracite and sand were 500 g/L, 799 g/L and 1,200 g/L, respectively. Samples were extracted from the top of each column with a sterile spatula before and after each temperature change.

3.2.4 Liquid Sample Analysis

Liquid samples were collected from the influent feed, a sampling port after column 1 (15 min EBCT) and the effluent of column 2 (30 min EBCT). Samples were collected in amber glassware that was previously cleaned with deionized water and muffled at 550 ˚ C for 3 hours.

3.2.5 Dissolved Organic Carbon

Biodegradable organic matter was measured as dissolved organic carbon (DOC). DOC concentrations were measured at the University of Colorado, Boulder on a Sievers 5310 C Laboratory Organic Carbon Analyzer using the ultraviolet irradiation/persulfate oxidation method (SM 5310C). The samples were collected and immediately filtered through a 0.45 μm membrane filter (Pall Life Sciences). Filters were first rinsed with 250 mL of reverse osmosis water to assure that carbon leaching from the filters did not occur. After filtration, the samples were stored at 4°C until DOC analysis. All DOC analysis was preformed within the hold time of 2 weeks of sample collection.

3.3 Results and Discussion

3.3.1 Biomass Depth Profiles Modeling

Higher concentrations of substrate are available at the top of the filter and support higher levels of biomass, which is followed by a decay of biomass due to substrate consumption throughout the filter (Carlson & Amy, 1998; Wang et al., 1995). When the depth of the filter was represented by the EBCT, the biomass distribution was independent of the loading rate (Wang et al., 1995; Zearley, 2012). Zearley (2012) developed a biomass distribution coefficient, ß, to determine the biomass concentration at any give EBCT as well as total and average filter biomass using an exponential decay.

The biomass distribution, both PL biomass concentration and ATP biomass activity, as a function of EBCT was evaluated for 117 data points reported in 26 biofilter studies (Carlson & Amy, 2001; Chowdhury et al., 2009; Dowdell, 2012; Dugan & Summers, 1997; Elhadidy et al., 2017; Hallé, 2009; Huck et al., 2000; Lee et al., 2010; Liao et al., 2013; Moll et al., 1999; Peldszus et al., 2012; Persson et al., 2006; Pharand et al., 2014; Rahman, 2013; Seredynska-Sobecka et al., 2005; Son et al., 2014; Urfer & Huck, 2001, 2000; Velten et al., 2011; Wang et al., 1995; Westerhoff et al., 2005). PL biomass concentrations and ATP biomass activity decreased with depth for filters with both ozonated and nonozonated feed waters. An unsuccessful effort was made to correlate the PL biomass concentration to the influent temperature, TOC (or DOC) concentration, EBCT and preozonation. However, a relationship of the relative ATP biomass activity or PL biomass concentration depth concentration relative to the concentration at the top of the filter, with EBCT was found, Figure 3.2, for both the preozonated and nonozonated waters.

After 11 minutes of EBCT, the relative biomass in the ozonated and nonozonated fed filters were both less than 50% of that at the top. However, the biomass gradient with EBCT was initially much steeper in the filters with ozonated water, as after 3 minutes of EBCT the biomass was about 50% of that at the top, while for nonozonated water this did not occur until after 9 minutes of EBCT.

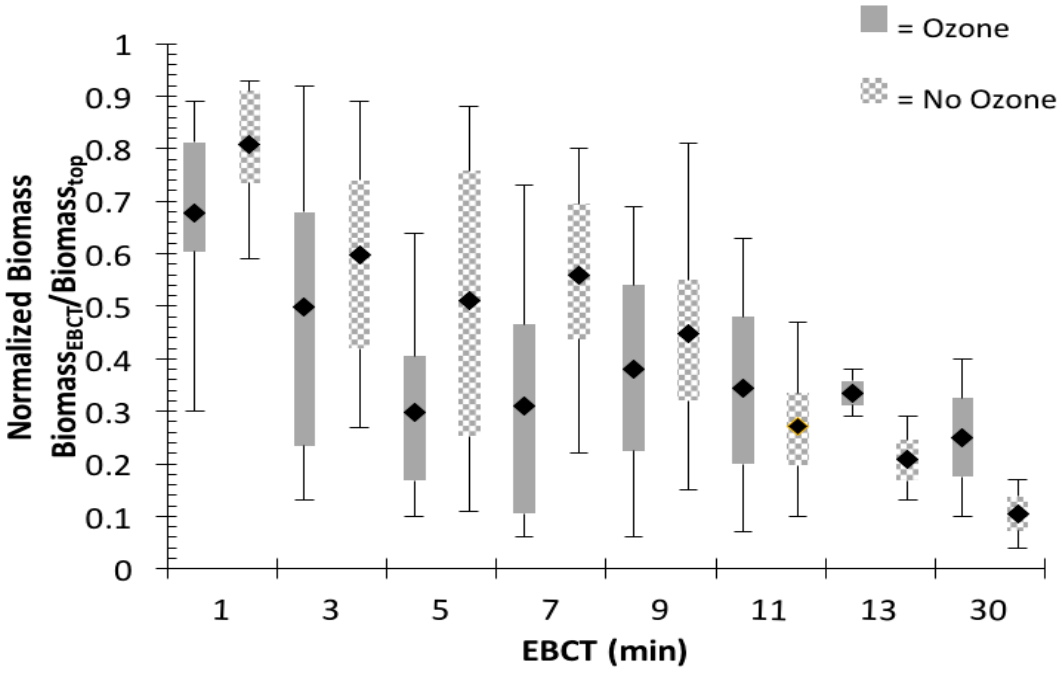


Figure 3.2 Normalized biomass concentration as a function of EBCT and ozonation. PL biomass concentration and ATP biomass activity decreased with increasing filter depth, with a sharper decrease for ozonation waters compared to nonozonated waters. The solid squares represent normalized biomass distribution in ozonated biofilters (n=77) and the dashed boxes represent nonozonated biofilters (n=78). The black diamonds represent the median values, the boxes represent the 25th and 75th percentiles and the whiskers represent the minimum and maximum values.

Based on literature data, the relationship of the normalized biomass concentration, or biomass activity, $X(EBCT)/X_{top}$, can be expressed as, Equation 3.1, with the correlation coefficient values for a and b.

$$X(EBCT)/X_{top} = a*\ln(EBCT) + b \qquad \text{Eq. 3.1}$$

The derivative of the correlation, expressed in Equation 3.2, demonstrated that longer filter EBCTs allow for a shallower slope, or drop-off, of biomass throughout the filter.

$$dx/d(EBCT) = a*X_{top}*1/EBCT \quad \text{Eq. 3.2}$$

The majority of the evaluated data were measured as PL biomass concentration, but including the ATP biomass activity results did not impact the resulting coefficient values, *a* and *b.* With ozone pretreatment, coefficient *a* value was higher and coefficient *b* value was lower than without ozone pretreatment. For all conditions, a ranged from -0.120 to -0.154 and b ranged from 0.69 to 0.76, as shown in Appendix B Table B2.

Caution must be taken when evaluating the biomass at the top of the filter. The ATP biomass activity results of sampling from the very top few grains of media compared to that of 3 inches into the biofilter can indicate up to an 80% reduction in biomass with depth (Appendix B Table B3). Taking a sample from the first three inches (5 cm) and mixing it to get a more homogenous and representative sample is recommended. In nearly all cited cases, the sample was taken from the top few inches of the filter.

Eq. 3.1 was solved to yield the biomass, X, as a function EBCT when the biomass concentration at the top of the filter, X_{top}, was known:

$$X(EBCT) = X_{top}(a*\ln(EBCT)+b) \quad \text{Eq. 3.3}$$

The total biomass in a filter, X_{Total}, to a given depth or EBCT was determined by integrating Eq. 3.3 with respect to EBCT.

$$X_{Total} = X_{top}(a*(EBCT*\ln(EBCT) - EBCT) + b*EBCT) \quad \text{Eq. 3.4}$$

The units for total biomass were product of biomass (nmol PO4 or ng ATP) and EBCT. The average biomass, X_{avg}, was the total biomass divided by filter EBCT.

$$X_{avg} = (X_{top}(a*(EBCT*\ln(EBCT) - EBCT) + b*EBCT)/(EBCT) \quad \text{Eq. 3.5}$$

3.3.2 Total Biomass

To evaluate the relationship between TOC removal and top of filter biomass, literature data from three studies reported in Appendix B Table B1 were used to determine if a relationship exist. The data were plotted and a linear correlation with an R^2 value of 0.69 was found. When additional data from the literature and in-house studies were added to the plot, a linear relationship with an R^2 value of 0.30 was found (Appendix B Figure B1). While the TOC removal was significantly related to biomass ($p<0.05$), a strong regression correlation was not found. The lack of a strong relationship between biomass and TOC removal has been reported by other authors (Pharand et al., 2014; Urfer & Huck, 2001; Wang & Summers, 1993). However, Wang and Summers (1993) found that TOC removal did not correlate to EBCT or biomass, yet there was a strong correlation between TOC removal and the product of EBCT and top of filter biomass, i.e., total microbial activity within the biofilter.

Since TOC removal was found to be a function of total microbial activity (Wang & Summers, 1993), the use of a kinetic model for predicting the biodegradable fraction was evaluated. The utilization rate describing contaminant degradation at low concentrations in biological filters is described by the following second-order reaction equation:

$$r = dC/dt = -k''*X*C \qquad \text{Eq. 3.6}$$

where r is the utilization rate, mass per volume per time, C is the mass concentration, t is the biofilter contact time, X is the biomass (or activity) concentration, and k'' is a contaminant utilization rate constant per biomass concentration per time. This model can be derived from Monod kinetics when the substrate concentrations were low, as is the case in drinking water treatment of BOM (Digiano et al., 2001). Zearley (2012) used a similar model to predict trace organic contaminant utilization.

Integrating this rate equation over filter depth or total EBCT resulted in the following integrated rate equation, which describes the resulting contaminant concentration for a filter with a set EBCT:

$$C_{eff}/C_{inf} = \exp(-k''*X*EBCT) \quad \text{Eq. 3.7}$$

where C_{inf} and C_{eff} are the biofilter influent and effluent concentrations, respectively.
To apply the model to the total organic matter, the biodegradable fraction, BOM_{frac}, must be known, as not all total organic carbon is biodegradable as expressed in Equation 3.8.

$$C_{eff}/C_{inf} = BOM_{frac}(\exp(-k''*X_{avg}*EBCT)) \quad \text{Eq. 3.8}$$

Since the product $X_{avg}*EBCT$ was the total biomass, Equation 3.8 can be expressed in total biomass. If EBCT is expressed in minutes and X_{avg} as nmolPO4/ml, then k'' has units of $ml*nmolPO4^{-1}*min^{-1}$. If X_{avg} is expressed as ngATP/mL, then k'' has the units of $ml*ngATP^{-1}*min^{-1}$.

Total biomass data was correlated to filter effluent TOC (or DOC) concentration for 119 paired values from 35 studies at warm and cold temperatures using Equation 3.8. PL biomass concentrations were used because most of the biomass results were developed with that method. For the data given in activity, ATP biomass activity was converted to PL biomass concentration using Dowdell (2012) conversion equation as seen in Figure 3.3, ATP (ng/mL) = $11*PO_4$ (nmol PO_4/mL). The conversion was developed for a range of media types and water quality scenarios.

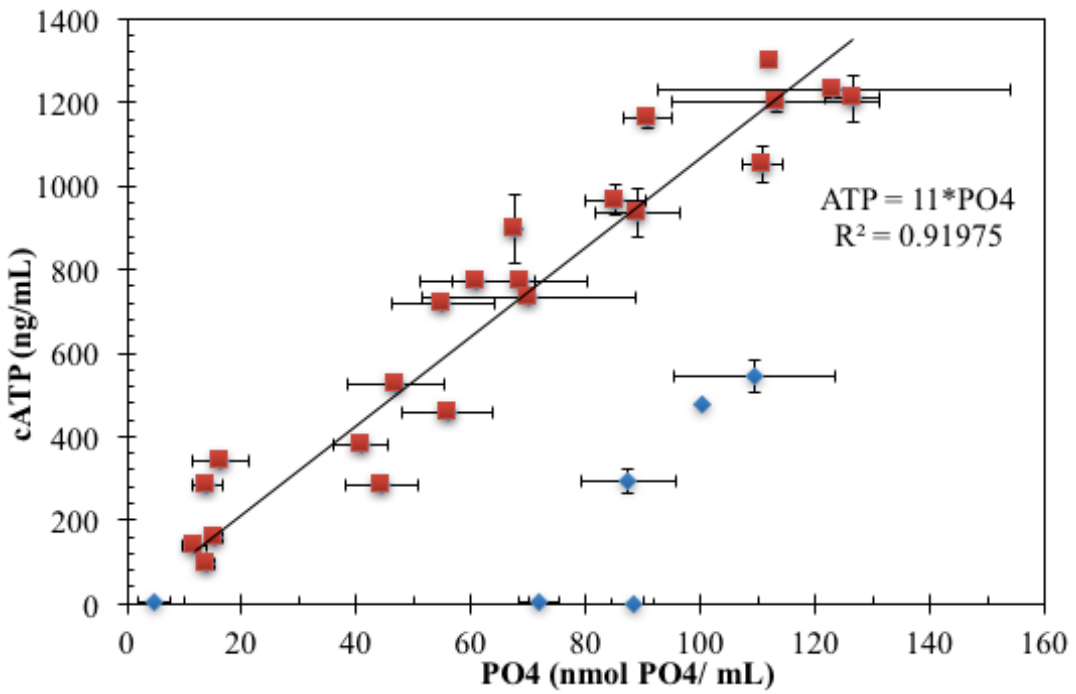
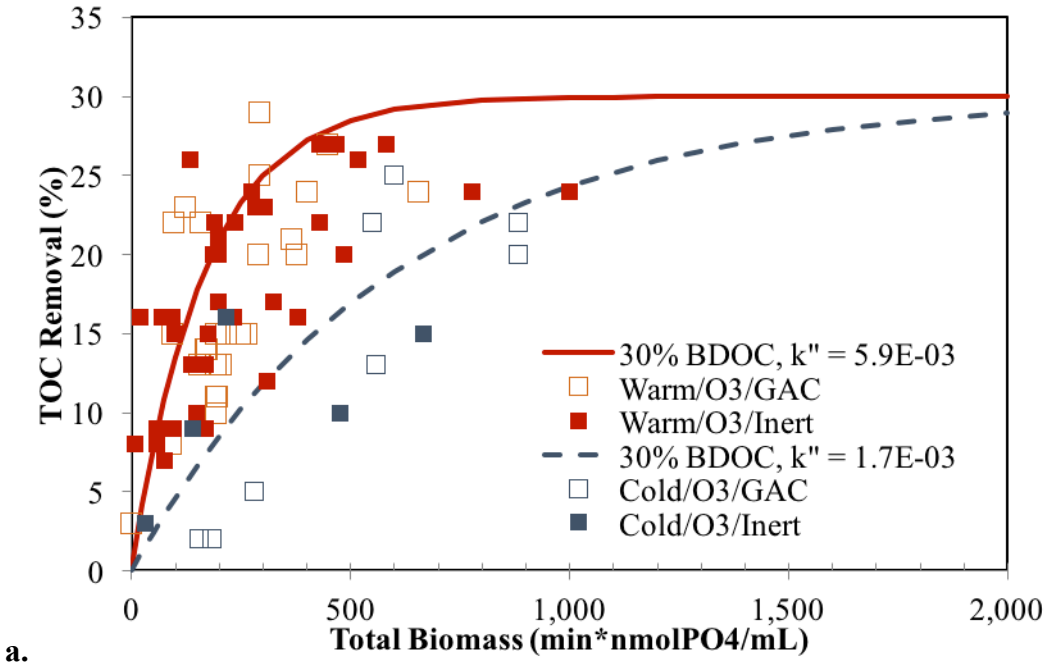


Figure 3.3 ATP biomass activity linear correlated with PL biomass concentration for multiple media types, source waters and influent temperatures. The conversion found is as follows: ATP (ng/mL) = 11*PO_4 (nmol PO_4/mL) (Dowdell, 2012).

The data were divide into four subsets; a) warm (≥10°C) ozonated waters (n=64), b) warm (≥10°C) nonozonated waters (n=36), and c) cold (<10°C) ozonated waters (n=14) and d.) cold (<10°C) nonozonated waters (n=5). (Carlson & Amy, 1998; Chen et al., 2016; Chowdhury et al., 2009; Dugan & Summers, 1997; Elhadidy, 2016; Emelko et al., 2006; Fonseca et al., 2001; Granger et al., 2014; Hallé et al., 2009; Huck et al., 2000; Klevens et al., 1996; Lauderdale et al., 2012; Miltner et al., 1996; Moll et al., 1999; Peldszus et al., 2012; Peleato et al., 2016; Persson et al., 2006; Pharand et al., 2014; Rahman, 2013; Stoddart et al., 2016; Vahala et al., 1998; Velten et al., 2011; Wang et al., 1995; Wang & Summers, 1993; Westerhoff et al., 2005). For each biofilter the average biomass, X_{avg}, was calculated from the reported top of the filter biomass using Equation 3.5. Application of the model to nonozonated and cold/ozonated waters is shown

in Figure 3.4a&b. The type of media, GAC vs. inert, did not impact the results as seen by the hollow and solid symbols representing GAC and inert media, respectively.



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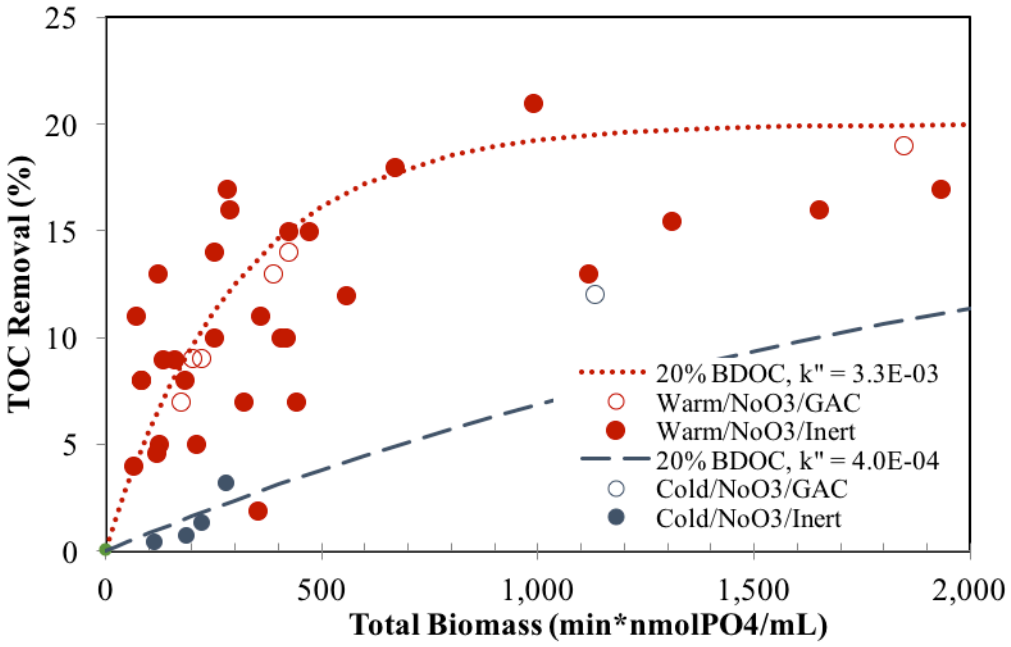


b.

Figure 3.4 TOC removal as a function of total biomass using literature data, (a.) ozonated and (b.) nonozonated, with media separation and multiple k" constant. Measured TOC removal includes the following: warm (≥10°C) ozonated waters (n=64), cold (<10°C)

ozonated waters (n=14), warm (≥10°C) nonozonated waters (n=36) and cold (<10°C) nonozonated waters (5).

For the ozonated waters, a BOM fraction of 0.30 was based on a literature review of ozonated waters in Chapter 2 (Cipparone et al., 1997; Terry & Summers, 2017). The rate constant k″ was adjusted to produce the best fit, which resulted in a k" value of 5.9E-03 $ml*nmolPO4^{-1}*min^{-1}$ for the warm ozonated waters.

The model fit the data well with a TOC removal average residual of 4.1%, i.e., the average of the absolute difference of experimental TOC removal and predicted TOC removal for all scenarios. Media type, GAC and inert media, had little impact on the model fit as TOC removal average residuals were 4.7% and 3.7%, respectively. A residuals analysis, showed that the model underestimated the removal for the first 200 units of total biomass, this result was not unexpected as several authors have reported a fast reacting BOM fraction, which would yield more removal in the top of the filter (Billen et al., 1992; Wang, 1995). The average biomass of these filters was 78 nmolPO4/ml, which if applied to the 200 biomass value would yield an EBCT of 2.5 minutes. Wang (1995) reported that the fast reacting BOM fraction was utilized in the first 3 minutes of EBCT. However, the utilization of this fast reacting BOM fraction was not explicitly considered herein.

For cold waters the BOM fraction was kept at 0.30 due to preozonation and the rate constant was decreased to yield a good fit, which resulted in a k″ value of 1.7E-03 $ml*nmolPO4^{-1}*min^{-1}$. The k″ value was what would be expected at temperatures less than 10° C. Again, the model fit the data well with a TOC removal average residual of 4.1%, with no difference due to media type.

For the nonozonated waters, a BOM fraction of 0.20 was used based on work from Chapter 2 and other literature (Frías et al., 1992; Ribas et al., 1992). The k″ constant best fit was

a k″ value of 3.3E-03 ml*nmolPO4^{-1}*min^{-1}. For the warm, nonozonated waters, the fit resulted in a TOC removal average residual of 3.1%. GAC and inert media average residuals were 1.0% and 3.6%; further suggesting the model worked equally well for GAC and inert media. For the cold, nonozonated waters, a BOM fraction of 0.20 was used and a k″ value of 4E-04 ml*nmolPO4^{-1}*min^{-1} was used to account for lower temperatures. This model fit the data well with an average residual of 1.6%, GAC and inert media TOC removal average residuals were 4.97% and 0.8%, respectively.

For the 119 data points simulated by all four cases, a regression of the measured versus simulated data was carried out. The regression yielded a slope of 0.82, indicating that the model under predicted the removal, and an R^2 value of 0.63, indicating an acceptable fit for a meta-analysis literature biofilter data due to uncontrollable variables, i.e., scale, location, water quality parameters, personal, and laboratories. However, this model approach depends on multiple k" constants for the differing temperatures.

3.3.3 Model Comparison

Models are developed to relate the behavior of biological filters to the kinetics of the microbiological processes occurring within the system, in order to assist optimal design and management of the biofilters. Different steady-state models (balanced growth and decay) have been developed over the years to predict biofilter performance based on operational parameters and water quality conditions (Rahman, 2013; Servais et al., 2005). Under wastewater treatment conditions, Rittmann and McCarty (1980) used Monod kinetics and Fick's law of diffusion and developed the concept of S_{min}, which is the minimum incoming substrate level at which the biofilm can survive. Other models have used Monod kinetics to simulate biomass profile and BOM removal in a biofilter (Hozalski & Bouwer 2001b, Rittmann & Stilwell 2002), and found

that a first order relationship provided a good approximation for removals seen in actual biofilters (Chen et al., 2016; Huck et al., 1994). Zhang and Huck (1996) modeled AOC removal from a packed bed biofilm reactor by adapting a pseudoanalytical solution by Saez and Rittmann (1988), which was built from a steady-state biofilm model developed by Rittmann and McCarty (1980). When modeling AOC removal, Zhang and Huck (1996) found the removal of substrate was proportional to the influent concentration and the average percentage removal of BOM was determined by EBCT. Thus, X^* was used to represent the "dimensionless EBCT of the column, which was measure of the EBCT in relation to the specific area of the biofilm column, diffusivity of the substrate and the rate of biodegradation" in order to provide practical application to the kinetic model. The authors saw the same BOM removal, albeit different combinations of hydraulic loading rate and column depth as long as EBCT remained the same (Zhang and Huck, 1996). Other models include the CHABROL Model, which estimates BDOC removal and biomass distribution by using 12 parameters to describe the kinetics during the biofiltration process (Billen et al. 1992), and the Integrated Biofilm Model (IBM), which is a spreadsheet program that produces iterative design and analysis of biofiltration process (Rittmann & Stilwell 2002).

3.3.4 Temperature Impacts

Temperature affected PL biomass concentration differently than ATP biomass activity, as shown in Figure 3.5. Each temperature value, tested in order of lowest to highest: 5°C, 10°C, 14°C, 18°C to 22°C, showed a corresponding PL biomass concentration, ATP biomass activity and DOC removal rate for top and middle of the filter. The 14°C condition was tested a second time after the 22°C condition to evaluate systematic effects and yielded results similar to those at the earlier 14°C run. The ATP biomass activity between each temperature range and top of the

filter versus that in the middle of filter was statistically different according to a 95% confidence interval. However, biomass concentration (measured in nmolPO_4/mL) remained relatively constant with no statistical difference, despite the temperature increase, as seen in Figure 3.5.

ATP biomass activity is higher at the top of the filter than in the middle and increases with increasing temperature. ATP biomass activity at the top of the filter was on average 124 pg ATP/mL higher than that in the middle of the filter. ATP biomass activity at the top of the filter at the 5°C was 510 ± 30 ng cATP/mL and at the 22°C, activity it increased to 1,392 ± 50 ng ATP/mL. The middle of the filter results demonstrated similar increases in ATP biomass activity. PL biomass concentration data showed higher concentrations at the top of the filter than that in the filter middle. On average, PL biomass concentration was 7.0 nmol PO4/mL higher at the top of the filter than the middle. PL biomass concentration data did not increase statistically between temperature ranges. At the top of the filter, PL biomass concentration at 5°C was 26 ± 3 nmol PO4/mL and at the 22°C, PL biomass concentration was 27 ± 3 nmol PO4/mL. Similar results were seen at the middle of the filter. This suggests that biomass activity was a better indicator of what was occurring in the filter rather than biomass concentration. Biomass activity increased with increasing temperature, but biomass concentration remained constant at certain temperature ranges.

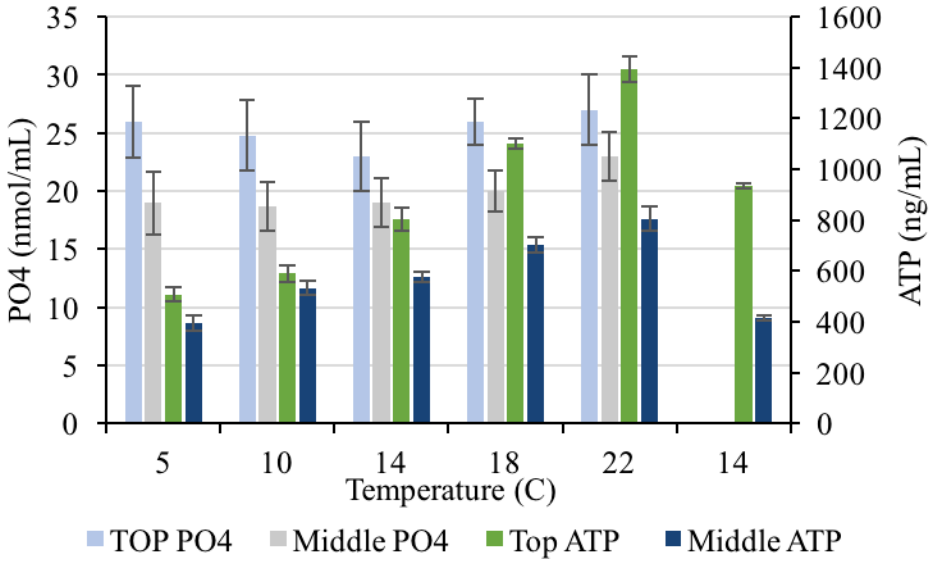


Figure 3.5 Temperature impact on PL biomass concentration (PO4) measurement, and ATP biomass activity measurement.

3.3.5 Temperature Correction

ATP biomass activity was often measured in a lab, but the temperature of full-scale plant filter media can fluctuate with seasonal changes. In addition, media samples were often shipped to labs over ice and then analyzed; therefore, experiencing multiple temperature changes since extraction from the filter. Therefore, a temperature correction factor to account for temperature difference between the filter and the lab was developed. For the top of the filter, Equation 3.10 was developed to determine the normalized ATP biomass activity with respect to 22 °C at the specific full-scale filter temperature. The "ATP_{filter}" value represents activity at the filter temperature normalized to ATP biomass activity at lab temperature, 22 °C. The "T_{filter}" value represents the temperature of the filter water, when the media was extracted. Temperature throughout the filter may fluctuate, therefore at an EBCT of 15 minutes, Equation 3.11 was

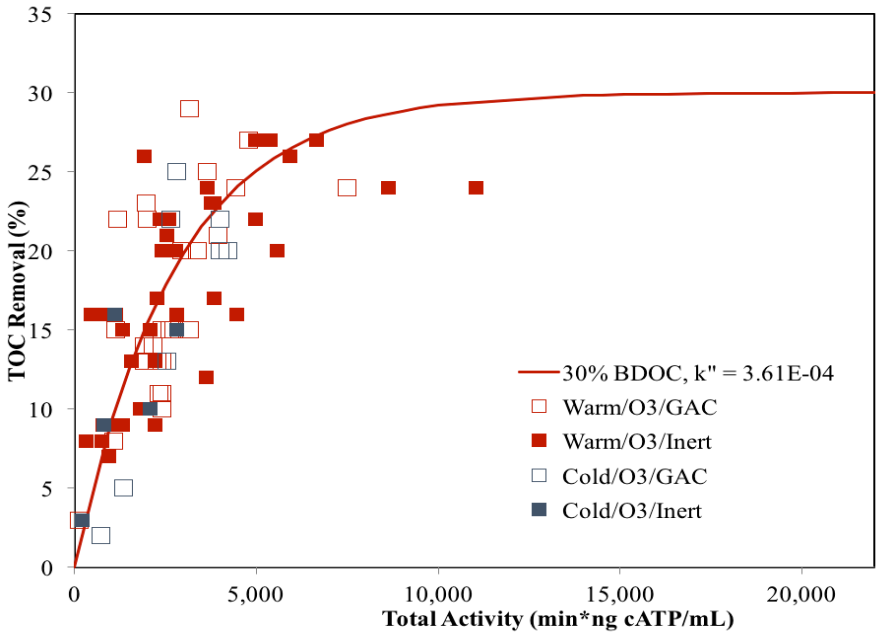
developed to determine the percent of normalized activity to 22 °C, as designated "$ATP_{15minEBCT}$".

$$ATP_{topoffilter} = 0.0385*T_{filter} + 0.0997;\ R^2 = 0.94 \quad \text{Eq. 3.10}$$

$$ATP_{15minEBCT} = 0.0292*T_{filter} + 0.0345;\ R^2 = 0.99 \quad \text{Eq. 3.11}$$

3.3.6 Temperature Correction Verification

To determine if the temperature correction was applicable to data other than in house data, the temperature correction factor was applied to the cold temperature literature data from Figure 3.4, after utilizing Dowdell (2012) conversion equation to convert PL biomass concentration data to ATP biomass activity data. The temperature corrected literature data had a much tighter fit applying BDOC fraction of 0.30 and 0.20 for ozonated and nonozonated data respectively, and applying only one k″ value (k″ = 3.61E-04 $mL*ngATP^{-1}*min^{-1}$), as seen in Figure 3.6. This suggests ATP biomass activity incorporates temperature within the measurement, therefore, one k″ constant, can be used for multiple temperatures. The k″ constant was calculated using the TOC removal, EBCT, and ATP biomass activity from the literature data. Therefore, care must be taken to record temperature when ATP biomass activity samples are taken. The 95% confidence level for k″ was 4.47E-05 $mL*ngATP^{-1}*min^{-1}$.



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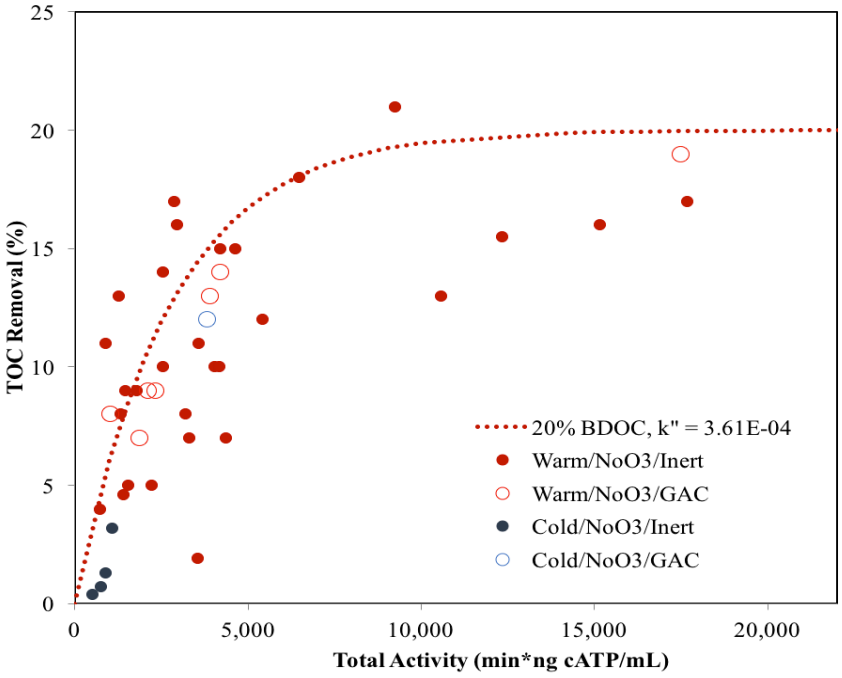

b.

Figure 3.6 TOC removal as a function of total activity using literature data, ozonated (a.) and nonozonated (b.), with temperature correlation factors, media separation and one k" constant (k" = 3.61E-04 mL*ngATP-1*min-1). Measured TOC removal includes the following: warm ozonated waters (n=64), cold ozonated waters (n=14), warm nonozonated waters (n=36) and cold nonozonated waters (5).

For the 119 data points simulated by all four cases and using only one k″ constant, 3.61E-4 $mL*ngATP^{-1}*min^{-1}$, a regression of the measured versus simulated data was carried out, as seen in Appendix B Figure B2. The regression yielded a slope of 0.70, indicating that the model under predicted the removal, and an R^2 value of 0.59, again indicating an acceptable fit for a meta-analysis of literature derived biofilter data. The TOC removal prediction average residuals ranged from 4.1 to 3.5 %. Type of media within the filter did not impact the results, as seen by media separation average residual in Appendix B Table B4. The model proved to be a good fit for multiple variables, while using one k″ value.

Obtaining an ATP biomass activity sample from the top of the filter may not be viable in certain situations. The following equation has been developed from the literature data to predict ATP biomass activity at the top of the filter using influent water quality parameters.

$$ATP_{top}=a*Temp^{b}*TOC_0^{c} \qquad \text{Eq. 3.12}$$

Where, ATP_{top} is the activity at the top of the filter, *Temp* is influent water temperature, and TOC_0 is influent TOC concentration (mg/L). From the literature data and experimental data (n=30), the following coefficient values were calculated: a = 44, b = 0.41 and c=0.99, R2 =0.5. Once ATP biomass activity at the top of the filter was predicted, TOC removal was predicted using the same approach as above, Eq. 3.4 to predict total filter activity and Eq. 3.8 to predict TOC removal. When ATP biomass activity value at the top of the filter was predicted, the TOC removal average residual was 6.29%, while the average residual for TOC removal using the measured biomass activity (and/or concentration) was 3.76% and the average residual for TOC removal using measured biomass and multiple k" constants was 3.69%. Therefore, predicted ATP biomass activity value at the top of the filter compared to measured ATP biomass activity at the top of the filter yielded an increase in average error by ~3%. PL biomass concentration data

converted to ATP biomass activity using Dowdell (2012) conversion, impacted the average residual by a decrease of 0.3%, which did not contribute to the overall results, as seen in Table 3.1. Considering 30% to 20% of the BOM is biodegradable and the variability in each literature data set, the average residuals were not large for the literature data prediction of TOC removal. Measuring ATP biomass activity at the top of the filter is the recommended route, but prediction of ATP at the top of the filter can be an extremely useful tool when acquiring an ATP measurement is not practical or too arduous a task. The relatively small residual further validates the prediction model and allows for significant advancements in modeling TOC removal with biofilters.

Table 3.1 Average residual analysis for TOC removal predictions using Eq. 3.8 for multiple scenarios.

	TOC Prediction with multiple k"s (all data)	TOC Prediction with one k" (all data)	TOC Prediction using one k″ and Predicted Top of Filter ATP biomass activity (all data)	TOC Prediction using one k" (PL biomass concentration only)	TOC Prediction using one k" (ATP biomass activity only)
TOC Percent Removal Average Residual	3.69	3.76	6.29	3.84	3.54
Count	119	119	100	87	32

3.4 Conclusions

This work advances biofiltration model capabilities through the prediction of biofilter performance for TOC removal based on design and operation parameters, which is a useful tool for drinking water utilities when optimizing treatment. The implications of this work suggest biomass distribution decreases exponentially with filter depth and temperature linearly impacts ATP biomass activity, but does not linearly impact PL biomass concentration. A direct

correlation was developed between biomass activity and temperature that allows a robust model, developed by literature data, to be applicable at specific utilities with varying temperatures and EBCTs. Lastly, influent water quality parameters, influent TOC and temperature, can be used to predict ATP biomass activity at the top of the filter.

Chapter 4
Understanding Biofiltration Performance Based on Extended EBCT and Biomass Acclimation and Distribution

4.1 Introduction

Biological filtration (biofiltration) is a treatment technology that can aid drinking water treatment utilities in controlling problematic biodegradable organic matter (BOM), which is found in all source waters (Hozalski et al., 1995; Wang et al., 1995; Carlson & Amy, 1998; Servais et al., 2005) and can lead to bio-regrowth in the distribution system, disinfection byproduct (DBP) formation or be a specific contaminant of concern (Volk et al., 1997; LeChevallier, 2014). Biofiltration occurs when indigenous, heterotrophic bacteria attach to granular media in the filtration process, via elimination of disinfectants in the biofilter influent, to oxidize organic chemicals, as well as nutrients, metal constituents and anthropogenic compounds (Bouwer & Crowe, 1988). Biologically active filters (biofilters) can achieve full removal of certain specific compounds of BOM and partial removal of other fractions of BOM (Zearley & Summers, 2012; Hallé et al., 2015, Carpenter & Helbling (2017), thus leading to drinking water quality with an improved biological stability and reduced toxins and aesthetic contaminants (Stoddart et al., 2016). Numerous factors can impact the effectiveness of biofiltration, i.e., backwashing regime, filter run time, filter media type, time since start-up, nutrient conditions, and peroxidation (Huck et al., 2000). Biofilter efficacy can be improved when design and operational parameters are optimized, i.e., empty bed contact time (EBCT) and the concentration and activity of the biomass throughout the filter (Kaplan et al., 1994; Basu et al., 2016; Selbes et al., 2017).

Slow sand filters (SSF), which are highly effective at BOM removal and widely used in Europe, are no longer commonly used in the U.S. due to their large footprint; however,

conventional filters have a smaller footprint and are required for surface water treatment in the U.S. Rapid media filters can be designed and operated as biofilters through the removal of prechlorination, yet the typical EBCT for a conventional filter in the U.S. is in the range of 3 to 9 minutes, which yields suboptimal performance for biofilters, especially under cold conditions. Residence times of biofilters vary from hours for a SSF to minutes for a rapid-rate filter, depending on the type of filter (Graham & Collins, 1996). There is a knowledge gap between BOM removal of SSFs and extended EBCT biofilters, yet some researchers have indicated that longer EBCT for biofilters will lead to enhanced BOM removal similar to SSF due to longer exposure time for degradation of slower degrading compounds (LeChevallier et al., 1992; Zearley & Summers, 2012). There is a potential for extended EBCT biofilters (EBCT = 30 minutes) as many water treatment utilities have additional capacity in their filter operations. Utilities operate at partial filter utilization by keeping some filters offline, but an alternative approach is to utilize all of the filters at a lower hydraulic loading rate (HLR), which would yield longer EBCTs.

Understanding when this extended EBCT approach is advantageous requires an understanding of the distribution of biomass throughout biofilters and relating it to biofilter performance, as biomass concentration profile is an important tool to enhance BOM removal by optimizing filter design and operations (Carlson & Amy, 1998). Biofilter biomass is often quantified via concentration, phospholipid analysis, or activity, adenosine triphosphate (ATP) analysis, as reviewed by Pharand et al. (2014) and in Chapter 1. Total microbial activity in biofilters is important for dissolved organic matter (DOM) removal efficacy, as first purposed by Wang and Summers (1993), and may be influenced by various water quality parameters and filter operations, such as influent BOM, temperature, and backwashing conditions. Acclimation

time for a conventional filter converted to a biofilter is not well understood in terms of biomass activity acclimation and biofilter performance at the pilot scale under seasonal variations.

The objectives of this work were to 1) evaluate biomass acclimation behavior from a conventional filter to a biofilter, 2) determine source water seasonal variations and extended EBCT effects on biofilter performance, 3) develop biomass filter depth profiles and 4) apply the models developed in Chapter 3 to predict DOC removal with pilot data that is susceptible to uncontrollable variables, such as the seasonal water quality variations.

4.2 Material and Methods

4.2.1 Biofilter Design and Operation

Pilot filters were set up at the City of Boulder's (CO) Betasso Water Treatment Plant. The experiments were operated from April 2015 to August 2015 during spring runoff (i.e., snowmelt) to capture surges in TOC and temperature. The pilot treatment train operated at a flow rate of 2 gallons per minute and consisted of rapid mix, three stage tapered flocculation, sedimentation, and filtration, as seen in Appendix C Figure C1. Source water, a combination of Barker Reservoir and Lakewood Reservoir (water quality data for duration of study seen in Table 4.1), was sent to the raw water tank, from which water was pumped to the treatment train. The experiments in this study were run as direct filtration; therefore, no chemicals were added prior to biofiltration. Biofilters were backwashed weekly to 30% bed expansion for ten minutes with finished plant effluent from the clearwell (i.e., chlorinated backwash). Chlorinated backwash was not preferred but employed solely due to existing pilot plant schematic. The chlorine residual never exceeded 1.1 mg/L.

Table 4.1 Barker and Lakewood Reservoir Combined Water Quality

DOC (mg/L)	UVA_{254} (cm^{-1})	SUVA (L/mg-C/m)	Alkalinity (mg-CaCO3/L)	Temperature (°C)	pH
2.4-7.0	0.07-0.23	2.86-3.49	18.5	5-18	7.3-7.6

DOC = dissolved organic carbon, UVA = ultraviolet absorbance, SUVA = specific ultraviolet absorbance

The original pilot system was modified and two new filter columns were fabricated with depth taps to achieve filter depth samples, as seen in Appendix C Figure C2. The placement of the sampling ports facilitated direct organic removal measurements associated with the biofilter and not the feed system. Filter 1 was packed with nonacclimated anthracite and Filter 2 was packed with bioacclimated anthracite from Longmont (CO) Nelson Flanders Water Treatment Plant, to compare the development and performance of the nonacclimated anthracite to the bioacclimated anthracite. The anthracite had an effective size of 1.0 mm and was uniformly placed into pilot scale clear PVC columns with 3-inch (76 mm) inner diameter to a depth of 3.30 feet (100 cm). The sample taps were located at the top of the media (the first 3 in (5 cm) and 0.55ft (17 cm), 1.66 ft (50 cm), and 3.30 ft (100 cm) below the top to represent EBCTs of 5, 15 and 30 minutes, calculated as the ratio of the depth to the hydraulic loading rate or column cross-sectional area to volumetric flow rate. The flow rate was measured using in-line flow meters after each filter and averaged at 0.04 gpm, which represents a HLR of 2 m/hr. The filters were online during winter, spring and summer months and captured surges in DOC during spring runoff (3 mg/L to 7 mg/L to 2 mg/L), with temperature fluctuations from 5 to 18 °C.

4.2.2 Biomass Activity

Media samples were extracted weekly from each column sample tap prior to backwashing in order to measure biomass activity throughout the column. Samples were taken at the top, 5 min, 15 min and 30 min sample ports and analyzed after liquid samples had been

collected and the filters drained. Activity was measured on-site, directly after each sampling period, which eliminated temperature changes in the media after extraction. A sterilized spatula was used to remove media from the sample port and immediately placed in a sterile conical tube. To measure ATP, approximately one gram of media was weighed into a test tube and extracted using the LuminUltra Deposit and Surface Analysis Test (DSA-100C, Fredericton, NB) following the manufacturer's instructions. The dry: wet ratio for the media and the bed density were determined to normalize all results to biomass per mL of dry media. Results were then converted to ngATP/cm^3 media using a dry:wet ratio of 57% and an anthracite bed density of 0.8 g dry weight/cm^3.

4.2.3 Water Quality Analysis

DOC samples were taken in duplicate at the influent (directly after sedimentation basin), 5 min, 15 min and 30 min EBCT sample ports. Samples were immediately filtered at the Betasso Water Treatment Plant through a 0.45 μm membrane filter (Pall Life Sciences). After filtration, the samples were transported to CU-Boulder and stored at 4°C until DOC analysis. Then samples were analyzed on a Sievers 5310 C Laboratory Organic Carbon Analyzer using the ultraviolet irradiation/persulfate oxidation method (SM 5310C). Biodegradable dissolved organic carbon (BDOC) was measured using the Mogren et al. (1990) method. A recirculating column with indigenous, acclimated bacteria attached to media was employed to measure the change in DOC over a period of 5 days at 20 °C. BDOC was measured as BDOC = DOC_{intial}-DOC_{final}. UVA_{254} samples were taken out of the same liquid sample as the DOC measurements, therefore, having identical sampling time and frequency. UVA_{254} was measured using a HACH DR/4000 Spectrophotometer in the CU-Boulder lab. Samples were analyzed using a 1-cm quartz cell and absorbance values were reported with units of cm^{-1} as spectral absorbance coefficients. Turbidity was measured daily on filter influent and effluent using online turbidity meters. Duplicate grab

samples for turbidity measurements were taken and analyzed at Betasso Water Treatment Facility in accordance with Method 2130 B (Standard Methods, 1998) using a HACH 2100N Turbidimeter on biofilter sample days. Turbidity measurements were reported as nephlometric turbidity units (NTU) and measured on unfiltered water. Filter influent was measured daily by an online pH meter and grab samples were taken as a duplicate and measured using a pH probe at Betasso Water Treatment Facility. Influent alkalinity samples were taken biweekly and measured at Betasso Water Treatment Facility using a titration apparatus with 0.020N Sulfuric Acid Standard Solution.

4.3 Results and Discussion

4.3.1 Biomass Acclimation and Behavior

Carlson and Amy (1998) reported that biofilter DOC removal was a function of biomass concentration or BOM formation, therefore, biomass acclimation was significant. Basu et al. (2015) reviewed acclimation periods ranged from 20 days to more than 16 months for biofilters under a wide range of source waters. For this study, the development of biomass in the nonacclimated filter began immediately upon startup contingent upon the lack of disinfectant residual in the influent of the filter. The acclimated biofilter media had been in full-scale biological operation for several years and the biomass was well established; however, seasonal changes in the source water increased the biomass in the acclimated filter over the course of this experiment. The comparison of biomass development for a filter with nonacclimated anthracite relative to a filter with acclimated anthracite at four depths is shown in Figure 4.1.

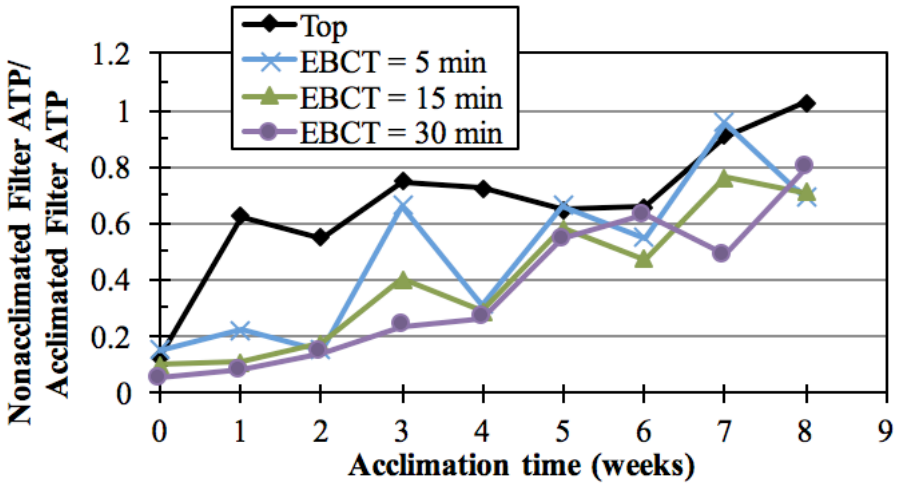

Figure 4.1 Biomass development (ATP, adenosine triphosphate) on an nonacclimated anthracite filter normalized to that of an acclimated anthracite filter for four sample depths, representing empty bed contact times (EBCTs) of 5, 15 and 30 minutes.

The top of the nonacclimated filter developed biomass activity relatively quickly, within the first week, and after week three, the biomass at the top of the nonacclimated filter was consistently 70% of the top of the acclimated filter. After week seven, the top of the nonacclimated filter had the same biomass activity as the acclimated filter. Biomass development was slower at the other depths and required 8 weeks to achieve similar activity levels (80%) to the acclimated filter at all depths. After 17 weeks of operation, the total biomass activity throughout the nonacclimated filter was 80% of the acclimated filter. Wang and Summers (1993) showed a similar trend whereas biomass in the bioacclimated system was twice that of the non-bioacclimated system after 77 days (Wang & Summers, 1993). Stoddart et al. (2016) also found a rapid initial biomass accumulation at the top of the biofilter once prechlorination was removed. Velten et al. (2011) observed a 90-day time frame for unacclimated GAC to reach a steady-state biomass concentration throughout all levels of the filter.

The average ATP results for the nonacclimated and acclimated filters, respectively, were 423 and 463 (ng ATP/cm^3 media) at the top of the biofilter over the 17-week operation period (Table 4.2). The results were in the range of the Pharand et al. (2014) survey of 16 published

biofilter studies reporting ATP concentrations of 10^2 to 10^3 ng ATP/cm^3 media. The filter bed depth samples showed activity decreased exponentially throughout the filter due to consumption of primary substrate, which was consistent with other literature studies (Wang et al., 1995; Carlson & Amy, 1998; Rahman, 2013). Biomass concentration, phospholipid analysis, have also been shown to decrease with biofilter bed depth (Hallé et al., 2009; Emelko et al., 2006; Wang et al., 1995; Urfer & Huck, 2001). Likewise, this trend was observed when protein concentration was used as a surrogate for biomass concentration (Carpenter & Helbing, 2017).

Table 4.2 Average biomass activity (ngATP/ cm^3 media) for the nonacclimated filter and the acclimated filter for the duration of the study.

	Average Activity (ng ATP/cm^3 media)			
Sample Point	Top	EBCT 5 min	EBCT 15 min	EBCT 30 min
Nonacclimated Filter	423	113	69	49
Acclimated Filter	463	154	116	97

EBCT-empty bed contact time

The overall biomass activity in each filter increased over the course of the experiment, as the temperature increased from 5 to 18°C and the influent DOC varied between 2 mg/L and 7 mg/L, both of which impacted biomass activity. A significant correlation was found between temperature and activity at the top of the filter (p value= 0.016), as well as influent DOC and top of filter activity (p value= 0.015). Other researches also observed a correlation with ATP and influent DOC (Pharand et al., 2014) and ATP with temperature (Hallé et al., 2015).

Preoxidation, variation in seasonal DOC, or increased HLR can impact the substrate (DOC) loading. The impact of DOC on biomass has been debated in the literature. An increased HLR, the rate of mass flow per unit area (flux), is one way to increase DOC onto the filter. Carlson and Amy (1998) concluded an increase in flux of substrate onto the biomass allowed substrate to infiltrate deeper into the filter bed, which positively impacted biomass growth. Carpenter and Helbling (2017) found a higher flux of substrate was positively associated with

total biomass within the filter and depth of the biological zone, yet the biofilter with the lower flux had more biomass at the surface of the filter. However, other researchers found a higher flux led to biomass shear and sloughing, which had a negative effect on the biomass (Horn et al., 2003). Wang et al. (1995) found biomass decreased by 50% in three filters when preozonation was turned off, suggesting BDOC loading affected biomass growth. Increased influent DOC without changing hydraulics, i.e. snowmelt & seasonal variations, was examined in this experiment and was found to impact biomass activity significantly (p value= 0.015). Biomass oxidize DOC as the primary substrate for regrowth and energy, so it is intuitive the positive correlation between influent DOC and biomass activity, although a threshold may exist.

Temperature proved to be highly correlated with total biomass activity, as temperature affects microbial growth and decay rates (Urfer et al., 1997). Temperature impacts on total biomass activity can be seen Figure 4.2. For both filters starting at 5 °C, there was an upward, linear trend between temperature and total activity; however, the relationship plateaued around 15° C. The R^2 value for the acclimated and nonacclimated filter (up to 15°C) was 0.85 and 0.77, indicating a good correlation between temperature and total biomass activity for water temperatures less than 15 °C. Interestingly, Stoddart et al. (2017) found media enhancement (GAC over anthracite/sand) on DOC removal was statistically significant up to 15°C, and >15°C the effect was no longer statistically significant, albeit still positive.

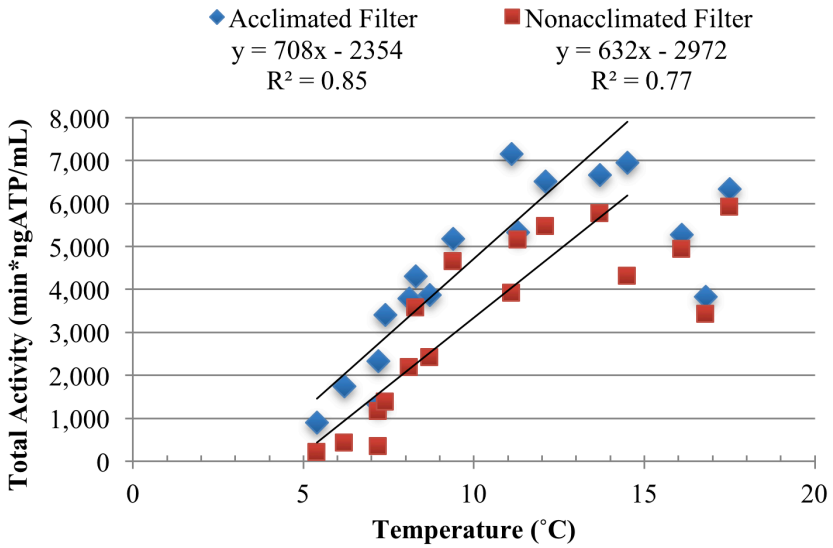

Figure 4.2 Effect of temperature on activity over a 17-week operation period. Trendlines associated with total activity and temperature up to 15 °C.

4.3.2 Organic Carbon Removal

The influent DOC concentrations ranged from 2.4 mg/L to 7.0 mg/L and the BDOC of the source water was 20%. The source water BDOC was similar to other source water BDOC values as the average BDOC percentage for 100 different nonozonated waters was 23%, with a median of 20% (Chapter 1). Throughout the study, BDOC removal increased with filter depth, i.e., 30 min EBCT sample removed more BDOC compared to the 5 min EBCT sample in both the acclimated and nonacclimated filter. The average BDOC removal for the 5, 15 and 30 min EBCTs for the nonacclimated filter was 14%, 23% and 33%, respectively, and for the acclimated filter was 16%, 23% and 50%, respectively. The average DOC removal for the 5, 15 and 30 min EBCTs for the nonacclimated filter was 2%, 4% and 7%, respectively, and for the acclimated filter was 3%, 5% and 10%, respectively. DOC removals from this study, Figure 4.3, fall within the range of the literature review from Chapter 1, which reported median removals of 7% TOC at ≤10 °C, 10% TOC for 10-20 °C, and 15% TOC for ≥20 °C for nonozonated waters at an average EBCT of 14 minutes.

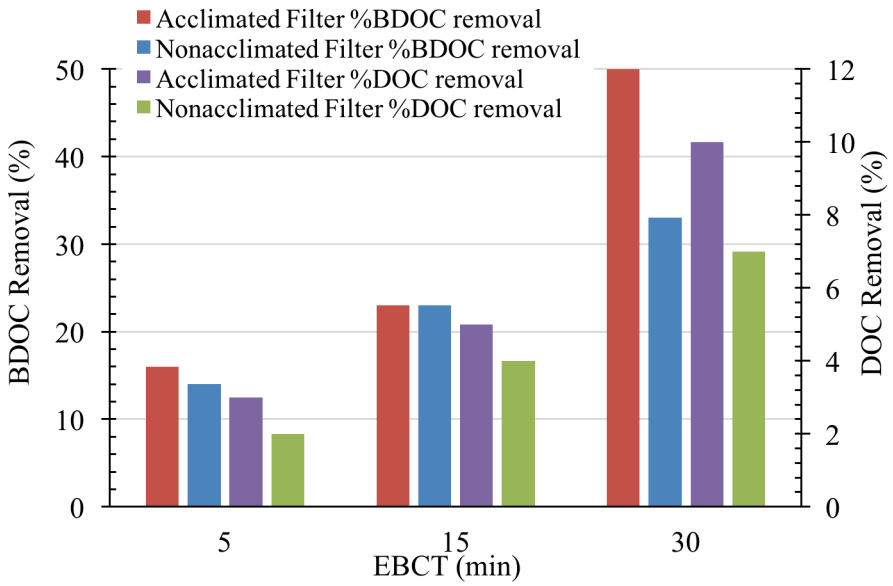

Figure 4.3 Average BDOC and DOC removal with respect to EBCT in the nonacclimated and acclimated filter.

4.3.3 Impact of Extended EBCT

EBCT has a strong correlation with DOC removal (Urfer et al., 1997; Terry & Summers, 2017). It has been shown that external mass transfer in the hydraulic loading rate range of 1.5 to 15 m/hr (0.6 to 6 gpm/ft^2) does not limit the removal of DOC, rather utilization of the substrate at the media surface limits DOC biodegradation (Wang & Summers, 1994; Carlson & Amy, 1998). Therefore, extended EBCTs allow for longer contact time and can increase DOC biodegradation. Servais et al. (1992) found increased EBCT increased BDOC removal within the HLR of 6-18 m/hr, but found EBCT needed to be doubled to maintain the same BDOC efficacy when the source water temperature dropped 12°C. LeChevallier et al. (1992) found TOC removal increased from 29% at 5 min EBCT to 33% at 10 min EBCT to 42% at 15 min EBCT to 51% at 30 min EBCT. Ko et al. (2007) ran three filters in parallel with preozonation at three different HLRs and found the longest EBCT to be the most efficient with DOC removal, i.e. 19% removal

at 5 min, 26% removal at 10 min, and 31% removal at 20 minutes. Similarly, Chen et al. (2016) ran two pilot filters in series with depth ports and found increased DOC removals with increased filter depth: 3% DOC removal at 2 min EBCT, 5% DOC removal at 4 min EBCT, 7.5% DOC removal at 12 min EBCT, 10% DOC removal at 16 min EBCT and 15% DOC removal at 23 min EBCT. In contrast, Hozalski et al. (1995) found no difference in TOC removal in the range of 4 to 20 minute EBCT, but attributed this to the rapid TOC biodegradation as a result of preozonation and warm temperatures.

In this study, EBCT was extended from 5 to 30 minutes without preoxidation and was evaluated over seasonal variations to determine the impact extended EBCT had on DOC removal. Extending the EBCT from 5 to 30 minutes increased DOC removal by 7% and BDOC removal by 34% in the acclimated filter, and increased DOC removal by 5% and BDOC removal by 19% in the nonacclimated filter, as seen in Figure 4.3. The results suggest an extended EBCT is desirable to enhance BOM removal for biofilters by allowing additional BOM removal to be achieved from longer exposure time.

4.3.4 Impact of Temperature

Previous research suggest temperature impacts biofiltration performance (Selbes et al., 2016; Hallé et al., 2015; Basu et al., 2015; Moll et al., 1999). This study found as influent water temperature increased from 5° C to 18° C, BDOC removal increased. As seen in Figure 4.4, for the acclimated filter, at temperatures higher than 10° C, BDOC removal averaged 62%; however, at temperatures lower than 10° C, the acclimated filter BDOC removal averaged 47%. Hozalski et al. (1999) found that decreased operating temperatures impaired BOM removal and lengthen biofilter acclimation time. Servais et al. (1992) found BDOC removal strongly decreased with decreasing temperature, with an average BDOC removal of 60% at 20°C, and an average 27%

BDOC removal at 8°C. The Basu et al. (2015) review concluded that the majority of published literature found lower operating temperatures decreased organic removals due to a decreased microbial activity within the filter and thus lower biodegradation of organics.

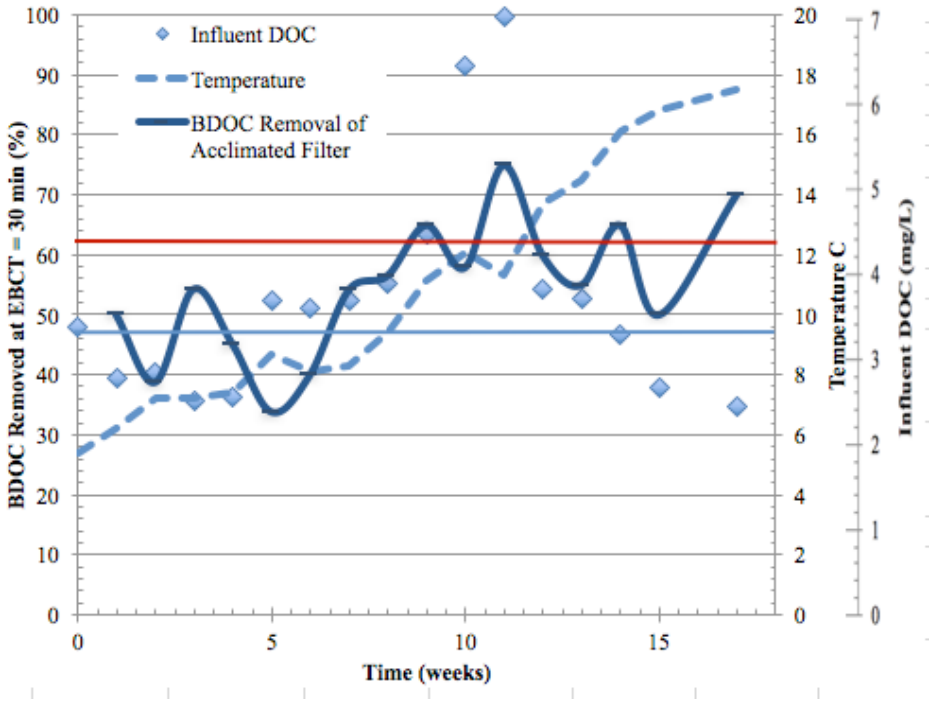

Figure 4.4 Impact of temperature and influent DOC on filter performance as measured by BDOC removal for the acclimated filter. Above 10 °C, average BDOC removal was 62% (red horizontal line) and below 10 °C, average BDOC removal was 47% (blue horizontal line).

4.3.5 Impact of Influent DOC Concentration

With time, the influent DOC concentration increased from 3 mg/L to 7 mg/L then decreased to 2 mg/L in the acclimated filter. However, BDOC removal, expressed as a percent, did not follow the same trend as influent DOC. BDOC removal gradually increased over time despite influent DOC decreasing after spring runoff. Thus, influent DOC did not have a direct correlation with BDOC removal throughout the filter, as seen in Figure 4.4. This trend seems accurate under the notion that external mass transfer does not limit DOC removal. Biomass is a

function of influent DOC, but DOC removal does not seem to be a direct function of influent DOC, once a steady-state biomass has been achieved.

4.3.6 Biomass Profile Model

It has been widely studied that biomass concentrations are highest at the top of the biofilter and exponentially decay throughout the filter as reviewed by Pharand et al. (2014) and in Chapter 3. The normalized concentration of literature data, the ratio of ATP or phospholipid biomass at a given EBCT to that at the top produced a correlation found in Chapter 3.

$$X(EBCT)/X_{top}=a*\ln(EBCT)+b \quad \text{Eq. 4.1}$$

The equations and coefficients from this study were similar to the derived equation from the literature data, further validating the model. Chapter 3 biomass distribution model for nonozonated waters (Eq. 4.1) reported an average *a* coefficient of -0.12 and an average *b* coefficient of 0.72. The pilot filters activity data show a similar correlation with an average *a* coefficient of -0.10 and 0.12 and an average *b* coefficient 0.53 and 0.46, respectively for the acclimated and nonacclimated filters, seen in Table 4.3. Biomass distribution of the acclimated and nonacclimated filter over the duration of the study normalized to the top of the filter was similar to the distribution seen in the 26 biofilter studies in Chapter 3 (Table 4.3). For the literature model, acclimated filter and nonacclimated filter, ATP normalized to the ATP at the top of filter was 0.47, 0.40, 0.30 at 5 minutes EBCT, 0.33, 0.30, 0.15 at 15 minutes EBCT, and 0.25, 0.20, 0.10 at 30 minute EBCT, respectively. In all biofilters, biomass decreased with increased EBCT.

Table 4.3 Biomass activity distribution for acclimated and nonacclimated filter compared to Chapter 3 biomass distribution model.

Biomass(EBCT)/Biomass at top = a*ln(EBCT)+b					ATP/ATP_{top}		
Study	**a**	**b**	**r^2**	**n**	**5**	**15**	**30**

Literature Model	-0.12	0.72	0.67	117	0.47	0.33	0.25
Acclimated Filter	-0.10	0.53	0.89	68	0.40	0.30	0.20
Nonacclimated Filter	-0.12	0.46	0.95	68	0.30	0.15	0.10

EBCT- empty bed contact time; ATP – adenosine triphosphate

Previous researchers have indicated biomass concentration controlled DOC removal during biofiltration (Carlson & Amy, 1998), but the removal of DOC was not linearly correlated to the amount of attached biomass (Urfer et al., 1997). Wang and Summers (1993) developed a correlation between DOC removal and the product of EBCT and biomass, total biomass. The concept of total biomass has also been used by other researchers (Zearley, 2012; Carpenter & Helbling, 2017). Chapter 3 developed Equation 4.2 from literature data to determine the total biomass in a filter, X_{total}, to a given depth or *EBCT*, using the same a and b coefficients as in Table 4.3, to better predict DOC removal within the biofilter.

$$X_{Total} = X_{Top}(a*(EBCT*\ln(EBCT)-EBCT)+b*EBCT) = X_{avg}*EBCT \quad \text{Eq. 4.2}$$

The authors then used second order rate equations, developed in Chapter 3, to model DOC removal using total biomass, Eq. 4.2. The contaminant remaining (C_{eff}/C_{inf}) is equal to the biodegradable fraction of the organic matter (BOM_{frac}) times the exponential function of a rate constant (k'') times total biomass ($X_{avg}*EBCT$). For full model development or other defined parameters refer to Chapter 3.

$$C_{eff}/C_{inf} = BOM_{frac}\ (\exp\ (-k''*X_{avg}*EBCT)) \quad \text{Eq. 4.3}$$

Integrating total activity (Eq. 4.2) with the organic removal prediction equation (Eq. 4.3) allowed for prediction of DOC removal as shown in Figure 4.5. ATP is a function of temperature, thus incorporating total ATP biomass activity in the predictive model incorporated temperature into the model. Therefore, one k" constant is required for any given temperature, with known BDOC

values. The model predicted DOC removal when EBCT (min) and ATP_{top} (ATP ng/cm^3 media) were known and a k″ constant of 3.61E-4 [cm^3*(ngATP*min)$^{-1}$], the average k" from the literature data (n=119) was used for both BDOC values. For this study, the model over predicted DOC removal, which could have been a result of chlorinated backwash. The chlorinated backwash inhibited biomass at the bottom of the filter more than biomass at the top of the filter. Biomass at the top, which was used to predict total activity, was more resilient to the chlorinated backwash due to higher levels of primary substrate and a more established biomass community. Wang et al. (1995) compared an anthracite filter backwashed with chlorine and an anthracite filter backwashed without chlorine and found the chlorinated water impaired the biomass growth as higher biomass concentrations were found in the filter backwashed without chlorine. In addition, Miltner et al. (1995) found a dual media filter backwashed with 1 mg/L chlorine maintained lower concentrations of biomass and achieved lower steady-state removals of the same contaminants than a dual media filter backwashed without chlorine. Therefore, the model is best applied when a chlorinated backwash is not used, as chlorinated backwash impairs biomass concentration/activity and organics removal.

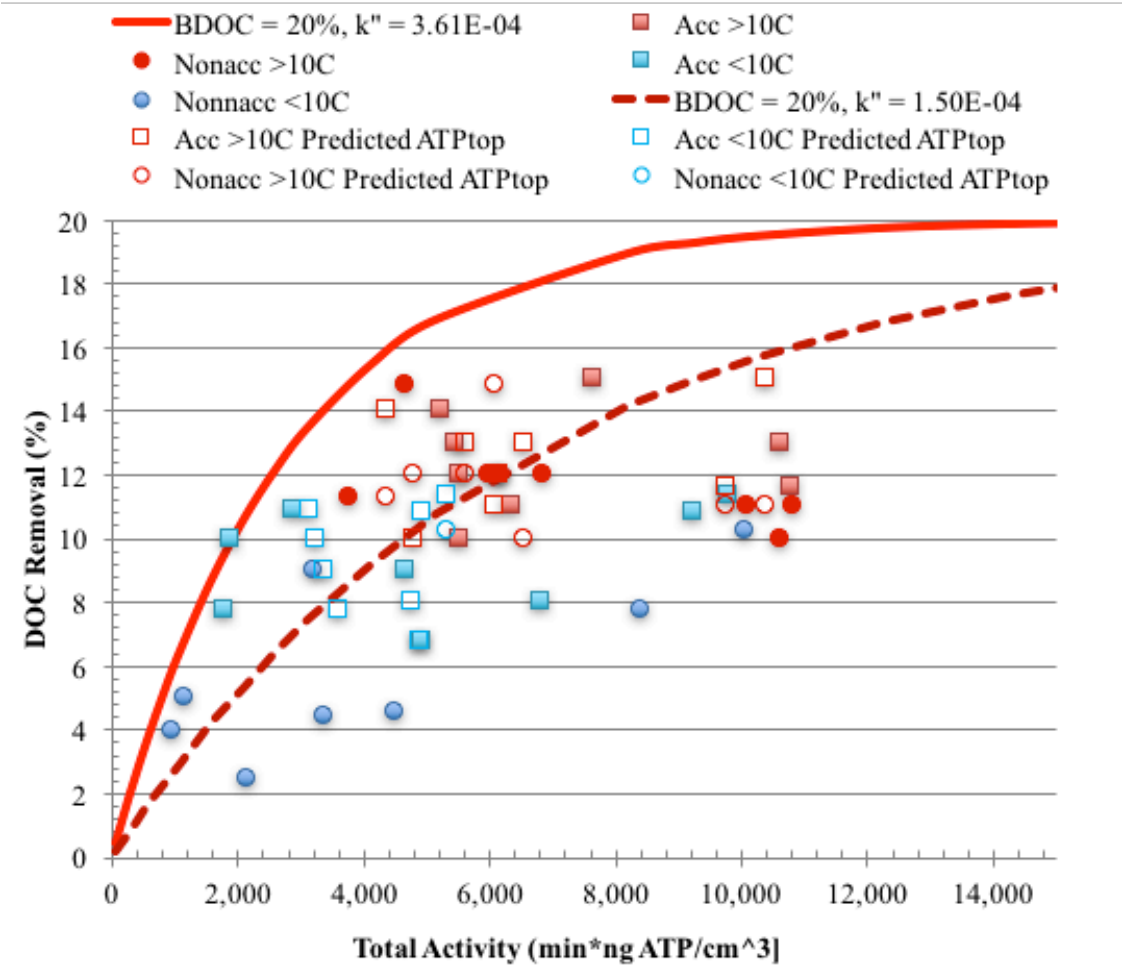

Figure 4.5 Measured and modeled DOC removal on an nonacclimated (nonacc) and acclimated (acc) filter as a function of total biomass activity for cold waters (n=17) and warm waters (n=23) for measured ATP at the top of the filter (solid symbols) and predicted ATP at the top of the filter (hollow symbols).

Total activity plotted against DOC removal for this study showed a better fit to the model using a lower k″ constant of 1.5E-4 [cm^3*(ngATP*min)$^{-1}$], Figure 4.5. The cold and warm data points fit the model well with TOC removal average residual of 2.9% for the acclimated filter and 3.3% for the nonacclimated filter.

Influent water quality parameters can be used to predict ATP at the top of the filter, when obtaining a biomass sample at the top of the filter is unfeasible. In Chapter 3 the following

equation from literature data (n=119) was developed to predict ATP at the top of the filter if temperature and influent TOC are known parameters.

$$ATP_{top}=a*Temp^{b}*TOC_0^{c} \quad \text{Eq. 4.4}$$

In the above equation, the coefficient values were as follows: a= 44, b = 0.41 and c=0.99. Once ATP at the top of the filter was predicted, the same process of finding total activity to predict TOC removal was followed. The data from this pilot study was applied to the prediction model to test its validity. The cold and warm data points fit the model well with TOC removal average residual of 1.6% (n=16) for the acclimated filter and 2.14% (n=9) for the nonacclimated filter, albeit Equation 4.4 was only applied after week 7 for the nonacclimated filter to ensure a healthy operating biomass. The results fit the model well and strengthened the concept of predicting ATP based off of influent water quality parameters. The authors recommend measuring activity at the top of the filter, but in special cases where a sample is not attainable, this model can be applied. The ability to predict ATP based off of influent water quality parameters significantly advances the field of biofilter performance modeling.

4.4 Conclusions

Longer EBCTs increased removal of the BDOC. The acclimated filter exhibited the following performance: at 5, 15 and 30 minutes of EBCT, 16%, 23% and 50% BDOC removal were measured. Extended EBCT biofilters have the potential to bridge the gap of BDOC removal between slow sand filters and conventional rapid media filters. Acclimation of the filters in terms of BDOC removal and activity occurred over a two-month time frame (8 weeks). Activity development on the nonacclimated filter relative to that of the acclimated filter occurred over three weeks for the top of the filter and over eight weeks for the depth of the filter (80% activity). Similarly, BDOC removal was only 10% different at week eight at the 30 minute EBCT. After

17 weeks of operation, the nonacclimated filter contained 80% of the activity the acclimated filter contained after 5 minutes of EBCT. Influent DOC concentration did not impact BDOC removal directly. When the influent DOC concentration increased from 3 mg/L to 7 mg/L and back down to 2 mg/L, BDOC removal at 30 minute EBCT did not follow the same trend. Whereas, temperature and depth of the filter did impact BDOC removal. At temperatures higher than 10° C, BDOC removal averaged 66%; however, at temperatures lower than 10° C, BDOC removal averaged 48%. Activity was a direct function of temperature ($p=0.016$) and influent DOC ($p=0.015$). A strong relationship between temperature and activity was found in the range of 5-15° C, then the relationship plateaued off at greater than 15° C. The pilot data fit the predictive TOC model (Chapter 3) well and improved the strength of the model due to low TOC removal residuals. Influent TOC and influent temperature can be used to predict activity at the top of the filter when a biomass sample is not feasible.

Appendix C

Appendix C is available at the end of this document. It includes a more detailed description of the schematic, turbidity data, coagulation data, EPS data, and stop-start operation effects.

Chapter 5
Biofiltration Performance: Evaluating and Modeling Effects of Extended Empty Bed Contact Time, Temperature, and Dissolved Organic Matter Character

5.1 Introduction

Understanding the removal of organic constituents, both specific and non-specific natural organic matter (NOM) removal is essential for the production of safe drinking water. NOM removal in drinking water utilities is important as it can serve as an organic disinfection byproduct (DBP) precursor, diminish the biostability of the finished water, or be a specific contaminant (Moll et al., 1998; Jarusutthirak et al., 2002; Lauderdale et al., 2012; Hozalski et al., 1999; Liu et al., 2017). A fraction of the NOM is biodegradable, and averages 20% biodegradable dissolved organic carbon (BDOC) in nonozonated waters and 30% BDOC in ozonated waters, as shown in Chapter 1.

One of the primary factors affecting the rate and extent of NOM biodegradation is its composition. NOM occurs in all water sources and is produced by the breakdown of plant (terrestrial, allochthonous) and animal (microbial, autochthonous) material. Effluent Organic Matter (EfOM) is another category of NOM and is the discharge from wastewater treatment plants and can be divided into three groups based on its origin: 1) recalcitrant NOM derived from drinking water sources, 2) synthetic organic compounds added by consumers and DBPs generated during disinfection of wastewater and water treatment, and 3) soluble microbial products derived during biological processes of wastewater treatment due to decomposition of organic compounds (Drewes et al., 1999; Jarusutthirak et al., 2002). Despite having a variety of chemical structures and biodegradabilities, drinking water utilities are faced with the task of removing NOM from the drinking water.

Biological filtration (biofiltration) is a treatment technology that can remove NOM from the source water. The biomass in the biofilters grows and maintains itself on the BDOC. With biofiltration, water is passed through a media bed (e.g. sand, anthracite, GAC) where it is treated by a combination of physical and biological processes. Most granular media filters can be converted into biological filters by not carrying an oxidant residual through the filters, thus allowing the naturally occurring microorganisms in the source water to attach to the media surface and develop a biofilm (Hozalski et al., 1999; Lee, 2014).

Specific ultraviolet absorbance (SUVA), the ratio of UV-to-TOC, is the UV absorbance of a water sample at 254nm normalized by dissolved organic carbon (DOC), which provides a measure of aromaticity of NOM (Weishaar et al., 2003). Higher SUVA indicates more aromatic structures and unsaturated carbon bonds, while lower SUVA indicates more aliphatic structures and saturated carbon bonds (Hozalski et al., 1999). Edzwald and Tobianson (1999) characterized SUVA the following: >4 L/mg-C/m composed of mostly aquatic humics, high hydrophobicity, and high molecular weight; 2 – 4 L/mg-C/m composed of a mixture of aquatic humics and other NOM, mixture of hydrophobicity and hydrophilic NOM, and mixture of molecular weights; <2 L/mg-C/m composed mostly of non-humics, low hydrophobicity and low molecular weight. Low SUVA waters (less than 4 L/mg-C/m) are thought to be more susceptible to higher biodegradability than waters with high SUVA (Volk & LeChevallier, 2002).

Parameters such as empty bed contact time (EBCT), temperature, and biomass impact the NOM removal efficacy of biofilters. EBCT is a measure of how long water is in contact with the biofilter media, and the longer exposure contaminants have in the filter, the more degradation and removal occurs (Juhna & Melin, 2006). True filter residence time is about 50% of EBCT as the media takes up about 50% of the volume. Source water temperatures can vary and Basu et al.

(2015) review demonstrated that lower operating temperatures decreased biological filtration performance of organics biodegradation. In addition, biomass concentrations can limit NOM removal if the biofilters are not operating at steady-state and steady-state can take from 20 days to 16 months to be reached (Basu et al., 2015). Biomass concentrations, phospholipids or activity, have been shown to decrease with increasing filter depth concomitantly with primary substrate utilization (Wang et al., 1995; Moll et al., 1998). Liu et al. (2017) reviewed the importance of operating parameters, filter media, EBCT, backwashing, temperature, nutrient augmentation, and pre-oxidation on biofiltration performance as measured by removal of regulated DBPs and emerging DBPs.

Modeling biofilter performance is advantageous for the ability to predict organics removal throughout the biofilter and optimize overall treatment performance. The model from Chapter 3 is used in this study to predict TOC removal based on total activity, the total biomass throughout the filter depth (EBCT). This study will not go into the details of the model, but the development can be found in Chapter 3.

The overall objectives were to evaluate and model the effects of extended EBCT, temperature, influent DOM, biomass and source water type have on biofilter NOM removal performance.

5.2 Material and Methods

5.2.1 Biofilter design and operation

Three biofilters were run in series with a flow rate of 7.2 mL/min and a target hydraulic loading rate of 2.4 m/hr (1.0 gal/ft2·min). Acclimated anthracite media with an average size of 1.0 mm was packed into 15 mm inner diameter glass chromatography columns (ACE Glass 5820-12). The aspect ratio was less than 15, in order to minimize short-circuiting. The anthracite media was provided by the Nelson Flanders Water Treatment Plan (Longmont, CO), which had

been used in the pilot studies described in Chapter 3. Each filter contained a layer of support media (2 mm glass beads) below the filter media. The support media was not included in the calculation of the empty bed contact time (EBCT). The columns were run using peristaltic pumps (Masterflex Models 7553-30 and 7518-10), and a needle valve after each column was used to control flow, as seen in Figure 5.1. Sampling ports were located immediately before and after each column to assess the removal associated directly with the biofilter. The biofilter columns were covered to prevent photosynthesizing microorganisms from growing in the filter.

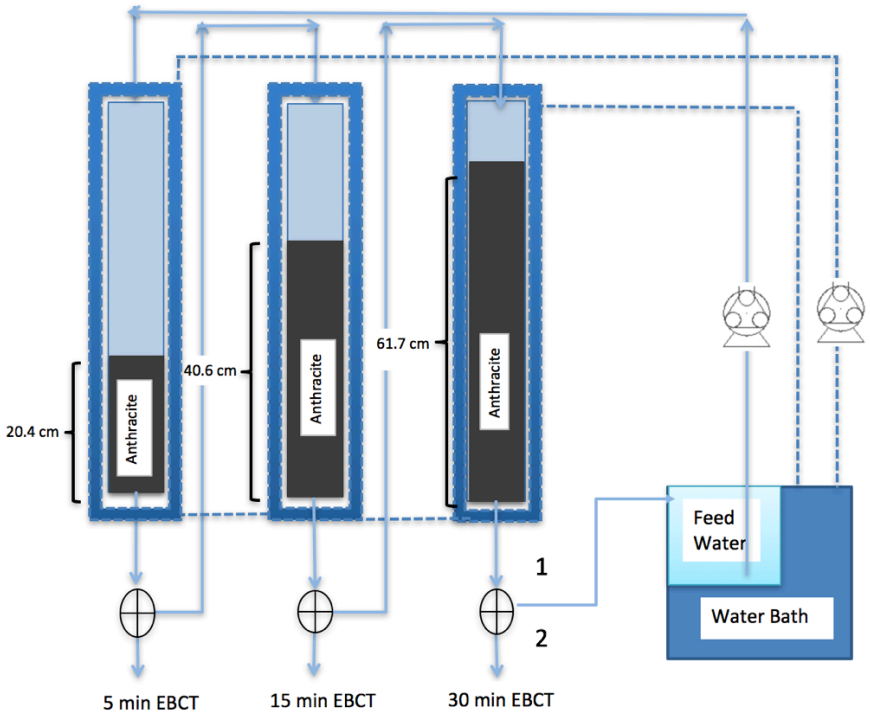

Figure 5.1 Biofilter apparatus bench scale biofilters, recirculation (valve 1 open) vs. single pass mode (valve 2 open).

The biofilters in normal operation were operated at lab temperature (20 ± 2 ° C). Experiments were ran at varying temperatures to determine temperature effects on biofilter performance. In order to run experiments at cold temperatures, biofilters were set up in the walk-in refrigerator with heating and cooling capacity (Bally Refrigerator Boxes, Inc. Model 3678) set for either 6 °C or 28 °C. In other scenarios, temperatures were controlled by a water chiller with

capacity for cooling and heating. The feed water was directly inserted into the chiller, acting as a temperature sink, and the biofilters were wrapped with temperature controlled tubing.

The batch/single pass method developed in Chapter 2 was employed for biofilter operation. In order to emulate a flow-through pilot scale experiment at the bench scale, biofilters were operated in recirculating batch mode (adapted from Joret & Levi, 1986; Mogren et al., 1990; Frias et al., 1992) followed by flow through single pass mode, then sampling. In order for microorganisms to adjust to each new influent condition, the biofilters were acclimated for 5 days through batch recirculation. The recirculation period reduced the total amount of source water needed for each experiment. Single pass mode was employed to simulate pilot scale flow through in order to allow for ample primary substrate to be consumed prior to sampling. Lastly, sampling occurred as would during a pilot flow through experiment, with samples taken directly after each biofilter column. In order to execute this method, the feed water was split into two batches. The first half of the feed water was recirculated to acclimate the media to the specific water matrix for a period of 5 days and the second half was stored at 4 °C until use. Following the acclimation period, the second half of the feed water was run in a single pass mode for 4 hours, then sampling occurred. Referring to Figure 5.1 showing the biofilter apparatus, valve 1 was open for recirculation and valve 2 was open for single pass.

5.2.2 Feedwaters

Three feed waters were used for this study to represent a terrestrial, microbial and wastewater effluent source. Barker Reservoir, located in Netherland, Colorado, served as the terrestrial source and receives snowmelt input yearly. Wonderland Lake, located in Boulder, Colorado, served as the microbial source and is not impacted by runoff or wastewater effluent. City of Boulder 75th Street Wastewater Treatment Plant (WWTP) tertiary effluent (Boulder WWEff), located in Boulder, CO, served as the wastewater effluent source. Water quality data at

the time of sampling for each source can be found in Table 5.1. The media was already acclimated to Barker Reservoir from the work in Chapter 4, thus Barker Reservoir was tested first. Next, the media was acclimated to Wonderland Lake for two weeks and then the experiments were operated. Finally, the media was taken to Boulder WWTP and allowed to acclimate in a flow through pilot system for two weeks. Then the media was brought back to the lab and experiments were run.

Table 5.1 Source Water Characteristics

Parameter	Barker Reservoir	Wonderland Lake	Boulder WWEff.
DOC (mg/L)	3.6	10.3	6.32
BDOC (%)	20	21	14
UVA_{254} (cm^{-1})	0.11	0.16	0.13
SUVA (L/mg-C/m)	2.9	1.6	2.1
Alkalinity (mg-CaCO3/L)	20	120	110
pH	7.2	8.4	7.14
Ammonium (mg/L NH_3-N)	0.01	0.045	0.04
Nitrite (mg/L NO_2-N)	0.006	0.204	0.048
Nitrate (mg/L NO_3-N)	0.285	0.006	9.68
Phosphorous (mg/L $PO4_3$)	0.109	0.127	9.68
Silicon (ppm)	2.13	7.11	3.93
Manganese (ppm)	BDL	BDL	BDL
Iron (ppm)	0.01	BDL	0.014
Magnesium (ppm	0.95	73.95	16.88
Calcium (ppm)	4.01	108.8	46.99
Sodium (ppm)	1.43	111.4	60.93
Potassium (ppm)	0.62	5.95	14.27
Fluorine (ppm)	0.10	0.37	0.53
Bromide (ppm)	0.05	0.46	0.13
SO_4 (ppm)	2.68	408.7	80.12
Acetate (ppm)	BDL	BDL	0.42

*BDL = below detection limit

In certain experimental runs, the feedwaters were coagulated with aluminum sulfate ($Al_2(SO_4)_318H_2O$, Mallinckrodt Chemicals, 3208-04) and/or diluted with low carbon tap water. The low carbon tap water was City of Boulder tap water treated with granular activated carbon

(GAC) to reduce background DOC (<0.3 mgC/L). Alum coagulation dose was determined by performing a jar test using a 6-jar programmable jar tester (Phipps & Bird model 7790-901) with 2-liter B-KER^2 jars. Mixing conditions included a rapid mix phase (1 minute at 290 rpm), two flocculation phases (10 minutes at 55 rpm and 10 minutes at 20 rpm) and a 30-minute sedimentation phase. The alum doses were selected from the alum dose *vs.* DOC response curves. Alum doses of 20 mg/L, 80 mg/L, and 120 mg/L were chosen for Barker Reservoir, Wonderland Lake and Boulder WWEffl. respectively. Results from the jar tests can be seen in Figure 5.2, 5.3, 5.4.

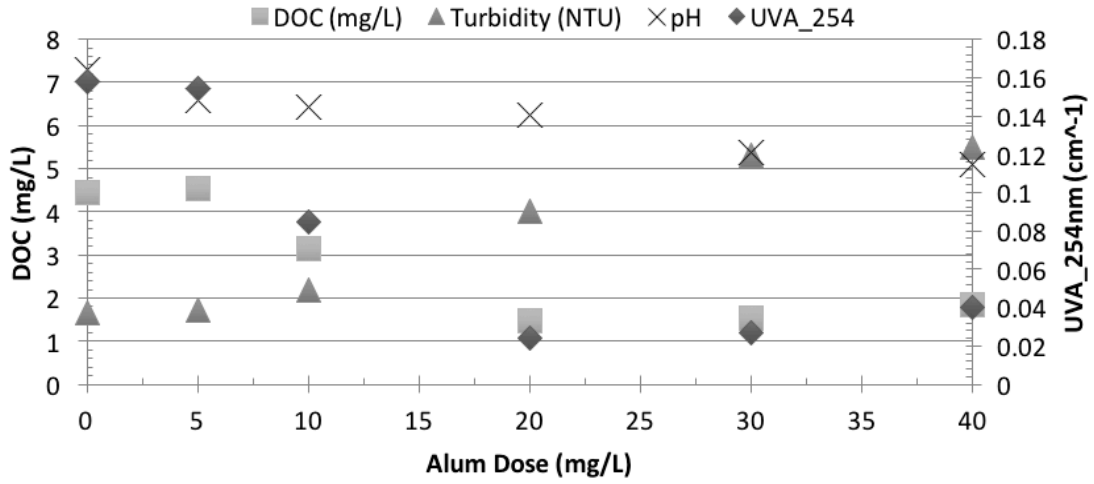

Figure 5.2 Barker Reservoir jar test results for DOC (mg/L), UVA_{254} (cm^{-1}), Turbidity (NTU) and pH.

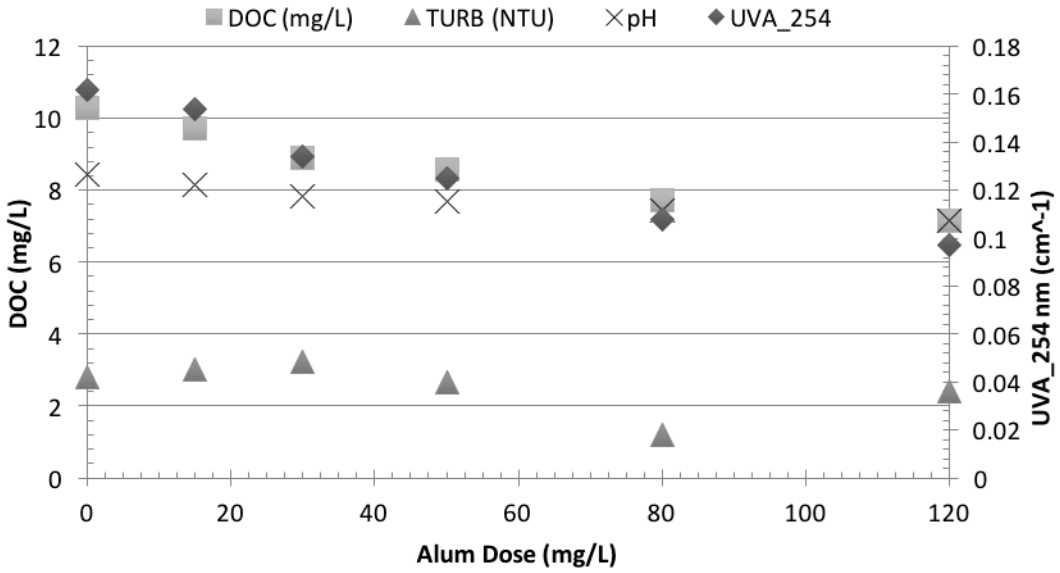

Figure 5.3 Wonderland Lake jar test results for DOC (mg/L), UVA_{254} (cm-1), Turbidity (NTU) and pH.

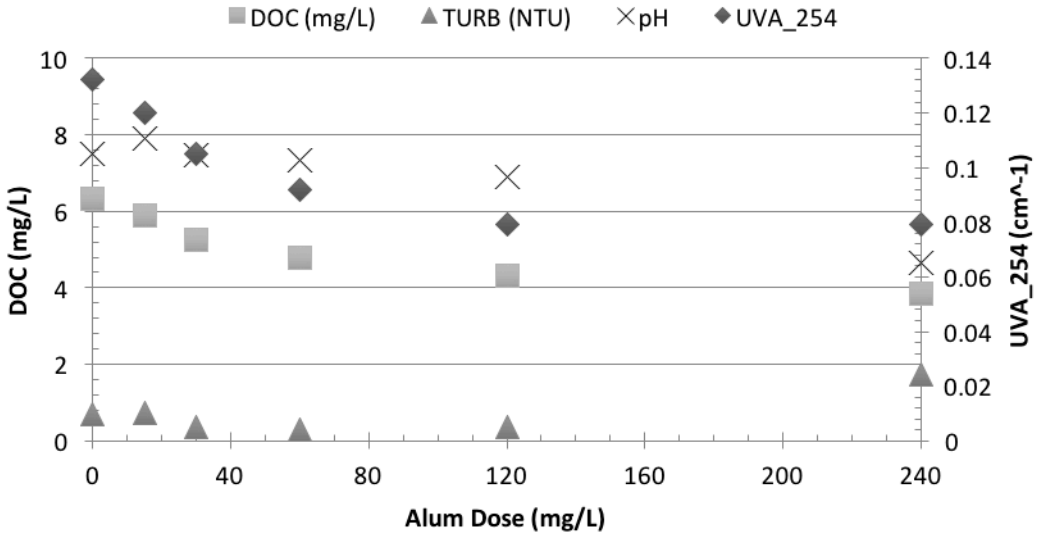

Figure 5.4 Boulder Wastewater Treatment Effluent jar test results for DOC (mg/L), UVA_{254} (cm-1), Turbidity (NTU) and pH.

5.2.3 Sample Analysis

Media samples were extracted weekly from the top of each column prior to changing experimental conditions. First liquid samples were taken, then the column was drained for biomass samples. Samples were taken at the top of each column representing EBCTs of <1 minute (top), 5 minutes and 15 minutes. Activity was measured on-site, directly after each sampling period, which eliminated temperature changes in the media after extraction. A

sterilized spatula was used to remove media from the top of the column and immediately placed in a sterile conical tube. Each media sample was first drip-dried using a vacuum with a 0.45-micron filter. To measure ATP, approximately one gram of media was weighed into a test tube and extracted using the LuminUltra Deposit and Surface Analysis Test (DSA-100C, Fredericton, NB) following the manufacturer's instructions. Activity samples were vortexed after each step to ensure sufficient mixing. A luminometer (Kikkoman C-110) was used to read light output from samples and results were given in relative light units (RLUs). The RLUs are converted to pg ATP/g using the ratio of the RLU's of the sample to the blanks and the mass of the sample. The dry:wet ratio for the media and the bed density were determined to normalize all results to biomass per mL of dry media. For this study, results were converted to ngATP/cm^3 media using a dry:wet ratio of 57% and an anthracite bed density of 0.8 g dry weight/cm^3.

Liquid samples were taken at the beginning and end of each column in order to measure biofilter performance for each filter EBCT. DOC samples were filtered through a 0.45 μm membrane filter (Pall Life Sciences). Filters were first rinsed with 250 mL of reverse osmosis water to assure that carbon leaching from the filters did not occur. Then samples were analyzed on a Sievers 5310 C Laboratory Organic Carbon Analyzer using the ultraviolet irradiation/persulfate oxidation method (SM 5310C). Biodegradable dissolved organic carbon (BDOC) was measured using the Mogren et al. (1990) method. A recirculating column with indigenous, acclimated bacteria attached to media was employed to measure the change in DOC over a period of 5 days at 20 °C. BDOC was measured as BDOC = DOC_{intial}-DOC_{final}. UVA_{254} was measured using a HACH DR/4000 Spectrophotometer. Samples were analyzed using a 1-cm quartz cell and absorbance values were reported with units of cm^{-1} as spectral absorbance coefficients. Filter influent was measured before each temperature run by a pH

probe, calibrated weekly with pH values of 4, 7, and 14. Measurements were made by rinsing the pH meter with deionized water and directly inserting the probe into each vial after sampling.

5.2.4 Disinfection Byproducts

DBP formation was evaluated on all three raw and treated source waters: Barker Reservoir, Wonderland Lake and Boulder WWeff. Bench scale chlorination was used to measure DBP formation following the uniform formation conditions (UFC) (Summers et al., 1996). A 24-hour chlorine residual of 1.0 mg/L (±0.4 mg/L) was determined by chlorine demand curves. The DPD (*N,N-diethyl-p*-phenylenediamine*)* colorimetric method (SM4500-Cl G) was used to measure the chlorine residuals and ammonium chloride was used to quench the samples, immediately following the 24-hour period. The samples were buffered with pH 8 borate buffer. Total trihalomethanes (TTHMs) and haloacetonitriles (HANs) were analyzed using EPA Method 551.1 (1995). Haloacetic acids (HAAs) were analyzed using EPA Method 552.2 An Agilent 6890 Gas Chromatography system with an electron capture detector (ECD) was used to analyze the samples. Total trihalomethanes included chloroform ($CHCl_3$), bromodichloromethane ($CHCl_2Br$), dibromochloromethane ($CHClBr_2$), and bromoform ($CHBr_3$). Haloacetonitriles included trichloroacetonitrile (C_2Cl_3N), dichloroacetonirtile (C_2HCl_2N), bromochloroacetonitrile ($C_2HBrClN$) and dibromoacetonitrile (C_2HBr_2N). Haloacetic acid data included the following HAA5s: chloroacetic acid ($ClCH_2COOH$), bromoacetic acid ($BrCH_2COOH$), dichloroacetic acid ($Cl_2CHCOOH$), trichloroacetic acid (Cl_3CCOOH) and dibromoacetic acid ($Br_2CHCOOH$); as well as the rest of the HAA9s: bromochloroacetic acid ($C_2H_2BrClO_2$), bromodichloroacetic acid ($C_2HBrCl_2O_2$), dibromochloroacetic acid ($C_2HBr_2ClO_2$), and tribromoacetic acid ($C_2HBr_3O_2$).

5.3 Results and Discussion

5.3.1 Biomass

Activity decreased throughout filter depth for each experiment, which was consistent with reviews on biofilter literature studies (Basu et al., 2015; Liu et al., 2017). At the top, 5 min EBCT and 15 min EBCT samples, Barker Reservoir activity measured 472 ngATP/cm^3, 215 ngATP/cm^3 and 111 ngATP/cm^3, respectively. Wonderland Lake measured overall higher activity with 560 ngATP/cm^3, 349 ngATP/cm^3 and 194 ngATP/cm^3, at the top, 5 min EBCT and 15 min EBCT sample points, respectively. Boulder Wastewater Treatment Effluent measured the highest activity with 904 ngATP/cm^3, 470 ngATP/cm^3 and 290 ngATP/cm^3, at the top, 5 min EBCT and 15 min EBCT, respectively, as seen in Table 5.2. The top of filter measured ATP values were in the 10^2 to 10^3 ngATP/cm^3 media range of literature data reported by Pharand et al. (2014). On average for all three water sources, activity at 5 min EBCT was 46% lower than activity at the top of the filter, and activity at 15 min EBCT was 69% lower than activity at the top of the filter. The biomass distribution was consistent with a review of literature activity distributions in Chapter 3 that found at 5 min EBCT activity was on average 40% lower than activity at the top of the filter and at 15 min EBCT activity was 66% lower than activity at the top of the filter (n=78 nonozonated biofilter studies). Overall, the wastewater effluent source had the highest activity measurements, followed by the microbial source then the terrestrial source. This suggest that biomass steady-state activity values differ depending on the water matrix.

Table 5.2 Average activity (ng ATP/cm^3), DOC removals, and UVA removals treating Barker Reservoir, Wonderland Lake and Boulder Wastewater Treatment Effluent.

	Average Activity (ng ATP/cm^3 media)			*DOC Removal (%)*	*UVA_{254} Removal (%)*
Sample Point	Top	EBCT 5 min	EBCT 15 min	EBCT 30 min	EBCT 30 min
Barker Res.	472	215	111	12	11
Wonderland Lake	560	349	194	11	6

WWEff	904	470	290	11	5

5.3.2 Dissolved Organic Carbon Removal

DOC removal throughout the biofilters were tested for multiple scenarios. For all temperatures (6°C, 22°C, 28°), Barker Reservoir removed an average of 12% DOC at 30-minute EBCT (n=5), Wonderland Lake removed an average of 11% DOC (n=4), and Boulder Wastewater Treatment Effluent removed an average of 11% DOC (n=5), as seen in Table 5.2. The results were similar to the values reported for 45 nonozonated waters that reported an average of 10% TOC removal for temperature range of 0.5°C to 35°C (Terry & Summers, 2017). Barker Reservoir had the lowest ATP values, yet exhibited similar DOC removals to Wonderland Lake and Boulder Wastewater Treatment Effluent. This suggest steady-state activity levels are unique to each water and it may not be useful to compare activity levels of differing NOM sources. In addition, Boulder Wastewater Treatment had the lowest overall BDOC of 14%, yet the biofilters removed similar levels of DOC (average 11-12%) from all the source waters. This suggest more DOC can be removed from each source water given further optimization.

5.3.3 Empty Bed Contact Time

Longer EBCTs increased DOC removal for all three water sources due to more exposure time which resulted in higher organics biodegradation, as seen in Figures 5.5, 5.6, and 5.7, for different temperatures and coagulation. All three source water yielded similar increase in removal, 5 to 6 %, when EBCT was increased from 5 minutes to 30 minutes. Wang and Summers (1996) saw similar, yet more pronounced trends as EBCT increased from 3 to 33 minutes, DOC removal increased from 16% to 24%. This study supports Urfer et al. (1997) literature review that concluded DOC removal increased with EBCT but in a less than a linear proportional way. A survey of 38 full-scale biofiltration facilities in North America found 50% of biofilters operated at an EBCT of 5-10 min (Brown et al., 2016), thus extending the EBCT can

increase organics removal, and the additional 5-6% of DOC removal can help utilities meet TOC removal and DBP regulations and reduce coagulant addition, albeit not interfering with water demand or particle removal goals.

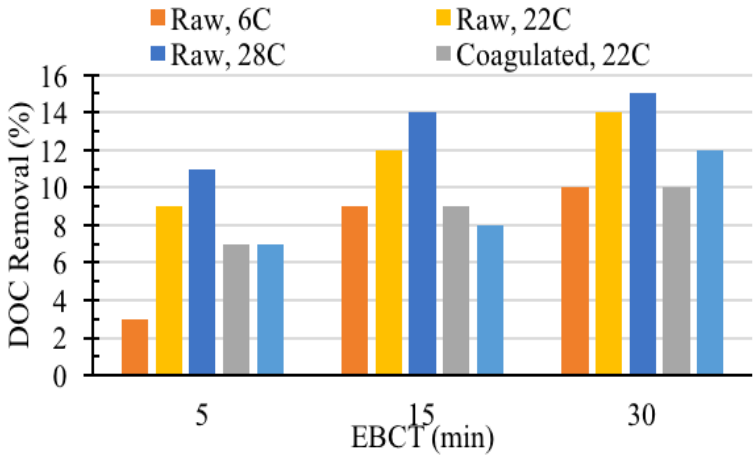

Figure 5.5 Barker reservoir DOC removal at EBCTs of 5, 15, and 30 minutes for multiple temperatures and scenarios: raw water at 6 °C, 22 °C, and 28 °C, coagulated at 22 °C, and diluted with dechlorinated tap at 22 °C.

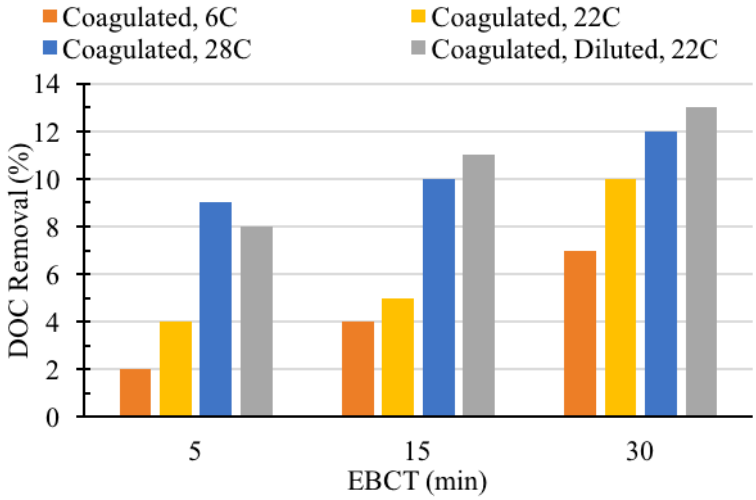

Figure 5.6 Wonderland Lake DOC removal at EBCTs of 5, 15, and 30 minutes for multiple temperatures and scenarios: coagulated at 6 °C, 22 °C, and 28 °C, and coagulated and diluted with dechlorinated tap at 22 °C.

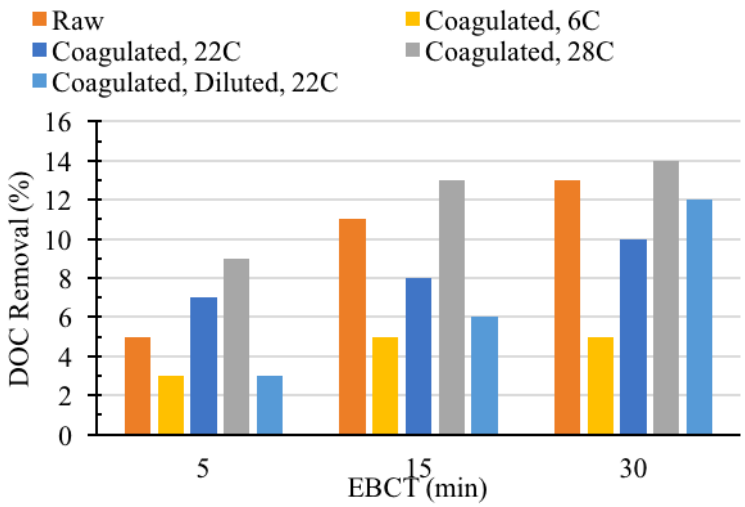

Figure 5.7 Boulder Wastewater Treatment Plant Effluent DOC removal at EBCTs of 5, 15, and 30 minutes for multiple temperatures and scenarios: raw water at 22 °C, coagulated at 6 °C, 22 °C, and 28 °C, and coagulated and diluted with dechlorinated tap at 22 °C.

5.3.4 Temperature

Temperature impacted DOC removal, as seen in Figures 5.5, 5.6, and 5.7. Biofilters exhibit a decrease in DOC removal performance at lower temperatures which can be attributed to decreased microbiological activity at the low temperatures (Liu et al., 2017). The experimental data was consistent with other studies that concluded temperature had an impact on biofilter performance due to temperature effects on degradation kinetics and microbial growth (Terry & Summers, 2017). Overall, a 22 °C increase in temperature was accompanied with a 5%-9% increase in DOC removal. Understanding the impact of temperature on biofilter performance is critical in the ability to optimize biofiltration and overall treatment performance during different seasonal temperature variations. Biofilter operation should be optimized for the winter and summer months to achieve the best performance out of the biofilter.

5.3.5 Influent Dissolved Organic Matter Characterization

Influent DOC was diluted to similar concentrations in all three source waters to determine effects of influent DOC characterization on DOC removal. Each of the source waters were diluted to a DOC of 3 mg/L and temperatures were held at 22 °C. At the influent of 3 mg/L,

DOC removals for all three source waters were the same; 12% to 13%. The similar removals of influent DOC despite different organic carbon origins suggests biofilters are robust in removing organic matter and can remove similar levels of organic matter from terrestrial, microbial and wastewater effluent sources. Utilities use a variety of source water matrixes and the robustness of biofilters to remove organics from different sources advances the knowledge of biofiltration performance and the applicability of biofiltration under different source water conditions.

SUVA values of the influent waters, 2.9 to 1.6 (L/mg-C/m), did not correlate well with DOC removal, which were not statistically different. Contrary to this study, Hozalski et al. (1999) found SUVA correlated inversely with DOC removal, as NOM sources with a lower SUVA value experienced more DOC removal by biodegradation in the SUVA range of 2.0 to 9.1(L/mg-C/m). In addition, Yapsakli and Çeçen (2010) found similar trends with higher biodegradability accompanied lower SUVA values, however, the GAC media was not exhausted and additional removal could have been a result of adsorption and not biodegradation. The trend of lower SUVA values being more susceptible to higher biodegradability applies to waters with SUVA less than 4 (L/mg-C/m) (Volk & LeChevallier, 2002), and all three water sources studied in this experiment were below 3 L/mg-C/m.

5.3.6 Disinfection Byproducts

Biofilters can remove DBPs and their precursors by biodegradation and biosorption, yet biofilters can contribute to DBPs and their precursors by biomass sloughing serving as a DBP precursor, sorption/desorption of DBPs and their precursors on the biomass, and form DBPs during nitrification/denitrication within the filter as review by Liu et al. (2017). In this study, DBP precursors were measured using the UFC approach (Summers et al., 1996). As seen in Table 5.3, Wonderland Lake and Boulder WWEff had high concentrations of bromide, which resulted in a shift of speciation towards HAA9s and TTHMs.

Table 5.3 Source water and DBP precursor data for Ter-NOM, Micro-NOM, and WWEff (average values)

	Ter-NOM	*Micro-NOM*	*WWEff*
SUVA (L/mgC/m)	2.9	1.6	2.1
Br^- (µg/L)	50	460	140
TOC (mg/L)	2.8	5	4.8
Br^-/TOC (µg/mg)	18	92	29
TTHM (µg/mL)	145	307	209
HAN4 (µg/mL)	4	20	17
HAA5 (µg/mL)	135	162	219
HAA9 (µg/mL)	145	361	316
TTHM/TOC (µgDBP/mgTOC)	45	24	24
HAN4/TOC (µgDBP/mgTOC)	1.4	3	3
HAA5/TOC (µgDBP/mgTOC)	41	10	18
HAA9/TOC (µgDBP/mgTOC)	44	31	35

After coagulation and dilution, on average the highest level of removals were seen at the longest EBCT and highest temperature due to longer exposure time for organics degradation and higher rates of enzymatic activity to contribute to organic degradation. DBP precursor removals at 28°C and 30 min EBCT for each water can be seen in Figure 5.8. Other studies showed similar results as a biofilter removed an average of 6% of THM4 and 12% of HAA9 (Chaiket et al. 2002), an aerated biofilter removed 70% of THM and 50% of HAA (Pramanik et al. 2015), and a biofilter treating river water removed 38% of THM and 73% of HAA9. Biofilters can help mitigate the risk of DBPs by removing their precursor material, organic matter, and lowering the chlorine dose needed.

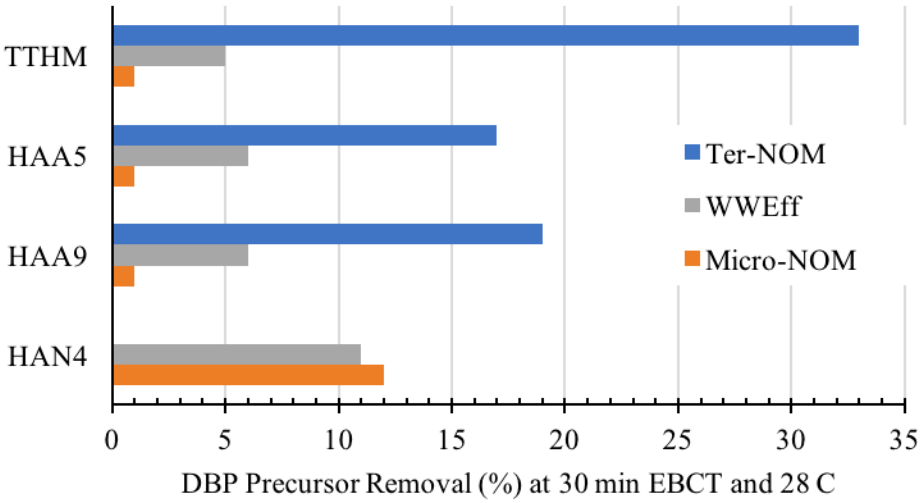

Figure 5.8 DBP precursor removals for TTHM, HAA5, HAA9, and HAN4 for Barker Reservoir, Wonderland Lake, and Boulder WWEff at 28 °C and 30 minute EBCT.

SUVA did not prove to be a useful indicator of reactivity in the formation of DBPs, as Barker Lake had the highest SUVA of 2.9 (L/mg-C/m), yet formed the lowest levels of DBPs, as seen in Table 5.3. Wonderland Lake had the lowest SUVA of 1.6 (L/mg-C/m) yet formed the most DBPs and Boulder WWEff had a SUVA of 2.1 (L/mg-C/m) and formed a similar number of DBPs to Wonderland Lake. Poor correlation could result because not all organic matter absorbs light at 254 nm, yet the organic matter can still contribute to the formation of DBP precursors. In addition, some organic matter may absorb light at 254 nm but not form DBPs. Weishaar et al. (2003) also found SUVA was a weak universal predictor of reactivity for the formation of DBP precursors s because DOC chemical compositions could have similar properties, elemental analysis, or size analysis, yet different.

Specific DBP yields give insight to DBP precursor formation based on TOC content of the water. SUVA values correlated linearly with DBP yields (excluding HANs). The R^2 values correlated SUVA to DPB yields (HAA9/TOC, TTHM/TOC, and HAA5/TOC) were 0.99, 0.85, and 0.98, respectively, as seen in Table 5.3 and Figure 5.9. This suggest the aromaticity of the

water is a good indicator for DBP yields, with higher SUVA values correlating with higher yields.

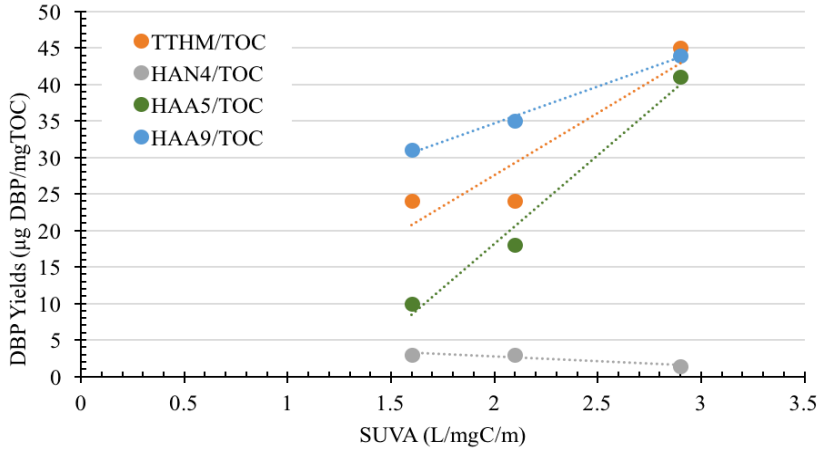

Figure 5.9 DBP precursor yields (µg DBP/mg TOC) as a function of SUVA for TTHMs, HAA5, HAA9 and HAN4, for all waters.

5.3.7 Modeling Biofilter Performance

The predictive model developed in Chapter 3 was applied to biofilters treating all three source waters. The model predicts TOC removal based on total biomass activity, which is total biomass within the filter found by measuring biomass at the top of the filter and using literature data derived equations to determine biofilter biomass profiles. For each of the experimental matrix scenarios, activity measurements at the top of the filter were measured and used to find total activity.

$$X_{avg} = \frac{X_{top}(a*(EBCT*\ln(EBCT)-EBCT)+b*EBCT)}{EBCT} \qquad \text{Eqn. 5.1}$$

Where, X_{avg} is the average biomass within the filter, X_{top} is the biomass measured at the top of the biofilter, and *a* and *b* are constants derived from literature studies (a=-0.12, b=0.72).

DOC removal measurements were measured and plotted with respect to total activity, as seen in Figures 5.10, 5.11 and 5.12. For the predictive model, BDOC percentages from each water were

measured and the best fit k″ constant was employed. The following equations were used to model performance:

$C_{eff}/C_{inf} = BOMfrac\ (\exp\ (-k'' * X_{avg} * EBCT))$ Eqn. 5.2

$X_{total} = X_{avg} * EBCT$ Eqn. 5.3

C_{eff}/C_{inf} is the contaminant fraction remaining, *BOMfrac* is the biodegradable fraction of organic carbon, k″ is the rate constant, and X_{total} is the total activity.

Barker Reservoir with 20% BDOC and a best k" fit of 1.8E-4 mL*ngATP^{-1}*min^{-1} fit the model well with an average DOC removal residual of 1.84% (n=18), as seen in Figure 5.10. Wonderland Lake with 21% BDOC and a best k″ fit of 1.3E-4 mL*ngATP^{-1}*min^{-1} fit the model well with an average DOC removal residual of 1.99 % (n=15), as seen in Figure 5.11. Boulder Wastewater Treatment Effluent with 14% BDOC and a best k" fit of 1.5E-4 mL*ngATP^{-1}*min^{-1} fit the model well with an average DOC removal residual of 1.71 % (n=15), as seen in Figure 5.12. Chapter 3 literature data best fit k″ for 20% BDOC nonozonated waters was 3.61E-4 mL*ngATP^{-1}*min^{-1} (n=41). The literature k″ applied to the model for each water with respective BDOC percentages instead of the best fit k″ resulted in average TOC removal residuals of 3.28 %, 5.63 %, 3.65%, for Barker Reservoir, Wonderland Lake and Boulder Wastewater Treatment Effluent, respectively. The average residuals are low considering 15-21% of the DOC is biodegradable. The ability to predict biofilter performance, on a range of different NOM sources, would allow optimization of treatment plant operations to improve overall plant efficiency and improve the acceptance of biofiltration as an appropriate treatment technology.

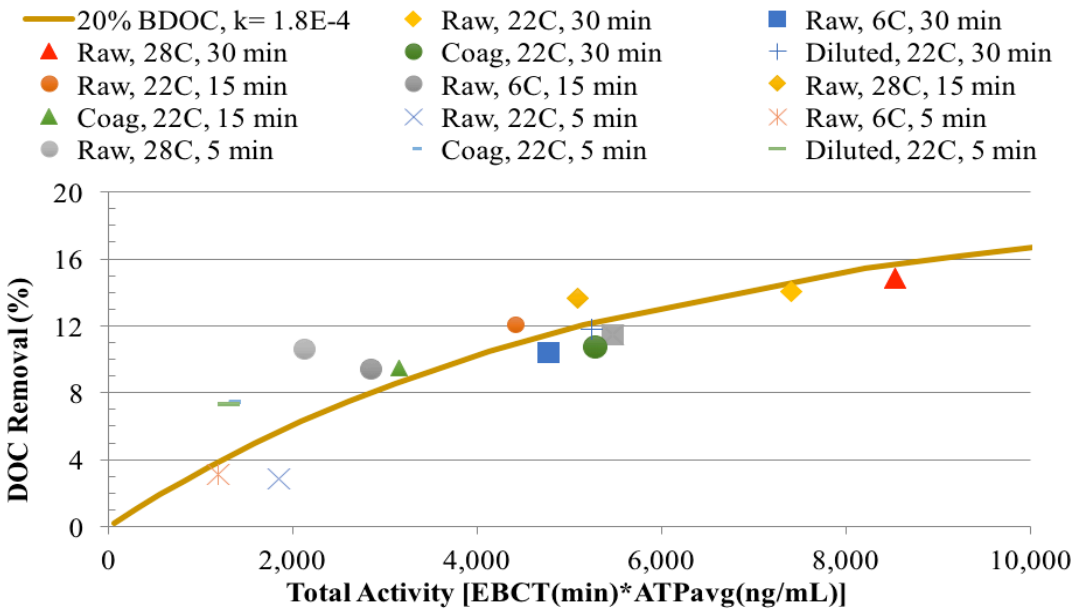

Figure 5.10 Barker Reservoir total activity (EBCT(min)*ATP$_{avg}$(ng/mL)) vs. DOC removal for multiple temperatures, EBCTs and experimental scenarios modeled with 20% BDOC and k" = 1.8E-04 mL*ngATP^{-1}*min^{-1}.

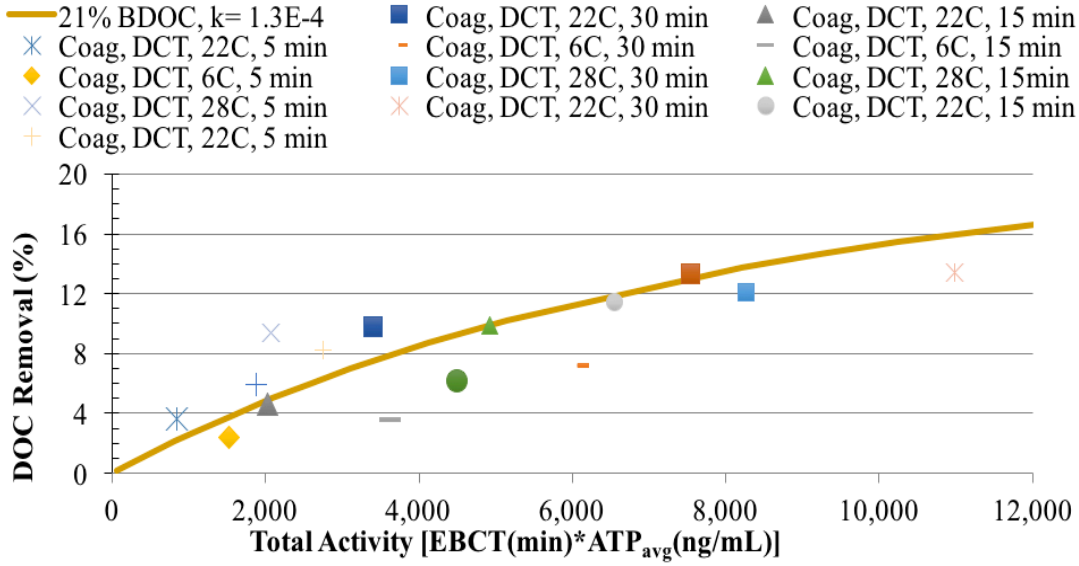

Figure 5.11 Wonderland Lake total activity (EBCT(min)*ATP(ng/mL)) vs. DOC removal for multiple temperatures, EBCTs and experimental scenarios modeled with 21% BDOC and k" = 1.3E-04 mL*ngATP^{-1}*min^{-1}.

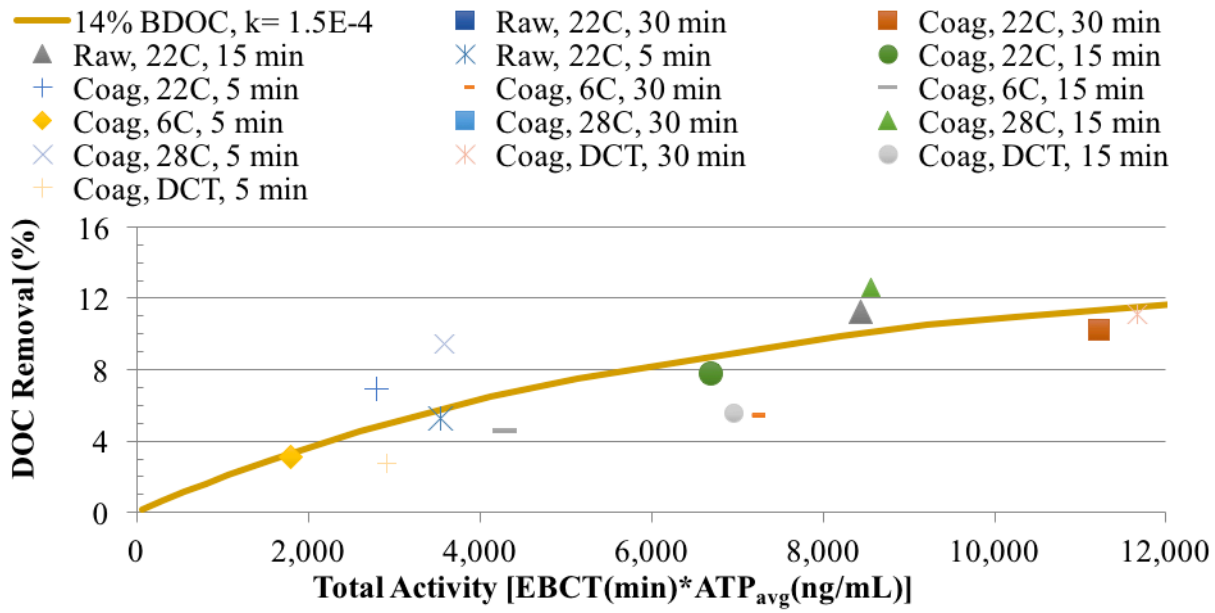

Figure 5.12 Boulder Wastewater Treatment Effluent total activity (EBCT(min)*ATP_{avg}(ng/mL)) vs. DOC removal for multiple temperatures, EBCTs and experimental scenarios modeled with 14% BDOC and k″ = 1.5E-04 mL*ngATP^{-1}*min^{-1}.

The models for each water compared to one another through the normalization of the extent of each reaction, i.e., BDOC fraction, demonstrated the kinetics of the NOM of each water, as seen in Figure 5.13. Barker Reservoir had the highest rate constant of 1.8E-4 mL*ngATP^{-1}*min^{-1}, which suggests a higher kinetic reaction of the NOM. Boulder wastewater effluent had a lower rate constant of 1.5E-4 mL*ngATP^{-1}*min^{-1}, which suggest the NOM kinetics were slower than the terrestrial source. Wonderland Lake had the lowest rate constant of 1.3E-4 mL*ngATP^{-1}*min^{-1}, which suggest the kinetics of the microbial source were the slowest out of all three sources. The lower kinetics for the wastewater effluent and microbial source can be explained by the origin of the NOM. The fast reacting NOM of the microbial source potentially degraded in the microbial system (Wonderland Lake), and the fast reacting NOM of the wastewater effluent potentially degraded in the wastewater treatment biological processes. However, the fast reacting NOM in Barker Reservoir had not yet been exposed to microbial activity prior to our biofiltration system, thus a higher rate constant was found.

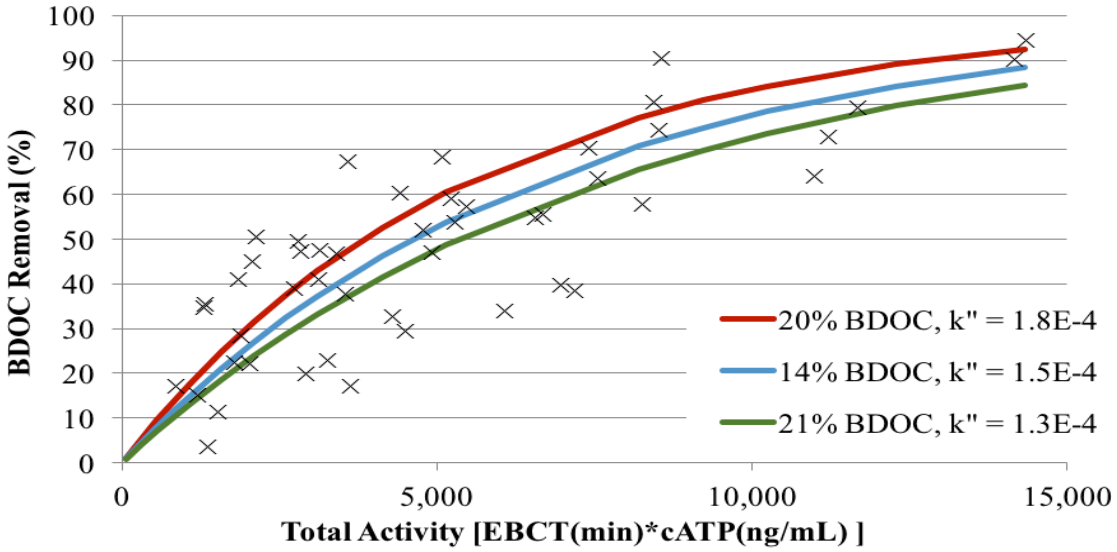

Figure 5.13 Total activity (EBCT(min)*ATP_{avg}(ng/mL)) vs. BDOC removal for each water at multiple temperatures, EBCTs and experimental scenarios modeled with respective k" (mL*ngATP^{-1}*min^{-1}) constant and normalized reaction extent, i.e., BDOC fraction.

5.4 Conclusions

This study evaluated operating parameters (EBCT, biomass) and water quality (temperature, DOM matrix) impacts on biofiltration performance. An increase in EBCT from 5 to 30 minutes improved DOC removals by 5-6% (n=14). An increase in temperature from 6°C to 28 °C improved DOC removals by 5-9% (n= 9). DOC origin, e.g., terrestrial, microbial or wastewater effluent, did not impact biodegradation, as the same DOC removal was observed for each source with the similar influent DOC. Each water had a different extent of reaction, as measured by BDOC fraction, yet Barker Reservoir had slightly higher kinetics than Wonderland Lake and Boulder Wastewater Treatment Plant Effluent. SUVA did not correlate well with biodegradation potential in the range of 1.6 – 2.9 (L/mg-C/m) nor did SUVA correlate with DBPs formed. However, SUVA did prove to be a useful indicator of DBP yields. DBP precursors were best controlled during longest EBCTs as higher biodegradation of the organic matter occurred. Modeling biofilters will prove to be a huge asset to water treatment plants, and the biodegradable fraction of the water and the contaminant utilization rate constant, k", are fundamental for improved model prediction accuracy.

Chapter 6
Environmentally Sustainable Scenarios for Drinking Water Biological Filtration

*This chapter has the following co-authors: Christopher Jones, Scott Summers and Sherri Cook. This work is currently under review with Water Research.

6.1 Introduction

Drinking water plants are facing the challenge of meeting increasingly stringent regulations, often at higher costs and operational complexity. In addition, the quality, such as total organic carbon (TOC), of source waters are expected to degrade as populations and use of non-traditional source waters increase (Delpla et al., 2009; Raseman et al., 2017; Todd et al., 2012; Vorosmarty et al., 2000). During conventional surface water treatment, chemical coagulation coupled with flocculation, sedimentation, and filtration is a common treatment approach to lower turbidity, remove organic matter, and control the formation of disinfection by-products (DBPs) (Howe et al., 2012). While effective, the excessive use of coagulation chemicals adds burdens to the water treatment plant, such as increased costs, chemical handling, and residuals management (U.S. EPA, 2005).

An alternative to conventional treatment, the biological treatment of drinking water by using biofiltration achieves organic matter removal by biodegradation, thus reducing coagulant use and pH adjustment chemicals. Previous studies have found that biofiltration can provide the same or better final water quality. For example, biofiltration has been found to achieve the same filtered water turbidity regulations (Emelko et al., 2006; Juhna and Melin, 2006; Stoddart and Gagnon, 2015; Urfer et al., 1997; Volk et al., 2000) and even better pathogen removal, including *Cryptosporidium* (Amburgey et al., 2005), as conventional filtration, and to achieve trace organic compound removal, such as 2-methylisoborneol and geosmin (Zearley, 2012). Also, previous

studies have experimentally compared biofiltration with conventional filtration, both using a dual media fixed bed configuration, and found similar unit filter run volumes and filter run times for each (LeChevallier et al., 1992; Stoddart & Gagnon, 2015), suggesting that changing to biofiltration could be an easy transition. While there are many benefits to biofiltration, there may be tradeoffs as well. For instance, biofilters require an acclimation period around 3 to 6 months before achieving steady-state contaminant removal (Servais et al., 1994). Also, seasonal variations in water quality can affect biofilter organic matter removal efficiency, especially temperature (Emelko et al., 2006; Liu et al., 2001). In addition, the integration of biofiltration into an overall treatment train has unclear systematic impacts. For example, compared to conventional filtration, biofiltration can decrease coagulant use due to biological TOC removal, but biofiltration may inadvertently increase the amount of chlorine needed since it cannot receive disinfection credits until after the biofilter. Also, while the alum dose may decrease, pH adjustment chemicals may increase due to the complex interactions between coagulation chemicals, pH, and alkalinity. Given the complexity of these treatment processes, a systems approach is needed to quantitatively evaluate the benefits of and tradeoffs between the different filtration options.

Multiple drinking water life cycle assessment (LCA) studies have compared diverse filtration alternatives using a systems approach, but these studies focused mostly on physical-chemical processes, such as using granular activated carbon for tertiary drinking water treatment (Amores et al., 2013; Barjoveanu et al., 2014; Barrios et al., 2008; Bonton et al., 2012; Garfí et al., 2016; Vince et al., 2008), and comparing membrane to conventional filtration (Bonton et al., 2012; Friedrich and Buckley, 2002; Garfí et al., 2016; Vince et al., 2008). There have been limited studies evaluating biofiltration, and a single study focused on sorptive attachment

materials, such as granular activated carbon (e.g., (Santana et al., 2014)). However, the use of non-sorptive media, such as anthracite over sand, can reduce costs (U.S. EPA, 2005) and possibly environmental impacts, especially since sorptive media LCAs found that media production was a significant source of environmental impacts (Bonton et al., 2012; Garfí et al., 2016; Santana et al., 2014). Also, a dual media filter using anthracite and sand could allow for a simple transition from a conventional filter to a biofilter by moving chlorine addition after the filter and allowing for acclimation (Stoddart and Gagnon, 2015). While LCA can be used to improve a process's performance, such as by identifying ways to reduce chemicals or costs, unfortunately, previous LCAs have not yet assessed this transition option. Furthermore, most previous drinking water LCAs have been based on case studies (Amores et al., 2013; Barjoveanu et al., 2014; Bonton et al., 2012; Friedrich and Buckley, 2002; Garfí et al., 2016; Santana et al., 2014), which can limit their wide application to multiple drinking water treatment plants and diverse source waters. General applicability is particularly important given the impact of seasonal water quality changes on treatment performance and given the expected degradation of source waters over time.

Therefore, the goal of this work was to identify and quantify the multiple tradeoffs between biofiltration and conventional filtration, for a wide array of source waters, using a systems approach. Specifically, a life cycle assessment (LCA) model was developed and used to elucidate the operational and source water quality conditions that can result in considerable environmental impact and cost reduction when using either biofiltration of conventional filtration. The biofiltration treatment train used the same processes as conventional filtration, except chlorine was added after the filter instead of directly before (Figure 6.1). Both treatment trains were designed to produce final water that met the same surface water regulations.

Sensitivity analyses were conducted to quantify uncertainty and identify important model assumptions. First, the relative environmental impacts were quantified for a source water representing average U.S. water quality parameters, in terms of TOC, pH, specific ultraviolet absorbance (SUVA), alkalinity, and temperature. Then, to evaluate the impact of source water quality on the environmental comparison, the LCA model was used to evaluate 135,000 unique source waters. Water treatment plants can use this new LCA model and data to assess the tradeoffs between using biofiltration or conventional filtration.

6.2 Methods

6.2.1 Life Cycle Assessment

To compare biofiltration with conventional filtration, comparative LCA methodology following the ISO 14040 framework was used (International Organization for Standardization, 1997). Both filtration alternatives were considered in the context of a full treatment train, which consisted of coagulation, flocculation, sedimentation, (bio)filtration, disinfection, and final water pH adjustment. The functional unit was to treat 1.0 Mm^3/yr (0.7 mgd) of surface water over 40 years to meet the following regulations: Surface Water Treatment Rule (SWTR) (U.S. EPA, 1989), which requires 3-log *Giardia* and 4-log virus removal/inactivation; Long Term 2 Enhanced Surface Water Treatment Rule (LT2ESWTR) (U.S. EPA, 2006), which requires 2-log *Cryptosporidium* removal/inactivation and effluent turbidity limits; and the Stage 1 DBP Rule (U.S. EPA, 1998), which requires organic matter removal (termed the enhanced coagulation requirement) between 15% to 50% TOC removal based on source water TOC and alkalinity (Appendix D Table D1).

Due to the dependence of treatment requirements on source water quality, this study generated 135,000 unique sets of water quality values to comprehensively evaluate water treatment plants

throughout the U.S. (Appendix D Table D2). Water quality included TOC, pH, SUVA, alkalinity (measured as calcium carbonate, $CaCO_3$), and temperature. There were three temperature ranges in the experimental literature that described percent TOC removal during biofiltration (Terry and Summers, 2017): 10% at 10 °C or lower, 12% between 10 °C and 20 °C, and 15% at 20°C or higher. There were 45,000 scenarios (i.e., unique combinations of TOC, pH, SUVA, and alkalinity) generated for each temperature range. The combination of water quality values for each scenario was generated using Latin Hypercube sampling of three continuous uniform distributions: 2.0 to 8.0 mg/L TOC; 6.0 to 8.5 pH; 2.0 to 5.0 L/mg/m SUVA; and one discrete uniform distribution: 0.0 to 125 mg/L $CaCO_3$ (in 1 mg/L increments). These water quality ranges were chosen to represent all possible source water scenarios that required enhanced coagulation.

Each scenario's combination of source water quality values was a unique input into the LCA model. The model framework and major assumptions were summarized below, with more details in Appendix D. Figure 6.1 shows the LCA system boundary, which includes hauling and the production of chemicals, electricity, and infrastructure materials needed for the treatment processes. Requirements that were equivalent for both alternatives were excluded from the comparative analysis (e.g., flow dependent requirements, such as sedimentation tank material, coagulation-flocculation mixing energy). Life cycle inventory data was collected from the Ecoinvent v3 (Swiss Centre for Life Cycle Inventories, 2014) and US-EI 2.2 databases (Earthshift, 2014) (Appendix D Table D4). Material, energy and chemical quantities for the entire functional unit for each filtration alterative were calculated (Appendix D Table D4). Life cycle emissions were based on the total amount and type of chemicals, energy, hauling, and materials required over 40 years of operation and accounted for typical replacement rates (U.S. EPA, 2004, 2003). The life cycle emissions were translated into 10 environmental impact

categories using the EPA's Tool for Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI) (Appendix D Table D5).

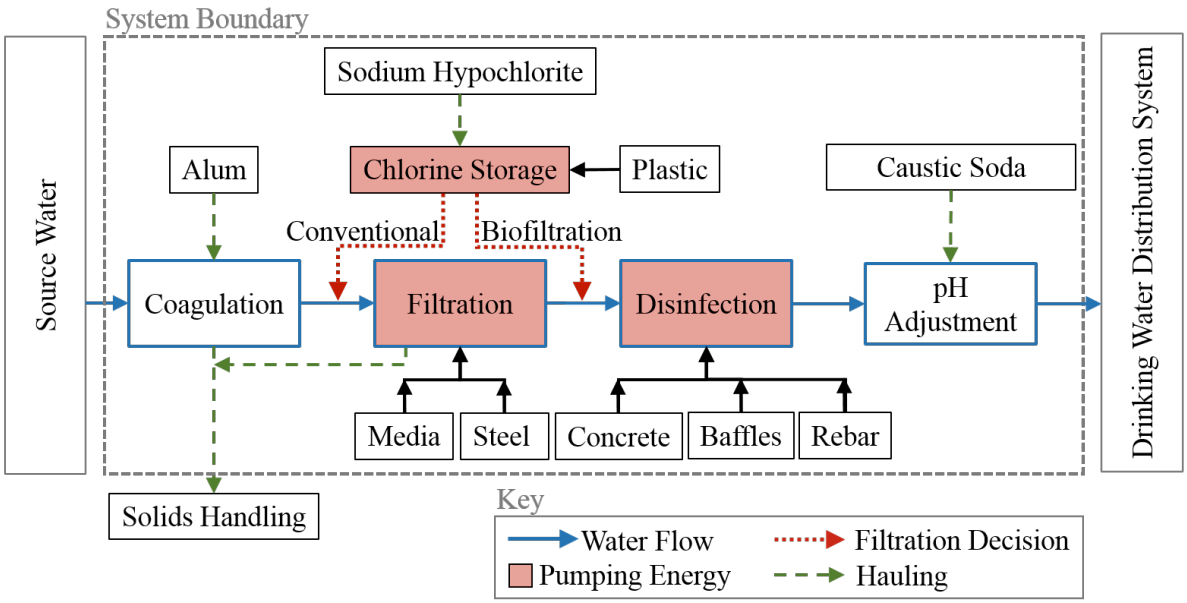

Figure 6.1 Process flow diagram for the LCA system boundary, which includes the hauling of chemicals and solids and the production of chemicals, electricity, and infrastructure materials needed for the treatment processes.

6.2.2 Treatment Process Modeling

6.2.2.1 Coagulation

Aluminum sulfate (alum), an effective and common coagulant, was used. The production and hauling of alum to the water treatment plant was included. The required alum mass was based on the turbidity and enhance coagulation requirements. The enhanced coagulation requirement uses bins of source water quality (combinations of TOC and alkalinity values) with different percent TOC removal requirements that account for economic limitations; for example, since source waters with high alkalinity and low TOC values are difficult to treat (i.e., can require unfeasibly high alum doses), the required percent TOC removal for these source waters is lower (McGuire et al., 2002; U.S. EPA, 1998) (Appendix D Table D1). This requirement specifies that one of three compliance criteria be met: (i) the bin's percent TOC removal, or (ii) 2 mg/L TOC, or (iii) 2 L/mg/m SUVA (U.S. EPA, 1998). The alum dose associated with each

criteria was calculated using the Edwards Langmuir-based semi-empirical model (Edwards, 1997), which accounted for influent TOC, pH, temperature, alkalinity, and SUVA. The smallest dose of the three compliance criteria was used to satisfy the requirement. However, that dose can be lower than the dose required for turbidity removal (i.e., LT2ESWTR requirements). The alum dose required for turbidity was based on typical alum doses used at U.S. water treatment plants before enhanced coagulation was common, which had a median value of 18 mg/L (Randtke et al., 1994). This turbidity dose represented the minimum allowable alum dose. Therefore, the modeled alum dose was the larger of the two requirements, turbidity and enhanced coagulation, to assure that both were met (Appendix D Section D3.1). For biofiltration, the requirements were the same, but the percent TOC removal needed by coagulation was reduced by the TOC removal achieved by biodegradation in the filter.

6.2.2.2 Filtration

Both the conventional filter and biofilter were rapid, dual media filters that used anthracite followed by medium sand. The stainless-steel filter housing, anthracite, and sand masses were based on filter dimensions (Appendix D Section D4.1). Area was based on typical hydraulic loading rates (10 to 25 m/h) (Kawamura, 2000). Height was based on the depth needed during operation (assuming typical 0.45 m anthracite and 0.3 m sand packed media depths, 1.5 to 2.5 m water height above the media, and freeboard) (Kawamura, 2000). A square filter cross section, steel thickness (Colorado Department of Public Health and Environment and Department of Civil and Environmental Engineering Colorado, 2014), and media densities (Urfer et al., 1997)were also assumed. The filter energy requirement included operation and backwashing (Appendix D Section D4.2). Operational pumping was based on the total filter height. Backwashing energy was based on a pumping pressure (9 m), flow (50 $m^3/h/m^2$), and frequency of once per day for 10 minutes (Howe et al., 2012). Pumping was required to overcome all head

losses throughout the plant (e.g., there was no excess head). Due to the lack of a common solids handling process, only solids hauling was included. It accounted for the total mass of alum used and TOC removed during the coagulation process (Appendix D Section D4.3). Since previous studies have shown that biofiltration achieved similar effluent turbidity as conventional filtration under identical operating conditions (Emelko et al., 2006; Juhna and Melin, 2006; Stoddart and Gagnon, 2015; Urfer et al., 1997; Volk et al., 2000), it was assumed that biofiltration achieved the same pathogen removal credits (U.S. EPA, 2006): 2-log *Giardia* and 2-log *Cryptosporidium* removal.

6.2.2.3 Disinfection

Chlorine disinfection used sodium hypochlorite to achieve the remaining 1-log *Giardia* and 4-log virus inactivation. Overall, the disinfection calculations were based on the LCA drinking water disinfection model developed by Jones et al. (2017). In summary, the concentration times time (CT) was determined based on the inactivation goals and the water's pH and temperature. The required dose was then calculated by assuming a 1.0 mg/L residual of free chlorine leaving the plant and by accounting for the water's chlorine demand (U.S. EPA, 2001). The amount of plastic for the storage tank and chemical hauling requirements were based on this dose and weekly chemical deliveries. Contact zone materials' masses and pumping requirements to overcome head loss were determined for a concrete basin with two steel baffles and based on the required contact time. Chlorine injection occurred after the filter during biofiltration and before the filter during conventional filtration. For biofiltration, the chlorine contact time was achieved only in the contact zone. The contact time achieved in the conventional filter was deducted from the total required contact time; this allowance varies by state, and this conservative assumption minimized chemical and infrastructure requirements for conventional filtration.

6.2.2.4 pH Adjustment

The final water's pH was raised to 8.2 at the end of the plant using caustic soda (sodium hydroxide) (Appendix D Section D5). To determine the amount needed, carbonate chemistry relationships from the U.S. EPA's Water Treatment Plant Model v2 (U.S. EPA, 2001) were used to first determine the pH after coagulation (Appendix D Eq. D1) and after chlorination (Appendix D Eq. D10) and then determine the caustic soda dose needed to achieve a final pH of 8.2 (Appendix D Eq. D11). Hauling was based on the total, required mass.

6.2.3 Uncertainty and Sensitivity Assessments

The impact of variability in major assumptions was assessed using a Monte Carlo analysis with the software Crystal Ball™ (Oracle, 2008). There were 24 uncertainty parameters that accounted for chemical and filter design variables (Appendix D Table D11). All parameters were assigned a uniform distribution, which best characterized the available data, except the minimum allowable alum dose (McGuire et al., 2002) (Appendix D Table D12) and biofiltration percent TOC removal (Terry and Summers, 2017) (Appendix D Figure D2) had triangular distributions. Uncertainty results were the output of 10,000 Monte Carlo simulations. The biofiltration and conventional filtration results were presented as a pair for each randomly generated set of uncertainty parameter values. The sensitivity of each TRACI category to the input parameters was determined by comparing Spearman's rank correlation coefficients; a category was defined as sensitive to a parameter if the corresponding correlation coefficient's magnitude was greater than 0.8 ($|\rho| > 0.8$).

6.3 Results and Discussion

6.3.1 Typical Source Water Quality Scenario

Figure 6.2 shows the environmental impacts of biofiltration relative to conventional filtration for a typical source water scenario based on national average values (McGuire et al., 2002): 3.2 mg/L TOC, 7.6 pH, 3.1 L/mg/m SUVA, 77 mg/L $CaCO_3$, and 15°C. A value less than one for a biofiltration total impact or for an uncertainty ratio signifies that biofiltration had better environmental performance than conventional filtration in that category. For this typical source water, biofiltration had better environmental performance than conventional filtration in all 10 categories. Specifically, biofiltration had lower negative impacts that were only 70% to 80% of the conventional filtration's impacts in all categories. Lower biofiltration environmental impacts were mostly due to biological TOC removal, which reduced the required alum dose and sequentially also reduced the caustic soda dose.

Alum production had the largest contribution to impacts, accounting for 22% to 72% of the conventional filtration impacts and 16% to 57% of the biofiltration impacts in any given category (dashed portions in Figure 6.2). Other major impact contributions were due to the production of caustic soda, filter operational energy, and filter steel (Appendix D Figure D4). Caustic soda impacts accounted for around 20% of environmental impacts in most categories, for both conventional filtration and biofiltration. In general, chemical production had the largest contribution to environmental impacts (up to 90% for conventional filtration and 87% for biofiltration). Other studies have also found that chemical production, especially of coagulants and pH adjustment chemicals, were a significant source of environmental impacts (Barrios et al., 2008; Bonton et al., 2012; Garfí et al., 2016; Mo et al., 2010; Venkatesh and Brattebø, 2011; Vince et al., 2008). Filter operational energy and steel production for the filter housing both

contributed around 15% of impacts for conventional filtration and 10% for biofiltration in most categories. The environmental impact contribution from filter media and baffle production were found to be negligible (less than 10% of any impact category). This is consistent with other studies that found steel dominated infrastructure impacts (Friedrich and Buckley, 2002; Jones et al., 2017; Lundie et al., 2004; Mo et al., 2010; Vince et al., 2008) but that overall, infrastructure impacts were minimal for drinking water treatment plants (Bonton et al., 2012; Friedrich and Buckley, 2002; Igos et al., 2014; Lemos et al., 2013; Loubet et al., 2014; Mery et al., 2013; Racoviceanu et al., 2007).

When accounting for uncertainty, biofiltration was still found to have better environmental performance (Figure 6.2 boxplots). The ranges of uncertainty ratios show that, even when conservative biofiltration values were considered (i.e., values leading to worse biofiltration environmental performance), biofiltration impacts were smaller and between 72% to 91% of the conventional filtration impacts, in all categories. The results for this typical source water scenario were not sensitive to any uncertainty parameters. Only two parameters had notable correlations with the results: minimum turbidity alum dose (ϱ=0.65) and biofilter percent TOC removal (ϱ=-0.5). Biofiltration's best environmental performance corresponded with the smallest minimum allowable alum dose (11 mg/L) and largest biofilter percent TOC removal (20%); this combination best emphasized biofiltration's capacity to minimize alum dosing due to biodegradation.

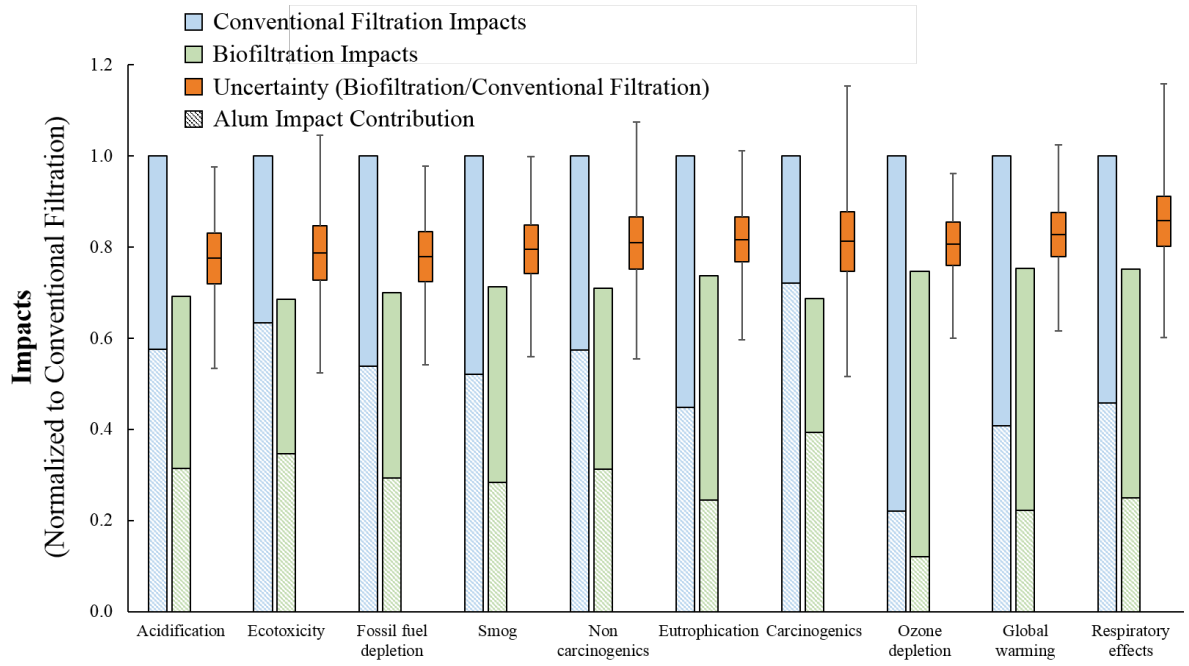

Figure 6.2 Comparison of conventional filtration (blue) and biofiltration (green) environmental impacts for the typical source water scenario for all 10 TRACI categories. Dashed region of each box shows the contribution of alum production and hauling to total impacts. Uncertainty results from the Monte Carlo analysis were shown as a box plot for each category (orange). All impacts were normalized to the conventional filtration impact in each of the 10 Traci categories. Source water values represented U.S. national averages (McGuire et al., 2002): 3.2 mg/L TOC, 7.6 pH, 3.1 L/mg/m SUVA, 77 mg/L $CaCO_3$, and 15 ˚C.

For this typical source water scenario, the use of biofiltration achieved at least a 25% impact reduction in all categories when compared to conventional filtration. The annual reduction in global warming impacts, for example, would be equivalent to reducing annual gasoline consumption by more than 1,800 gallons or 40,000 miles driven per year (U.S. EPA, n.d.). In particular, the use of biofiltration reduced the amount of chemicals by about 36%, specifically alum by 45% and caustic soda by 25%. When accounting for typical unit costs of alum, caustic soda, and sodium hypochlorite (U.S. EPA, 2005) (Appendix D Section D6), the cost savings expected with biofiltration was $7,500 per year. Also, more savings could be realized due to the reductions in solid waste generation from coagulation and in chemical hauling

and storage capacity. While there were significant environmental and cost benefits found for this typical source water scenario, the relative biofiltration benefits were largely dependent on source water quality.

6.3.2 Comprehensive Source Water Quality Scenarios

Since regulatory treatment requirements are based on source water quality (Appendix D Table D1), which also impacts chemical doses and environmental impacts, a comprehensive set of source water scenarios were evaluated to compare biofiltration and conventional filtration. Figure 6.3 shows the results for 45,000 unique source waters at the national average temperature, 15°C. These scenarios were sorted by the enhanced coagulation treatment bins, with bins for 4 to 8 mg/L TOC split into two sub-plots, a moderate (4 to 6 mg/L) and high (6 to 8 mg/L), for better resolution. Bins for TOC greater than 8 mg/L followed the same trends as the 6 to 8 mg/L bins (Appendix D Figure D5) but represented fewer than 2% of applicable water treatment plants. The global warming impact category was representative of all the TRACI categories (Appendix D Figure D4). It was found to have the same trends for all source water scenarios even when compared to the most dissimilar carcinogenic impacts category (Appendix D Figures D6). Overall, biofiltration had the same or better environmental performance than conventional filtration for almost all source water scenarios (Figure 6.3). The source water conditions that resulted in the best environmental performance for biofiltration depended on a variety of water quality parameters and combinations. As seen before, chemicals were the main driver of the environmental impact trends; specifically, alum had the largest influence since it was related to TOC removal and impacted the required caustic soda dose.

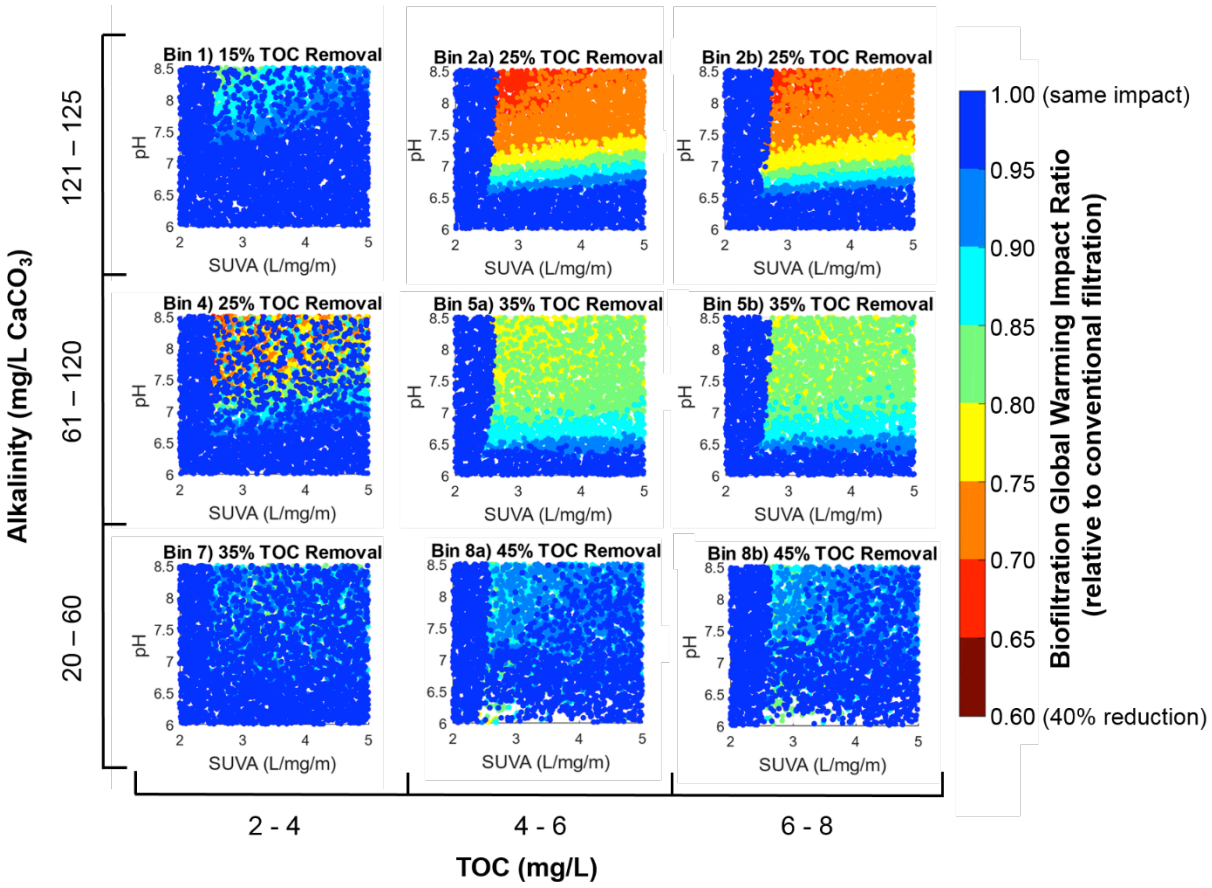

Figure 6.3 Biofiltration global warming impact ratio, relative to conventional filtration, for 45,000 unique source water scenarios at 15°C (12% biofilter TOC removal). The impact ratio color scale ranges from 1.0 to 0.6, and lower ratio values indicating lower impacts and better environmental performance for biofiltration. Bins are numbered to represent the enhanced coagulation bins, which have a low, moderate, and high range for alkalinity and TOC.

6.3.2.1 Influent TOC

As expected, higher influent TOC concentrations required larger alum doses. It was the difference in the alum doses, and therefore also caustic soda doses, between biofiltration and conventional filtration that dominated the relative environmental impact trends. This trend was most clear when influent TOC was between 4 and 8 mg/L (Figure 6.3: bins 2, 5, and 8). The large differences in relative environmental impacts between seemingly similar source water scenarios in Figure 6.3, especially for low influent TOC bins (2 to 4 mg/L; bins 1, 4, and 7), was caused by the inability to show all source water quality dimensions. When exact TOC and alkalinity values were indicated on a scatter plot, while each scenario had a pH between 8.5 and

7.3 and SUVA between 5 and 2.7, then there is a clear environmental trend associated with TOC values (Figure 6.4). Biofiltration had better, relative environmental performance when the source water had greater than 2.5 mg/L TOC (Figures 6.3 and 6.4).

At very low TOC values (< 2 mg/L), the turbidity requirement controlled the alum dose because the enhanced coagulation compliance criteria was already met, so biofiltration and conventional filtration used the minimum allowable alum dose. For very high TOC values (> 8 mg/L), the relative benefit of biofiltration was minimized because the percent difference in alum dose decreased with increasing alum doses. For example, for a source water with 3.0 mg/L TOC (and 15°C, 3.1 L/mg/m SUVA, 77 mg/L $CaCO_3$, 7.6 pH), the conventional filtration alum dose was 45% higher alum dose than the biofiltration dose of 18 mg/L. For the same source water scenario but with 6.0 mg/L TOC, the conventional filtration alum dose was only 35% higher than biofiltration dose of 33 mg/L. Overall, in terms of relative environmental impacts, biofiltration had the best performance when source water TOC values were 2.5 to 8.0 mg/L (Figures 6.3 and 6.4). It is expected that the majority of water treatment plants will have an influent TOC value in this range since the U.S. average surface water TOC is 3.2 mg/L (McGuire et al., 2002).

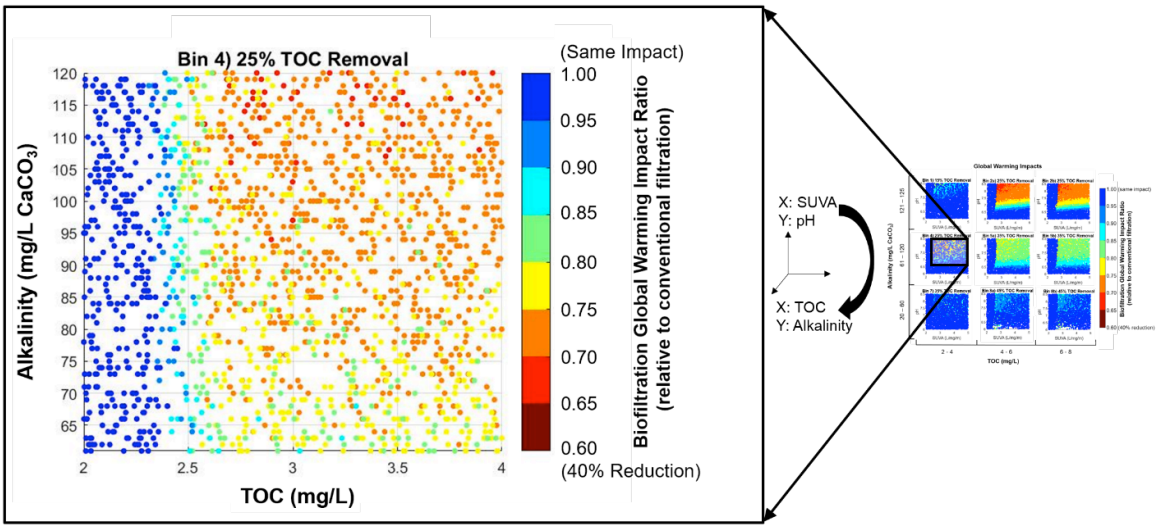

Figure 6.4 Biofiltration global warming impact ratio, relative to conventional filtration, for the enhanced coagulation bin 4. Data (from same as Figure 6.3) was shown as a function of alkalinity and TOC and only included bin 4 scenarios that had a pH between 8.5 and 7.3 and SUVA between 5 and 2.7. The impact ratio color scale ranges from 1.0 to 0.6, and lower ratio values indicate lower impacts and better environmental performance.

6.3.2.2 Influent Alkalinity

Due to the impact of alkalinity on pH changes, the relative environmental impact trends associated with alkalinity were mostly based on differences in caustic soda doses. Biofiltration had similar or slightly worse environmental performance than conventional filtration for lower alkalinity influent waters (0 to 60 mg/l) because of high caustic doses for biofiltration even though it required lower alum doses (Appendix D Figure D7). For these waters, the required percent TOC removal was highest (35% to 50%), so the conventional filtration's alum dose was responsively high. For many of these scenarios, the alum dose was so high that all alkalinity was consumed, and a low caustic soda dose was able to adjust the pH of the final water. On the other hand, the biofiltration alum dose was not high enough to consume all alkalinity, so the caustic dose was substantially higher to account for the remaining buffering capacity. However, source waters in the high alkalinity treatment bins required similar caustic doses for both biofiltration and conventional filtration due to the waters' high buffering capacities. For these scenarios,

though, biofiltration had better relative environmental performance because it was able to meet the lower TOC requirements mostly through biodegradation and have considerably smaller alum doses than conventional filtration. Therefore, biofiltration had the best relative environmental performance when the source water had alkalinity values above 60 mg/L $CaCO_3$, in particular at very high alkalinities (≥121 mg/L as $CaCO_3$) (Figures 6.3 and 6.4). It's also expected that the majority of water treatment plants will have an influent alkalinity value in this range since the U.S. average surface water alkalinity is 77 mg/L as $CaCO_3$ (McGuire et al., 2002).

6.3.2.3 Influent SUVA

In general, a low SUVA indicates more non-aromatic compounds, which are harder to coagulate and require larger alum doses (White et al., 1997). Since it has been found to be too costly and difficult to coagulate at very low SUVA values, the enhanced coagulation requirement can be satisfied by reducing the SUVA to 2 L/mg/m. This compliance criteria required the lowest alum dose compared to the other criteria when the source water SUVA was less than 2.75 L/mg/m. Therefore, for these source water conditions, conventional and biofiltration had the same alum doses and environmental impacts because the goal of coagulation was only SUVA reduction; also, it was conservatively assumed that biofiltration did not change the water's SUVA, although some studies have found reductions in SUVA when using sorbable media in the biofilter (Peleato et al., 2017). At SUVA values of 2.75 L/mg/m and higher, the smallest alum dose was instead associated with the percent TOC removal compliance criteria. So, the biofiltration had a lower relative alum dose due to TOC biodegradation, resulting in better environmental performance than conventional filtration. For surface waters, the U.S. average annual SUVA value is 3.1 L/mg/m (McGuire et al., 2002), which corresponds with this better biofiltration environmental performance.

6.3.2.4 Influent pH

At low pH values, less alum was needed because coagulation was more effective (Edwards, 1997). Due to this effectiveness, both the biofiltration and conventional filtration alum doses for enhanced coagulation were usually so small that the minimum allowable alum dose had to be used instead. At high pH, coagulation was more difficult to achieve, so higher alum doses were needed. For pH values greater than 6.5, biofiltration had better environmental performance because either (i) the biofiltration dose was the minimum allowable alum dose while the conventional filtration alum dose increased with higher pH values (Appendix D Figure D8); or (ii) the water quality required that both doses be higher than the minimum allowable alum dose, but the biofiltration dose was always less due to biological TOC removal (Appendix D Figure D9). The best environmental performance for biofiltration was seen at SUVA values around 2.75 L/mg/m coupled with high these pH values. It's expected that the majority of water treatment plants will have a source water in this pH range since the U.S. average surface water pH is 7.5 (McGuire et al., 2002).

Due to the influence of the minimum allowable alum dose on these results, the impact of this uncertainty parameter was evaluated. U.S. water treatment plant data, before 1994 and the common practice of enhanced coagulation, shows that a minimum dose for turbidity could range from 11 to 30 mg/L alum, with 18 mg/L being the median and most-likely value (Appendix D Table D12) (Randtke et al., 1994). Compared to results using the median value, when the minimum allowable dose was set to 11 mg/L, biofiltration performed even better and achieved up to 40% reduction in global warming impacts compared to conventional filtration (Appendix D Figure D10). This improved performance was true even at lower pH values because the biofiltration alum dose required for enhanced coagulation was no longer below the minimum allowable alum dose, and the biological TOC removal benefits were realized. When the

minimum allowable dose was set to 30 mg/L, the biodegradation benefit was masked even more by this larger minimum allowable dose as it was used instead of the relatively lower dose needed for enhanced coagulation. So biofiltration more often had a dose that was similar to the conventional filtration's dose (Appendix D Figure D11). Therefore, since a model to determine alum doses for turbidity does not currently exist, the minimum allowable dose needs to be experimentally determined for each source water to more accurately determine the relative environmental impacts.

6.3.2.5 Biofilter Performance

One of the largest impacts on biofilter percent TOC removal is temperature (Fu et al., 2017; Terry and Summers, 2017; Urfer et al., 1997). Figures 6.2 to 6.4 represented 12% TOC removal in a biofilter, which was typical of water temperatures between 10°C and 20°C. At temperatures greater than 20°C, 15% TOC removal was expected and biofiltration had even better environmental performance (Appendix D Figure D12). The opposite was also true. The environmental benefits of biofiltration decreased with decreasing temperatures, and respective percent TOC removal (Appendix D Figure D13). In general, an increase in temperature was found to improve biofilter percent removal, but temperature can change over time, even seasonally. Also, there were large ranges of percent TOC removal observed experimentally observed for a given temperature range; for example, a range of 5% to 20% TOC removal has been experimentally observed for water temperatures between 10°C and 20°C. This was because data was collected from full scale, to bench scale experiments and for a range of empty bed contact times. The empty bed contact time can be adjusted to maintain TOC removal; for example, during colder months, TOC removal may slightly decrease unless the empty bed contact time is extended. However, increasing the empty bed contact time may increase the environmental impacts, specifically the impacts associated with the production of filter media

and steel for the housing. Therefore, more full scale biofilter TOC removal performance data is needed, especially over temperature changes, to more accurately estimate environmental benefits.

6.3.3 Biofiltration Operational Considerations

The decision to use or to transition from conventional filtration to biofiltration, by adding chlorine only after the filter, requires several important considerations, including cost, water quality, and operational complexity. Also, even greater relative benefits are expected because the LCA model used conservative assumptions. For example, disinfection CT credit was given in the conventional filter, which is not expected for most drinking water plants, to underestimate relative biofilter benefits by requiring larger disinfection infrastructure. In terms of final drinking water quality, this LCA focused on the biofiltration benefit of TOC removal. However, many full scale and pilot scale studies have shown additional advantages of biofiltration, including the ability to: decrease microbial regrowth potential in distribution systems (Urfer et al., 1997), minimize residual disinfectant demand (LeChevallier, 2014; Pharand et al., 2014), improve the control of DBP precursors and DBP formation (LeChevallier et al., 1992), and remove organic contaminants (Hallé, 2009; Urfer et al., 1997). In particular, drinking water biofilters have been shown to biodegrade algal metabolites (Zearley and Summers, 2012), which cause taste and odor issues, such as geosmin (Elhadi et al., 2006; Lauderdale et al., 2012; McDowall, B., Ho, L., Saint, C.P. & Newcombe, 2007; Nerenberg et al., 2000; Westerhoff et al., 2005) and microsystins (Ho et al., 2006), as well as specific organic contaminants such as pesticides (Meffe et al., 2010; Zearley, 2012), pharmaceuticals (Zuehlke et al., 2007) and personal care products, which can be endocrine disrupting compounds (Halle et al., 2009; Snyder et al., 2007).

Overall, many source water qualities resulted in similar environmental performance between biofiltration and conventional filtration. For these circumstances, conventional filtration may be considered more advantageous because of perceived biofiltration limitations, which include additional operator training, increased operational complexity, lack of technology specific regulations, and decreased reliability. Despite these possible limitations, the majority of drinking water treatment plants are expected to have source waters that would result in significant biofiltration benefits. In general, biofiltration had considerably lower environmental impacts and costs than conventional filtration when a source water had: TOC > 2.5 mg/L, alkalinity > 60 mg/L $CaCO_3$, SUVA > 2.75 L/mg/m, and pH > 6.5. In addition, if water quality continues to degrade, as expected due to the use of alternative source waters and climate change (Delpla et al., 2009; Raseman et al., 2017; Todd et al., 2012; Vorosmarty et al., 2000), then it is even more important to consider biofiltration. With this new LCA model and data, water treatment plants will be able to decide if and when they can employ biofiltration to reduce environmental impacts and costs while maintaining, or improving, drinking water quality.

6.4 Conclusions

This comparative LCA of conventional filtration and biofiltration for drinking water treatment found biofiltration had lower environmental impacts than conventional filtration for average U.S. source waters by about 25%. Even under uncertainty, these trends were the same. While the simple and practical transition from conventional filtration to biofiltration can achieve these environmental benefits and result in the same drinking water quality, it can have barriers to implementation, especially due to perception issues. Chemicals, in particular alum and caustic soda, had the largest contributions to environmental impacts. The most effective way to substantially decrease negative environmental impacts of either filtration system is to optimize

chemical doses. Higher temperatures can support greater TOC biodegradation, which increases the environmental benefits of biofiltration. Higher levels of biodegradation can also be achieved at lower temperatures when biofilter parameters are optimized. Benefits of biofiltration increased with deteriorating source water quality; specifically, biofiltration had better environmental performance when a source water had TOC > 2.5 mg/L, alkalinity > 60 mg/L as $CaCO_3$, SUVA > 2.75 L/mg/m, and pH > 6.5. Source waters are expected to continue to deteriorate in quality over time, which increases the opportunity for biofiltration to reduce the environmental impacts and costs of drinking water treatment.

Chapter 7
Conclusions

7.1 Remarks

The foremost objective of this effort was to evaluate the efficacy of biological filtration to remove biodegradable organic matter, BOM, from surface water. The research focused on how operating parameters and water quality parameters impact biofilter removal efficacies. Chapter 1 summarized the literature and through a meta- analysis provided insight into operating and water quality parameter impacts. Chapter 2 developed a novel scale up method to simulate pilot flow through systems at the bench scale. Chapter 3 developed a predictive model to determine DOC removal based on operating and water quality parameters. Chapter 4 evaluated biofilter performance based on EBCT, temperature, and influent DOC, as well as biomass activity acclimation and distribution at the pilot scale. Chapter 5 focused on biofilter performance based on EBCT, temperature, influent DOC, biomass activity, NOM origin, and disinfection by-product (DBP) precursor removals of three different source waters at the bench scale. Chapter 6 investigated a LCA comparison of biofiltration and conventional filtration in terms of environmental impacts.

Overall, specific conclusions are provided at the end of each respective chapter; to avoid redundancy, conclusions here are discussed in context of the hypotheses developed in Chapter 1, that largely steered efforts throughout this research.

7.2 Hypotheses

Hypothesis 1: Extended EBCT will lead to higher removal efficacy of slowly degrading organic contaminants and organic matter. DOC removal increased as EBCTs extended from 5

minutes to 15 minutes to 30 minutes over multiple source waters and experimental conditions (Chapter 3,4,5). Thus, longer exposure time allowed for more degradation of DOC.

Hypothesis 2: Larger accumulation of biomass will lead to more degradation and removal of contaminants. Biofilters operating at steady-state for biomass activity achieved higher DOC removals than biofilters that had not reached acclimation or steady-state (Chapter 4). For the same source water, higher levels of biomass activity were congruent with higher levels of DOC removal (Chapter 4, 5). However, this congruent relationship did not hold true across different NOM matrix source waters, as different amounts of biomass activity levels were associated with different NOM matrixes, yet similar DOC removals were seen across the different NOM matrixes. (Chapter 5).

Hypothesis 3: Increasing temperatures will yield increasing removal of contaminants in biological filtration. The impact of temperature was evaluated in two parts:

Hypothesis 3a: Increasing temperatures result in increasing biomass activity within the filter. ATP biomass activity was a function of temperature and higher ATP activity levels were associated with higher temperatures. In the temperature range of 5°C to 28°C, biomass activity levels increased with increasing temperature and were statistically different (Chapter 3, 4, 5).

Hypothesis 3b: Increasing temperatures lead to increasing degradation rate constants of contaminants. Contaminant rate constant, *k″,* is a contaminant utilization rate constant per biomass concentration per time. Phospholipid biomass concentrations were not impacted by temperature in the range of 5°C to 22°C, as biomass concentrations were not statistically different. Therefore, k″ did increase with increasing temperature when phospholipids were employed as the biomass measurement (Chapter 3). ATP biomass activity is a function of temperature, so k″ did not increase with increasing temperature when ATP was employed as the biomass measurement, as temperature change was incorporated in the ATP activity measurement (Chapter 3).

Hypothesis 4: Biofiltration performance of BOM removal can be modeled using Monod kinetics when BDOC of the source water, total biomass of the filter and a contaminant utilization rate constant are known. Chapter 3 results developed a kinetic modeling approach, based on Monod kinetics, to successfully model biofiltration performance. Biomass distribution decreases throughout the filter at an exponential rate and a correlation was developed between biomass at the top of the filter and total biomass of the filter when EBCT is known. In addition, a correlation between temperature and ATP biomass activity was developed that allows a robust predictive model, developed by literature data, to be applicable at specific utilities with varying temperatures and EBCTs (Chapter 4, 5).

Another objective of this work was to complete a comparative LCA evaluating conventional filtration and biological filtration to help facilitate the design of a more sustainable drinking water treatment plant. Chapter 6 results concluded biofiltration had lower environmental impacts than conventional filtration for average U.S. source waters by about 25%. Even under uncertainty, these trends were the same. Chemicals, in particular alum and caustic soda, had the largest contributions to environmental impacts. The most effective way to substantially decrease negative environmental impacts of either filtration system is to optimize chemical doses. Higher temperatures can support greater TOC biodegradation, which increased the environmental benefits of biofiltration. Higher levels of biodegradation can also be achieved at lower temperatures when biofilter parameters are optimized. Benefits of biofiltration increased with deteriorating source water quality. Source waters are expected to continue to deteriorate in quality over time, which increases the opportunity for biofiltration to reduce the environmental impacts and costs of drinking water treatment plants (Chapter 6).

Extended biofiltration will improve the sustainability of drinking water utilities by providing additional DOC removal. Plant operations should be optimized to receive the most organics removal from the biofilter throughout different seasonal variations. Albeit, in some situations it would not be a practical operation to extend EBCT, i.e. filter run time constrained or problematic headloss. If utilities decide to convert a conventional filter to an extended EBCT biofilter by lowering the hydraulic loading rate and moving the chlorine disinfection point after the filter, biomass will begin to develop immediately. Once biomass is present, it will take about eight weeks to reach acclimation or steady state under similar water quality scenarios as this study. ATP, an indicator of activity, is a great gage for "health" of the biofilter and media samples should be taken three inches below the top of the filter. Normal filter monitoring and maintenance should continue with biofilters in addition to activity measurements. The biomass distribution profile allows utilities to predict DOC removal based on biomass activity at the top of the filter. If a biomass sample at the top of the filter is unattainable, influent TOC and influent temperature can predict top of filter ATP. Prediction of biofilter TOC removal will give utilities the unique opportunity to optimize plant operations, which will help reduce environmental impacts.

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Appendix A
Biodegradable Organic Matter and Biofilter Performance: A Review

Table A1. AOC data for nonozonated surface waters.

Water	TOC (mg/L)	AOC (µg/L)	AOC/TOC (%)	Reference
Reservoir	2.83	62	2	Volk et al., 2000
Reservoir	12.28	112	1	
River	6.89	482	7	
River	7.91	133	2	
River	2.69	152	6	
Creek	2.7	242	9	
River	16.27	120	1	
River	2.75	113	4	
Reservoir	3.39	138	4	
River	5.35	302	6	
River	5.7	16	0.3	Van der Kooij et al., 1989
River	5.7	12	0.28	
raw water	5.5	300	5	Liao et al., 2017
Shallow Aquifer with significant surface influence and salt water intrusion	11	83	1	Escobar and Randall, 1999a
Shallow Aquifer with significant surface influence and salt water intrusion	10	142	1	
River	7	400	6	Dugan et al., 1997
River, Lake, Reservoir (n=79)	1.99	139	23	Kaplan et al., 1994
River, Lake, Reservoir (n=79)	2	150	9	
River, Lake, Reservoir (n=79)	3.12	55	2	
River, Lake, Reservoir (n=79)	3.48	253	7	
River, Lake, Reservoir (n=79)	3.09	191	6	
River, Lake, Reservoir (n=79)	3.12	76	2	
River	3.5	30	1	Krasner et al., 1992
Lake	4.29	44	1	Persson et al., 2006
Lake	1.27	399	31	Miltner et al., 1996
Lake	0.53	203	38	
River	2.6	14	0.5	Van der Kooij et al., 1992
River	1.9	25	1.3	
River	2.2	6.3	0.3	
River	4.4	260	6	Rachwal et al.,

River	5.1	100	2	1992
River	6.3	40	1	Mitton et al., 1993
River	3	44	1	
River	3	75	3	
River	10.5	400	4	
River	2.75	158	6	
River	4.5	55	1	
River	4.4	157	4	Amy et al., 1992
Delta	4	175	4.4	Daniel et al., 1995
River	7.2	15	0.2	Van der Kooij et al., 1984
River	4	33	0.8	
River	3	12	0.4	
River	5.7	16	0.28	
River	4.3	12	0.28	
Reservoir	4.31	222	5.15	Reasoner et al., 1990
Reservoir	3.71	146	3.94	
Reservoir	3.71	234	6.31	
Reservoir	4.06	246	6.06	
Reservoir	3.7	322	8.70	
Reservoir	3.78	176	4.66	
Reservoir	4.09	224	5.48	
Reservoir	3.68	229	6.22	
Reservoir	3.51	246	7.01	
Reservoir	3.33	261	5.39	
Reservoir	2.8	190	6.79	Bradford et al., 1994
Reservoir	2.8	125	4.46	
Reservoir	2.8	200	7.14	
Reservoir	2.8	275	9.82	
Reservoir	2.8	185	6.61	
Reservoir	2.8	200	7.14	
Surface Water	3	25	0.83	Anderson et al., 1990
Surface Water	5	40	0.80	
Surface Water	3	40	1.33	
Surface Water	2.8	24	0.86	
Surface Water	2.2	10	0.45	
Surface Water	2.1	25	1.19	
Surface Water	2	30	1.50	
Surface Water	3.5	75	2.14	

Surface Water	11	225	2.05	
Surface Water	7.5	225	3.00	
Surface Water	5	75	1.50	
Surface Water	6	125	2.08	
River	3	55	2	Reckhow et al., 1992
River	3.9	350	9	LeChevallier et al., 1991b
Shallow Aquifer with significant surface influence and salt water intrusion	1.2	70	6	Escobar and Randall, 1999a
River	4	70	2	van der Kooij et al., 1990
River	3.9	71	2	
River	3.1	98	3	
River	3.8	105	3	
River	5.8	110	2	
Surface Water	3.0	299	10	Liu et al., 2002
Surface Water	3.81	275	7	
Surface Water	4.55	263	6	
Surface Water	2.5	250	10	Zhang et al., 2017
River	3	75	3	Gramith et al., 1991
River	2.7	75	3	
Lake	4.09	27.3	1	Charnock and Kjonno, 2000
River	3.2	75	2	Coffey et al., 1995

Table A2. AOC data for ozonated surface waters. In some cases, the ozone dose was reported, while in other cases the ozone / TOC ratio was reported and the ozone dose calculated.

Water	Ozonation Dose (mg/L)	TOC (mg/L)	AOC (µg/L)	AOC/TOC (%)	Reference
River	1.6	3.2	200	6	Siddiqui et al., 1997
River	3.2	3.2	300	9	
River	5	3.3	650	20	
River	6.4	3.3	1150	35	
Lake	0.5	5.5	436	9	Liao et al., 2017
River	1.6	3.2	300	9	Van der Kooij et al., 1989
River	3.2	3.2	400	13	
River	5	3.2	410	12	

River	6.4	3.3	500	15	
River	3	5.5	134	2.4	
River	1.4	1.46	35	2.4	
River	2 - 3.5	7.1	120	1.7	Van der Kooij et al., 1984
River	2 - 3.5	3.8	106	3	
River	2 - 3.5	2.9	91	3	
River	1.6	3.2	450	14	Kang and Kim, 1993
River	3.2	3.2	220	7	
River	5	3.3	550	17	
River	6.4	3.3	400	12	
River	1.8	3.5	585	17	Krasner et al., 1992
River	0.7	2.26	69	3	Swertfeger et al., 1993
River	0.7	2.39	69	3	
River	0.7	2.29	99	4	
River	1.2	3.5	84	2	Wang et al., 1995
River	1.2	3.5	84	2	
Surface Water	1.0		34		Mitton et al., 1993
Surface Water	1.0		95.5		
Surface Water	1.0		444		
Surface Water	1.5		450		
Surface Water	1.0		27.5		
River	0.36	1.42	100	7	Miltner et al., 1992
River	0.71	1.42	125	9	
River	1.07	1.42	125	9	
River	2.13	1.42	150	11	
River	3.2	1.42	180	13	
River	1.0	3.6	240	7	Rachwal et al., 1992
River	1.0	4.7	195	4	
River	1.0	4.5	185	4	
River	N/A		320		Gramith et al., 1991
River	N/A		430		
Delta	3.0		679		Daniel et al., 1995
River	1.6		405		Coffey, et al., 1995
Reservoir	1.6	4.4	207	5	Reasoner et al., 1990
Reservoir	1.6	4.07	98	2	
Reservoir	1.6	3.8	239	6	

Reservoir	1.6	3.73	266	7	
Reservoir	1.6	3.55	332	9	
Reservoir	1.6	4.42	317	7	
Reservoir	1.6	3.66	173	5	
Reservoir	1.6	3.92	232	6	
Reservoir	1.6	3.88	229	6	
Reservoir	1.6	3.12	241	8	
Surface Water	1		110		McEnroe et al., 1992
Surface Water	2		110		
Surface Water	3		170		
Surface Water	1		140		
Surface Water	2		100		
Surface Water	3		120		
Surface Water	7	2.9	550	19	Bradford et al., 1994
Surface Water	7	1.39	200	14	
Surface Water	7	0.66	300	45	
Reservoir	7	2.8	350	13	
Reservoir	7	2.8	190	7	
Reservoir	7	2.8	310	11	
Reservoir	7	2.8	400	14	
Reservoir	7	2.8	300	11	
Reservoir	7	2.8	400	14	
Surface Water	4	2.5	600	24	Zhang et al., 2017
Shallow Aquifer with significant surface influence and salt water intrusion	4.7	1	80	8	Escobar and Randall, 1999b
River	2.7	2.9	800	28	LeChevallier et al., 1991b
River	1.25		115		Reckhow et al., 1992
River	3.4	4.8	841	18	Amy et al., 1992
Lake	5.6	2.94	1300	44	Miltner et al., 1996

Table A3. BDOC data for nonozonated surface waters.

Water	TOC (mg/L)	BDOC (mg/L)	BDOC/TOC (%)	Reference
Lake	6	1.25	21	In-House
Lake	4.53	0.96	21	

Lake	4.5	1.03	23	
Lake	3.2	0.67	21	
Reservoir	3.6	0.7	19	
Reservoir	3.66	0.69	19	
Reservoir	3.48	0.72	19	
Reservoir	2.3	0.45	20	
Creek	6.65	1.02	15	
Creek	4.88	0.75	15	
Creek	4.89	0.62	13	
Creek	6.65	0.67	10	
River	6.8	1.35	20	Ribas et al., 1992
River	4.39	0.48	11	
River	3.2	0.6	19	Solarik et al., 1996
River	7.5	1.95	26	Vasyukova et al., 2014
River	2.6	0.42	16	
River	4.8	0.82	17	
Lake	1.4	0.17	12	So et al., 2017
Lake	5.5	1.4	25	Liao et al., 2017
Lake	4.09	0.91	22	Charnock and Kjonno, 2000
River	2.78	0.55	20	Allgeier et al., 1996
River	5.45	2	36	Frias et al., 1992
River	7.22	2	22	
River	7	1.5	20	Ribas et al., 1992
River	7	1.47	20	
River	5.06	0.98	19	
River	5.06	1.4	26	
River	4.82	0.95	19	
River	4.82	1.29	26	
River	3.5	1.2	34	Servais et al., 1987
River	3.6	0.7	19	
River	4.94	1.18	24	
River	8.91	3.07	34	
River	8.86	1.51	17	
River	11.93	4.89	41	
River	13.13	7.8	59	
Reservoir	2.83	0.87	31	Volk et al., 2000
Reservoir	12.28	2.44	20	

River	6.89	1.75	25	
River	7.91	1.96	25	
River	2.69	0.71	26	
Creek	2.7	0.73	27	
River	16.27	2.14	13	
River	2.75	0.39	14	
Reservoir	3.39	0.7	21	
River	5.35	1.08	20	
River	3.04	1.22	40	Joret et al., 1991
River	3.02	1.22	40	
River	3.02	1.13	37	
River	3.05	1.1	36	
River	3.09	1.35	44	
River	2.98	2.15	72	Joret et al., 1988
River	3.2	2.1	36	
River	2.9	1	34	
River	4.5	1.8	40	
Creek	4.52	1.31	29	Volk et al., 1997
Shallow Aquifer with significant surface influence and salt water intrusion	11.3	2.4	20	Escobar and Randall, 1999a
Shallow Aquifer with significant surface influence and salt water intrusion	10.0	1.3	12	Escobar and Randall, 1999a
Creek	0.23	2.84	8	Volk et al., 1997
Creek	1.72	0.50	29	Kaplan and Newbold, 1995
Creek	2.98	0.78	26	
River, Lake, Reservoir	1.99	0.24	27	Kaplan et al., 1994
River, Lake, Reservoir	2.27	0.3	12	
River, Lake, Reservoir	3.12	0.25	1	
River, Lake, Reservoir	3.48	0.29	8	
River, Lake, Reservoir	3.09	0.14	5	
River, Lake, Reservoir	3.12	0.39	13	
River	3	0.73	28	Kaplan et al., 1994
Lake	3.83	0.52	13.5	Sondergaard and Worm, 2001
Creek	6	3.92	34	Trulleyova and Rulik, 2004
Creek	6.1	3.56	40	
Creek	6	3.42	43	
River	2.8	0.28	10	Treguer et al., 2010

River	2.8	0.48	17	Summers, 1993
River	2.8	0.56	20	
River	4.14	0.95	23	
Lake	4.29	1.06	25	Persson et al., 2006
Lake	2.7	0.24	9	Cipparone et al., 1997
River	2.5	0.2	8	Swertfeger et al., 1993
Lake	5.83	1.27	22	Miltner et al., 1996
Lake	2.77	0.53	19	
River	2.6	0.47	18	Mogren et al., 1990
River	2.71	0.32	12	
River	2.96	0.45	15	
Reservoir	2.8	0.28	10	Fonseca et al., 2003
Shallow Aquifer with significant surface influence and salt water intrusion	1.2	0.11	9	Escobar and Randall, 1999b
River	3	1	33	Chaiket et al., 1999
River	2.7	0.2	7	
Source Water	2.5	1.5	60	Zhang et al., 2017
River	2.46	0.49	20	Kaplan et al., 1993
River	11.6	3.24	28	
Lake	2.7	0.23	8.5	Speitel et al., 1992
River	4.4	0.46	10.5	Amy et al., 1992
River	6.8	0.15	2.2	Cushing et al., 1996
River	2.5	0.35	14	Morisette et al., 1995
River	2.27	0.15	6.6	
Reservoir	3	0.6	20	Schneider et al., 1998
River	6.3	1.15	18	Yavich et al., 2004
Lake	10.1	5.04	50	

Table A4. BDOC data for ozonated surface waters. In some cases, the ozone dose was reported, while in other cases the ozone / TOC ratio was reported and the ozone dose calculated.

Water	Ozonation Dose (mg/L)	TOC (mg/L)	BDOC (mg/L)	BDOC/TOC (%)	Reference
Lake	0.5	2.4	0.82	34	Cipparone et al., 1997
Lake	1	2.4	0.41	17	

Lake	2	2.4	0.91	38	
Lake	3	2.4	0.72	30	
Lake	5	2.4	1.22	51	
River	3	2.65	0.75	28	Wricke et al., 1996
Reservoir	2.1	3.6	1.08	30	Carlson et al., 1996
Lake	N/A	1	0.14	14	So et al., 2017
Lake	0.5	5.5	2.2	40	Liao et al., 2017
River	0.83	3.3	0.8	24	Siddiqui et al., 1997
River	1.65	3.3	1.4	42	
River	5.6	5.6	2.5	45	
River	4.95	3.3	1.4	42	
River	6.8	3.4	1.6	47	
River	5.6	5.6	2.4	43	
River	5.6	5.6	2.4	43	
River	N/A	2.05	0.74	36	Summers, 1993
River	N/A	2.05	0.74	36	
River	N/A	2.05	0.51	25	
River	2.56	2.9	1.2	41	Solarik et al., 1996
River	2.56	2.9	1.2	41	
River	1.04	2	0.4	20	
River	1.58	2.8	0.3	11	
River	1.31	10.5	1.1	10	
River	1.03	9.8	0.8	8	
River	0.4	2.8	0.4	14	Treguer et al., 2010
River	0.6	2.9	0.6	21	
River	1	2.8	0.65	23	
River	1.2	2.7	0.6	22	
River	0.5	6.1	2.3	38	Yavich et al., 2004
River	0.75	6	2.55	43	
River	1	6	2.78	46	
Lake	0.75	9.5	6.06	46	
Lake	1.5	9.6	6.86	46	
Lake	3	9.5	7.39	46	
Lake	N/A	5.5	2.43	44	Allgeier et al., 1996
River	N/A	2.05	0.64	31	
River	N/A	6.95	1.41	20	
River	0.4 Residual	3.19	0.6	19	Niquette et al., 1998
River	0.4 Residual	3.28	0.7	21	
River	0.4 Residual	3.2	0.7	22	
River	0.4 Residual	3.25	0.8	25	
River	0.4 Residual	2.9	0.5	17	
River	0.7	2.39	0.43	18	Swertfeger et al., 1993
River	0.7	2.26	0.62	27	
River	0.7	2.29	0.43	19	

River	0.8	2.2	0.70	32	Wang et al., 1995
River	0.8	2.2	0.44	20	
River	6.2	3.09	1.16	38	Mogren et al., 1990
Creek	2	2.84	0.6	21	Volk et al., 1997
Creek	3.5	2.84	0.8	28	
Creek	5	2.84	0.85	30	
Creek	6.5	2.84	0.85	30	
River	N/A	3.1	1.17	38	Volk et al., 1994
River	N/A	3.1	0.6	19	
River	N/A	3.1	0.43	14	
River	N/A	3.1	0.47	15	
River	N/A	1.8	0.72	40	Volk et al., 1994
River	N/A	1.8	0.6	33	
River	N/A	1.8	0.45	25	
River	N/A	1.8	0.47	26	
River	0.36	1.42	0.15	11	Miltner et al., 1992
River	0.71	1.42	0.15	11	
River	1.07	1.42	0.2	14	
River	2.13	1.42	0.2	14	
River	3.2	1.42	0.3	21	
River	0.36	1.42	0.4	28	
Lake	1	2.3	0.69	30	Speitel et al., 1992
Lake	3	2.3	0.81	35	
Lake	5	2.3	1.13	49	
Lake	3	3.4	1.36	40	
Lake	5	3.4	1.7	50	
Reservoir	1.5	3	1.2	40.0	Schneider et al., 1998
River	3.4	4.8	1.42	29.5	Amy et al., 1992
River	8.0	2.75	0.5	18.2	Cushing et al., 1996
River	0.4	3.2	0.63	19.7	Prevost et al., 1995
River	0.4	3.2	0.7	21.9	
River	0.4	3.2	0.7	21.9	
Reservoir	1.2	2.5	0.83	33.2	Fonseca et al., 2003
Reservoir	0.77	2.3	0.67	29.1	
Lake	3.5			23	LeCourt et al., 1997
Lake	7.0			30	
Lake	14			35	
Lake	1.8	2.5	1.8	32	
Shallow Aquifer with significant surface influence and salt water intrusion	3.2	2	0.8	40	Escobar and Randall, 1999b

Shallow Aquifer with significant surface influence and salt water intrusion	4.7	1	0.14	13.5	Escobar et al., 2001
River	N/A	2.1	1.4	35	Joret et al., 1988
River	N/A	2.1	1.4	35	
River	N/A	3.25	1.5	43	
River	1.0	3.4	0.9	26	Chaiket et al., 1999
River	0.73	3.2	1.1	34	
River	1.0	3.2	1	31	
River	2.8	4.5	1	22	
River	2.4	3	0.5	17	
River	2.5	5	1.75	35	
River	3	3.2	1.3	41	
Source Water	4	2.9	1.8	62	Zhang et al., 2017
Lake	5.6	2.74	1.21	44	Miltner et al., 1996
River	0.8	2.28	0.39	17	Summers et al., 1998
River	N/A	2.05	0.74	36	Summers, 1993
River	N/A	2.05	0.51	25	
River	N/A	2.06	0.62	30	Servais et al., 1991
River	1	3.64	1.53	42	Moll and Summers, 1996
River	N/A	3.68	0.76	20	Ribas et al., 1992
River	0.4 residual	2.5	0.3	12	Morissette et al., 1995
River	0.4 residual	2.27	0.2	9	

Table A5. AOC and BDOC data for nonozonated groundwater.

TOC (mg/L)	BDOC (mg/l)	BDOC/TOC (%)	AOC (µg/L)	AOC/TOC (%)	Reference
1.2	0.14	19			Escobar et al., 2001
1.2			69	6	Escobar et al., 2001
7.6			13	0.2	Van der Kooij et al., 1992
2.3			4.5	0.2	
1.8			4.2	0.2	
0.3			1.9	0.6	
1.8			7.5	0.4	
3.2	0.16	5			Rittman et al., 2002
2.19			51	2	Bradford et al., 1994

2.9			92	3	
1.39			56	4	
0.66			34	5	
1.1			50	5	
1.1			50	5	
1.1			75	7	
3.5			50	1	
0.9			75	8	
0.9			80	9	
0.9			50	6	
0.9			50	6	
0.9			24	3	
0.9			55	6	
0.9			50	6	
0.9			100	11	
2.6			25	1	
2.6			100	4	
2.6			55	2	
2.6			34	1	
1.1			50	5	
1.1			100	9	
1.1			30	3	
7			300	4.29	
11	349	3			Noble et al., 1994
11	388	4			
4.14	0.47	11			Allgeier et al., 1996
17.5	0.24	1			
11.7	1.53	13			

Table A6. AOC and BDOC data for ozonated groundwater.

Ozonation Dose (mg/L)	TOC (mg/L)	BDOC (mg/L)	BDOC/TOC (%)	AOC (µg/L)	AOC/TOC (%)	Reference
4.7	1			85	9	Escobar et al., 2001
1	2.9	0.84	29			Rittman et al., 2002
1.4	2.9	0.99	34			
1.8	2.9	1.19	41			
N/A	3.64	1.53	42			Wang and Summers, 1996

7				300		
7				100		
7	2.19			125	6	
7				600		
7	0.98			100	10	
10				350		
10				300		
10				900		
1				75		
7				300		Bradford et al., 1990
7	1.2			150	13	
7	1.2			175	15	
7	1.2			100	8	
7	1.2			85	7	
7	1.2			105	9	
7	1.2			150	13	
7	1.2			90	8	
7	1.2			150	13	Bradford et al., 1994
7	2.6			290	11	
7	2.6			225	9	
7	2.6			200	8	
7	2.6			200	8	
7	1.1			200	18	
7	1.1			400	36	
7	1.1			900	82	

Table A7. Biofilter data for nonozonated surface waters.

Study	Temperature Range (°C)	EBCT (min)	Media	TOC Removal (%)	k′ (min^{-1})	Reference
Pilot	≤10	4	S	10	0.173	Elhadidy, 2016
Pilot	≤10	5	A	3	0.027	In-House
Pilot	10 --18	5	A	4.5	0.051	In-House
Pilot	10 -- 20	6	A/S	17	0.316	In-House
	≤10	7	A	4	0.032	Collins et al., 1993
Pilot	12	7	GAC (exhausted)	9	0.085	Chowdhury et al., 2009
Pilo	12	7	GAC	9	0.085	Chowdhury et al.,

t			(exhausted)			2009
Pilot	12	7	GAC (exhausted)	9	0.085	Chowdhury et al., 2009
Pilot	12	7	A	7	0.062	Chowdhury et al., 2009
Piot	20 -- 25	7	A	13	0.150	Chen et al., 2016
Pilot	≤10	8	A/S	6	0.045	Elhadidy, 2016
Pilot	≥20	8	A/S	12	0.115	Elhadidy, 2016
Pilot	10 -- 20	9	GAC (exhausted)	11	0.089	Klevens et al., 1996
Pilot	1--7	10	A/S	2	0.009	Morissette et al., 1995
Pilot	1 -- 7	10	S	2	0.009	Summers et al., 2006
Pilot	1 -- 7	10	A/S	10	0.069	Morissette et al., 1995
Pilot	1 -- 7	10	S	14	0.120	In-House
Pilot	10 -- 18	10	A/S	12	0.092	Morissette et al., 1995
Pilot	10 -- 18	10	S	12	0.092	Chowdhury et al., 2009
Pilot	16 -- 20	10	GAC (24 months)	14	0.120	In-House
Pilot	10 -- 18	10	A/S	8	0.049	Morissette et al., 1995
Pilot	10 -- 18	10	S	10	0.071	Rahman, 2013
Pilot	10 -- 24	10	A	12.5	0.098	Morissette et al., 1995
Pilot	10 -- 18	10	A/S	8	0.050	Morissette et al., 1995
Pilot	10 -- 18	10	S	12	0.089	Pelato et al., 2016
Pilot	13.7 -- 25.4	10	GAC (>120 months)	8	0.051	Hubele, 1995
Pilot	≤10	15	A	7	0.029	In-House
Bench	≤10	15	A	7	0.029	In-House
Bench	10 -- 20	15	A	8	0.034	In-House
Pilot	10 --19	15	A	10.5	0.050	In-House
Bench	≥20	15	A	10	0.046	In-House
Pilo	≤10	16	A/S	7	0.027	Elhadidy, 2016

t						
Pilot	≥20	16	A/S	15	0.087	Elhadidy, 2016
Pilot	≥20	17	A	14	0.071	Fonseca et al., 2003
Pilot	10 -- 20	17.5	A	7	0.025	Morissette et al., 1995
Pilot	≥20	23	A/S	15	0.060	Elhadidy, 2016
Pilot	≤10	24	A/S	8	0.021	Morissette et al., 1995
Bench	21 -- 22	24	A/S	17	0.079	In-House
Pilot	≤10	30	A	13	0.035	In-House
Bench	≤10	30	A	12	0.031	Wulfeck and Summers, 1994
Bench	10 -- 20	30	A	22	0.177	Wulfeck and Summers, 1994
Pilot	10 -- 20	30	A	18.5	0.086	Morissette et al., 1995
Pilot	≥20	30	GAC	19.9	0.177	Summers et al., 2006
Bench	≥20	30	A	22	0.177	Fonseca et al., 2003
Pilot	10 -- 20		A/S	10		Halle, 2009

Table A8. Biofilter data for ozonated surface waters.

Study	Temperature Range (°C)	EBCT (min)	Media	DOC Removal (%)	k′ (min^{-1})	Reference
Pilot	10 --13	2.1	A/S	7	0.127	Coffey et al., 1995
Pilot	20 -- 25	2.1	A/S	6	0.106	Coffey et al., 1995
Full	20 -- 25	4	A	6	0.056	Evans et al., 2010
Full	21 - 22	5.1	A	15	0.136	Selbes et al., 2017
Full	11 -- 21	6	A	17	0.139	Daniel et al., 1995
Full	11 -- 21	6	A	26	0.336	Daniel et al., 1995
Full	11 -- 21	6	A	8	0.052	Daniel et al., 1995
Full	11 -- 21	6	A	6	0.037	Daniel et al., 1995
Ful	11 -- 21	6	A	7	0.0	Daniel et al., 1995

l					44	
Full	11 -- 21	6	A	3	0.018	Daniel et al., 1995
Full	11 -- 21	6	A	13	0.095	Daniel et al., 1995
Full	11 -- 21	6	A	9	0.059	Daniel et al., 1995
Full	11 -- 21	6	A	5	0.030	Daniel et al., 1995
Full	11 -- 21	6	A	34		Daniel et al., 1995
Full	11 -- 21	6	A	26	0.336	Daniel et al., 1995
Pilot	10 -- 20	6	A	9	0.059	Carlson et al., 1996
	15 -- 19	6	A	16	0.127	Melin et al., 2006
Full	11 -- 21	6	A	16	0.127	Daniel et al., 1995
Full	11 -- 21	6	A	7	0.044	Daniel et al., 1995
Pilot	11 -- 30	6	GAC (exhausted)	11	0.076	Lauderdale et al., 2012
Pilot	21 -- 25	6	A/S	20	0.183	Dugan et al., 1997
Pilot	21 -- 25	6	S	20	0.183	Dugan et al., 1997
Pilot	≤10	7	S	15	0.099	Moll et al., 1999
Pilot	≥20	7	S	24	0.230	Moll et al., 1999
Pilot	≥20	7	S	24	0.230	Moll et al., 1999
Full	10 -- 20	7.1	A/S	5	0.026	Selbes et al., 2016
Full	20 -- 25	7.1	A/S	24	0.227	Selbes et al., 2016
Full	21 -- 22	7.1	A	21	0.176	Selbes et al., 2017
Pilot	≥20	7.4	A/S	20	0.148	Miltner and Summers, 1992
Pilot	21 -- 23	8.5	A	14	0.072	Selbes et al., 2017
Pilot	11 -- 13	8.8	GAC (exhausted)	22	0.153	Selbes et al., 2017
Pilot	≥20	9	A	18	0.102	Wang et al., 1995
Full	10 -- 20	9	GAC (>2years)	16	0.080	Servais et al., 1991

Pilot	≥20	9.5	S	24	0.172	Solarik et al., 1996
Bench	≥20	9.5	S	29	0.358	Solarik et al., 1996
Pilot	8 -- 10	10	A	9.8	0.040	Carlson et al., 1995
Full	8 -- 21	10	A, GAC, S	16	0.076	Evans et al., 2013
	10 -- 20	10	GAC (48 months)	20	0.110	Uhl and Gimbel, 1996
Pilot	13.7 -- 25.4	10	GAC (>120 months)	11	0.046	Pelato et al., 2016
Pilot	13.7 -- 25.4	10	GAC (>120 months)	10	0.041	Pelato et al., 2016
Pilot	13.7 -- 25.4	10	GAC (>120 months)	15	0.069	Pelato et al., 2016
Pilot	13.7 -- 25.4	10	GAC (>120 months)	13	0.057	Pelato et al., 2016
Full	≤10	10.5	GAC (exhausted)	3	0.010	Wang & Siembida-Losch, 2013 (via Pharand et al., 2014)
Pilot	≤10	13	S/GAC	9	0.029	Prevost et al., 1995
Pilot	≤10	13	S/GAC	9	0.029	Prevost et al., 1995
Pilot	10 -- 20	13	S/GAC	12.5	0.041	Prevost et al., 1995
Pilot	≤10	15	S	7	0.019	Singer et al., 1999
Pilot	4 -- 10	15	GAC (exhausted)	13	0.038	Vahala et al., 1998
Pilot	≤10	15	S	15	0.048	Singer et al., 1999
Full	10 -- 20	15	A/S	17	0.056	Staf et al., 2014
Pilot	10 -- 20	15	S	5	0.012	Singer et al., 1999
Pilot	≤10	16	GAC (exhausted)	22	0.083	Velten et al., 2011
Pilot	≤10	17	A	17	0.049	Fonseca et al., 2003
Pilot	≥20	17	A	26	0.119	Fonseca et al., 2003
Pilot	7 -- 8	17.5	A/S	24	0.093	Chaiket et al., 1999
Full	21 -- 25	20	A/S	20	0.055	Emelko et al., 2006
Pilot	10 -- 20	24	A	20	0.046	Wricke et al., 1996
Pilot	8 -- 10	25	A	9.8	0.016	Carlson et al., 1995

Full	1 -- 3	26.5	A/S	14	0.024	Emelko et al, 2006
	≤10	28	GAC (exhausted)	16	0.027	Odegaard et al., 2006
	10 -- 20	28	GAC (exhausted)	25	0.064	Odegaard et al., 2006
Pilot	≥20	30	GAC	45	0.190	Hubele, 1995
Pilot	≥20	30	S	20	0.037	Hubele, 1995
Full	3 -- 28	38	A	12	0.013	Pharand et al, 2013
Pilot	≤10		A/S	7		LeChevallier et al., 1991b
Pilot	≥20		A	11		Booth et al., 2001
Pilot	15 -- 20		A/S	47		Reckhow et al., 1992
Bench	≥20		A/S	13		Swertfeger et al., 1993
Pilot	≥20		A	11		Amburgey et al., 2001
Pilot	>20		A	11		Booth et al., 2001
Pilot	>20		A	11		Booth et al., 2001
Pilot	>20		A/S	16		Swertfeger et al., 1993

***Some cases BDOC/TOC was given, while in other cases BDOC concentration was given. Similarly, in some cases AOC/TOC was given, while in other cases AOC concentration was given.**

Appendix B
Modeling biological filtration performance for organic matter removal

Table B1. Phospholipid biomass concentrations and TOC removal as a function of preozonation, media type, chlorinated backwash, chlorine residual in the influent and depth for Wang et al. (1995) and Dugan (1998) both at EBCT = 9.2 min and Chowdhury et al. (2010) at EBCT = 7.1 min

Pre-oxidation	Media	Backwash	Phospholipid Biomass (nmol P/mL)			TOC removal (%) at steady-state
			Top	Middle	Bottom	
Wang et al.						
O_3 & Cl_2 Influent	Anthracite	Cl_2	2 (0.5)	-	-	8
O_3	Anthracite	Cl_2	15 (0.5)			16
O_3	Anthracite	-	50 (2)			20
O_3	Sand	-	108 (2)			20
O_3	GAC1	-	128 (4)			29
O_3	GAC2	-	172 (4)			27
O_3	GAC3	-	80 (5)			23
Dugan						
O_3	Anthracite	Cl_2	22	7	4	15
O_3	Anthracite	NH_2Cl	96	16	11	22
O_3	Anthracite	-	104	25	20	27
O_3	Sand	-	115	26	17	26
-	Anthracite	Cl_2	13	5	3	11
-	Anthracite	-	53	33	16	17
Chowdhury et al.						
Cl_2 influent	GAC	Cl_2	41(5)	-	32(2)	7
-	GAC	Cl_2	47(3)	-	27(2)	9
-	GAC	-	52(4)	-	29(3)	9
-	Anthracite	-	31(1)		25(1)	9

Values in parenthesis are standard deviations.

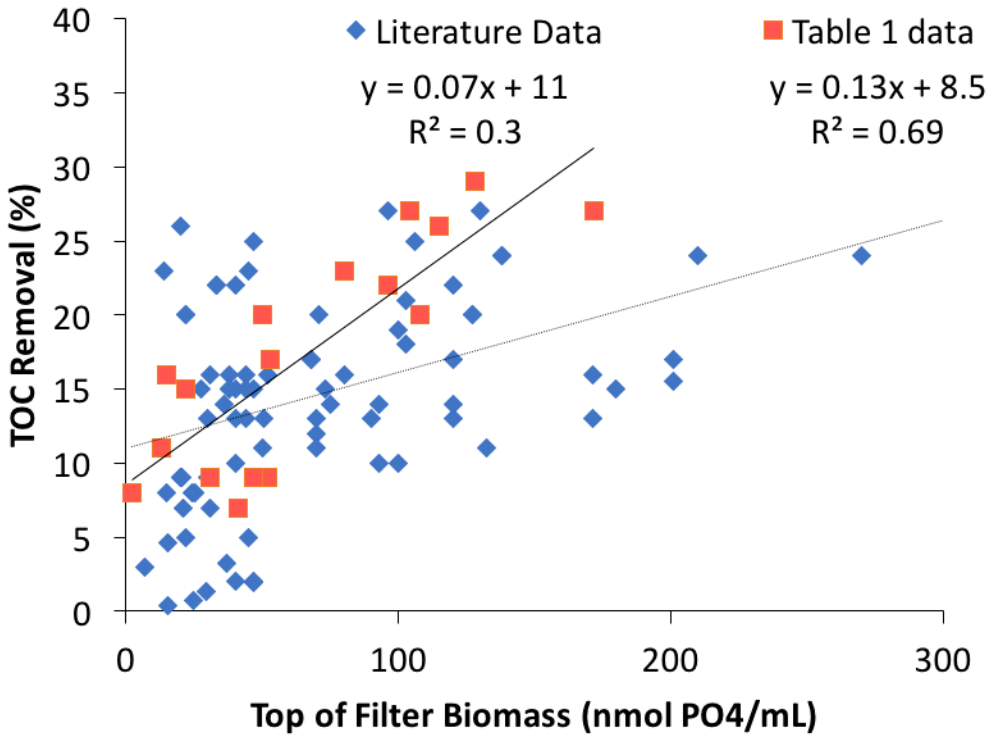

Figure B1. TOC removal as a function of top of filter biomass (n=108) at various temperatures with and without preozonation for Table B1 and other literature data.

Table B2. Biomass distribution equation coefficient values

Biomass Method	Pre-Oxidation	a	b	R^2	n
Phospholipid & ATP	No ozone	-0.12	0.72	0.67	117
Phospholipid & ATP	Ozone	-0.155	0.65	0.67	115
Phospholipid only	No ozone	-0.13	0.7	0.73	88
Phospholipid only	Ozone	-0.16	0.65	0.68	112
All	All	-0.135	0.681	0.65	232

Table B3. Biomass concentrations as a function of depth at the top of the filter.

ATP biomass (ng cATP/g media)			
	very top	3" deep	Difference (%)
Unacclimated Filter	1,762	220	88
Acclimated Filter	2,460	556	77

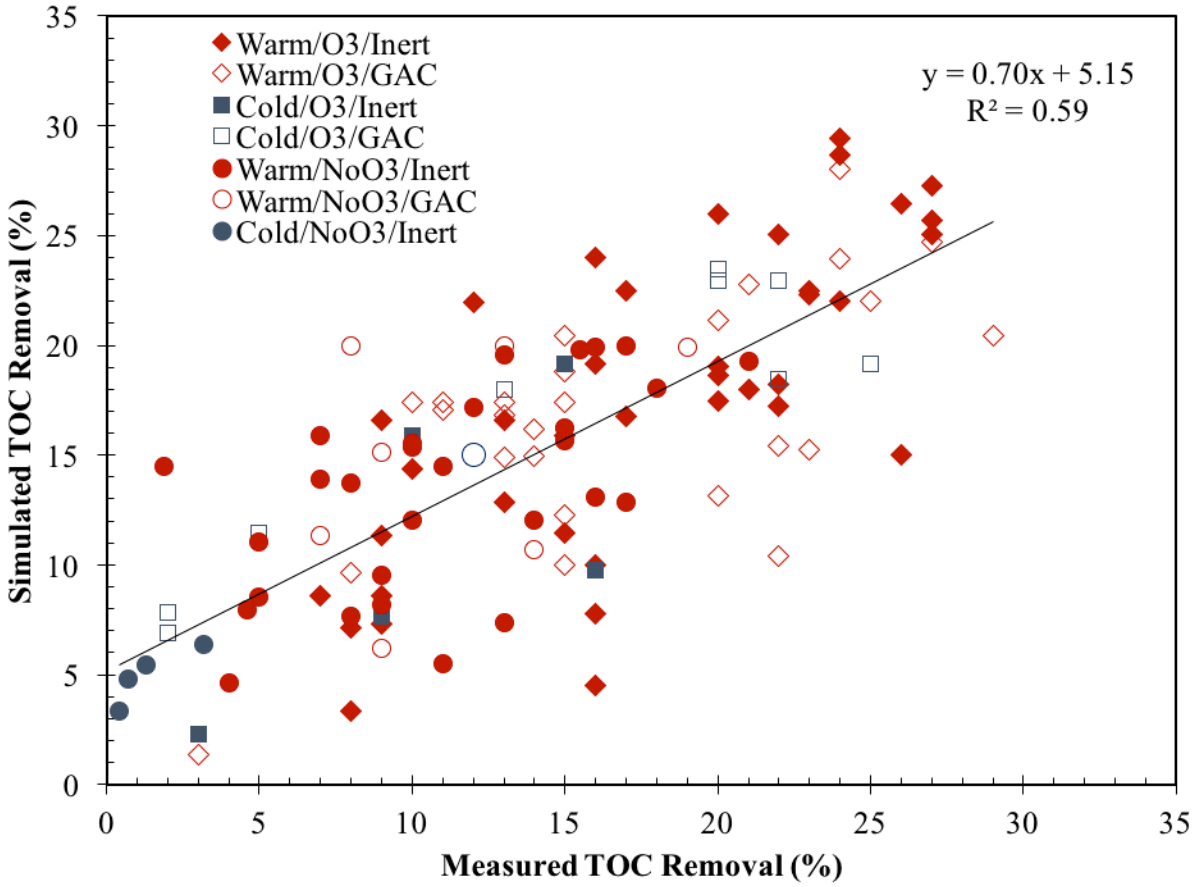

Figure B2. Regression analysis of simulated and predicted percent TOC removal at all conditions evaluated. GAC represented as hollow symbols and inert media represented by solid symbols.

Table B4. Average Residuals for Models

	Average Residuals for TOC % Removal – using individual k" constants			
	GAC	**Inert**	**Total**	**n**
Warm/O3	4.7	3.7	4.1	64
Warm/NoO3	1.0	3.6	3.1	36
Cold/O3	4.2	3.9	4.1	14
Cold/NoO3	4.97	0.8	1.6	5
TOTAL:	3.7	3.0	3.7	119
	Average Residuals for TOC % Removal using one k" constant			
	GAC	**Inert**	**Total**	**n**

Warm/O3	4.2	3.6	3.9	64
Warm/NoO3	1.9	3.9	3.5	36
Cold/O3	4.3	3.7	4.1	14
Cold/NoO3	2.99	3.6	3.5	5
TOTAL:	3.4	3.7	3.8	119

Appendix C
Understanding Biofiltration Performance Based on Extended EBCT and Biomass Acclimation and Distribution

C.1 Pilot Plant Schematic

The pilot plant schematic is represented by Figure C1 and a detailed schematic of the biofilters can be seen in Figure C2. Source water to the City of Boulder's Betasso Drinking Water Treatment Plant was diverted to the pilot plant. The water passed through the static mixer, into three flocculation basins with tapered paddles, into a sedimentation basin with plate settlers, then split into one of two filters. When the pilot plant was operating in direct biofiltration, no chemicals were added. When the pilot plant experiment incorporated coagulation, aluminum sulfate was added at the front of the pilot plant. Chlorine was added at the front of the second filter to allow comparison of a biofilter (BF) and a conventional filter, in the last set of experiments. The biofilter schematic is explained in the following chapter of this dissertation: "Understanding Biofiltration Performance Based on Extended EBCT and Biomass Acclimation and Distribution".

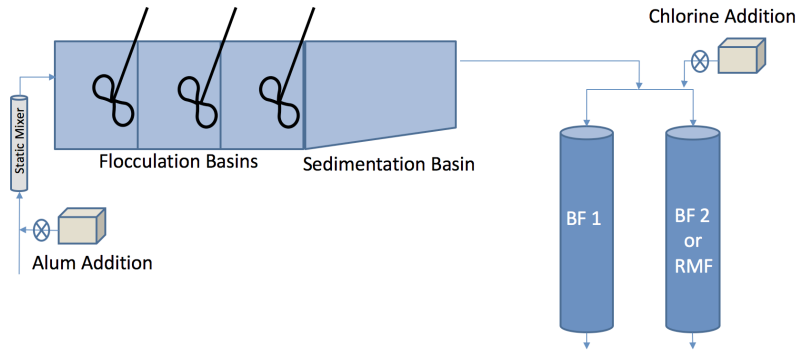

Figure C1. Pilot Plant Schematic

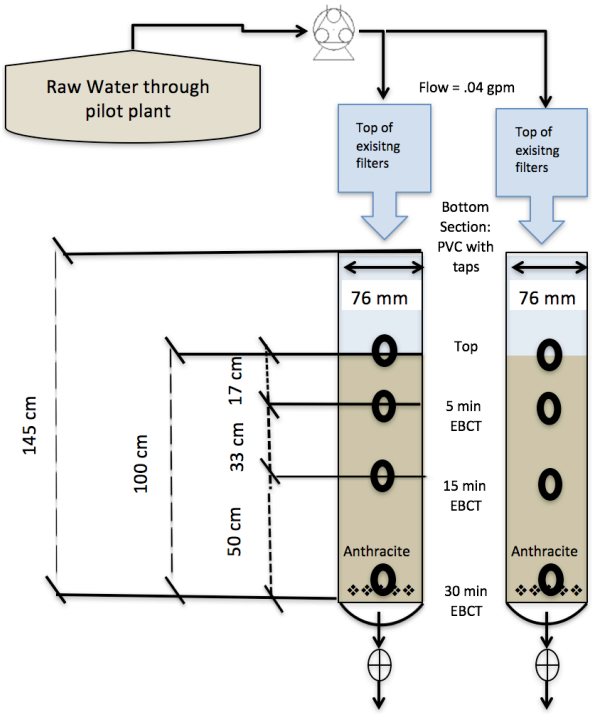

Figure C2. Biofilters Pilot Schematic

C.2 Biofiltration Optimization Based on Coagulant Dosing

Biofilter efficacy can be improved when the coagulant dose is optimized. Biofilter DOC removal as a function of empty bed contact time (EBCT) for a biological filter and a rapid media filter at three different aluminum sulfate (alum) doses (10, 15, and 20 mg/L) can be seen in Figure C3. The coagulant doses were chosen after jar test experiments. The rapid media filter does not remove any dissolved organic carbon (DOC), as expected. The biofilter with the smallest

coagulant dose (10 mg/L) removed the most DOC at 13% DOC removal. The biofilter removed less DOC with increasing coagulant dose, 11% DOC removal accompanied the 15 mg/L alum dose and 5% removal with the 20 mg/L alum dose. To get the most removal from your biofilter, optimizing coagulant dose for specific source waters is recommended.

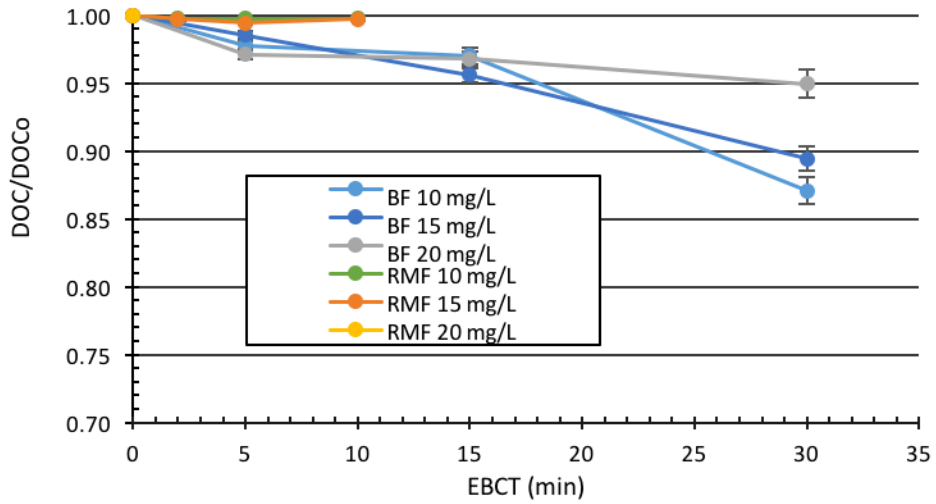

Figure C3. DOC removal throughout the filter as a function of EBCT for a biological filter (BF) and a conventional rapid media filter (RMF) at 3 different aluminum sulfate doses: 10, 15 and 20 mg/L.

Drinking water treatment utilities are required to remove a certain percentage of influent TOC (15 – 50%) based on source water TOC and alkalinity per the Stage 1 DBP Rule (termed the enhanced coagulation requirement). Utilities have multiple ways of meeting this regulation. The pilot plant was used to determine the trade-offs between DOC removal from coagulant addition and biofiltration. Figure C4 demonstrates the best optimization of biofiltration and coagulant addition to meet the Stage 1 DBP Rule requirements. If the utility is required to remove 30% of the influent TOC, the utility can either dose at 20 mg/L alum to achieve 33% DOC removal or dose at 15mg/L alum and run a biofilter with an EBCT of 30 minutes to achieve 32%. If the source water changes and the utility needs to remove 20% of the influent TOC, the utility can

dose 15 mg/L alum to achieve 25% DOC removal or dose 10 mg/L alum and run a biofilter with an EBCT of 30 minutes to achieve 23% DOC removal. Understanding removal requirements and how coagulation and biofiltration combined can meet regulations can save the utility cost associated with coagulant: transportation, storage, mass and energy.

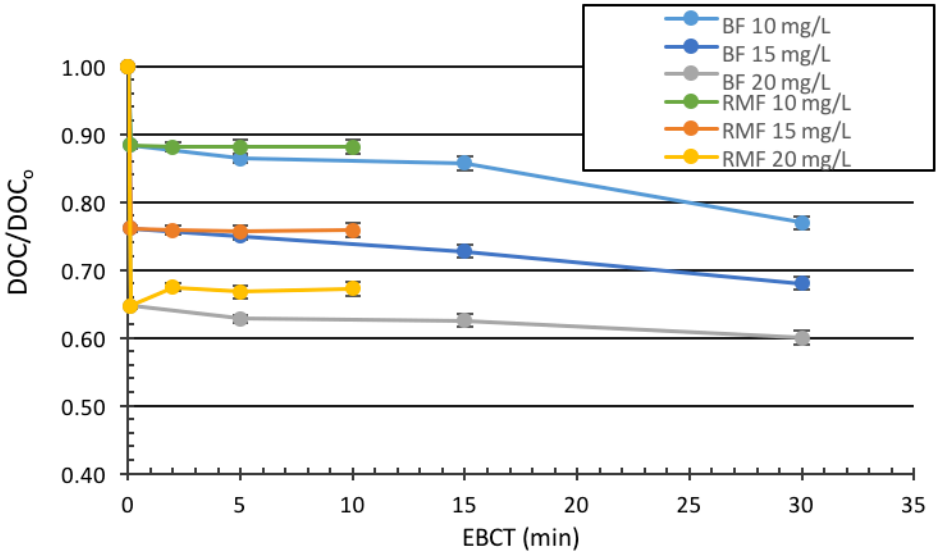

Figure C4. DOC removal throughout the coagulation and filtration process as a function of EBCT for a biological filter (BF) and a conventional rapid media filter (RMF) at 3 different aluminum sulfate doses: 10, 15 and 20 mg/L.

C.3 Biofiltration Response to Turbidity Perturbations and Intermittent Alum Dosing

The pilot plant was employed to test the response of biofilters to turbidity perturbations and spikes in particulate matter. The influent to the biofilters had an average turbidity of 0.8 NTU and the effluent averaged between 0.3 and 0.5 NTU for direct biofiltration. After the pilot plant had been in operation for 1 year, the sedimentation basin was stirred manually for 15 minutes to allow spikes of particulates to enter the biofilter. Online SCADA turbidity units measured NTU values every minute. The sedimentation effluent (influent to biofilters) spiked to 35 NTU and decreased slowly. The biofilters handled the perturbation well as a small peak in effluent turbidity was released as shown in Figure C5. Biofilter 2 effluent did not exceed 1.5 NTU and returned to 0.3 NTU within two hours. Biofilter 1 effluent did not exceed 1.75 NTU and returned

to 0.5 NTU within two hours. This experimental data suggest biofilters can handle surges and spikes in particulate matter within a reasonable time frame.

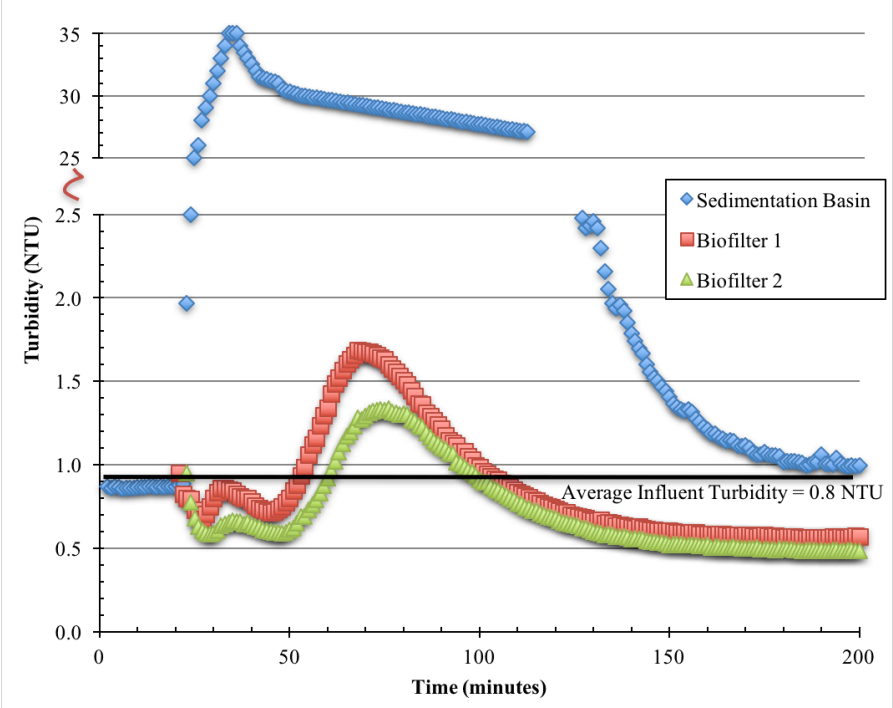

Figure C5. Biofilters response to turbidity perturbations from 1 NTU up to 35 NTU. Both biofilters were able to respond quickly and efficiently to the spike in turbidity and particle surges.

Next, the pilot plant was employed to examine the effects of intermittent aluminum sulfate (alum) dosing on biofilter effluent turbidity and rapid media filter effluent turbidity to represent start/stop coagulation addition. For this study, chlorine was added before the second filter, and no biomass activity was detected in the rapid media filter during this experiment. Online SCADA turbidity units measured NTU values every minute. The results of the start/stop alum addition for the sedimentation basin (Sed Basin), the biofilter (BF) and the conventional rapid media filter (RMF) can be seen in Figure C6. A 15mg/L alum dose began on Day 1 and the turbidity reached steady state when all three turbidities were under 0.4 NTU. On day 3, the alum addition was turned off and turbidity immediately started to rise in all locations. Without the addition of coagulant, the turbidity rose to 1 NTU at the end of the sedimentation basin, 0.8 NTU

at the rapid media filter effluent and 0.6 NTU at the biofilter effluent. Next, this study was repeated for a higher hydraulic loading rate of 6 m/hr. At the higher HLR, the results were similar as the lower HRL, as turbidity increased when the coagulant dosing was terminated, and the biofilter turbidity effluent was lower than the rapid media filter turbidity effluent. However, the effluent turbidity for the biofilter and rapid media filter were higher compared the turbidity results with the lower HLR. All experiments were performed in triplicate and the average turbidity results can be seen in Table C1. The biofilter outperformed the rapid media filter in effluent turbidity levels in every scenario and better controlled intermittent alum dosing.

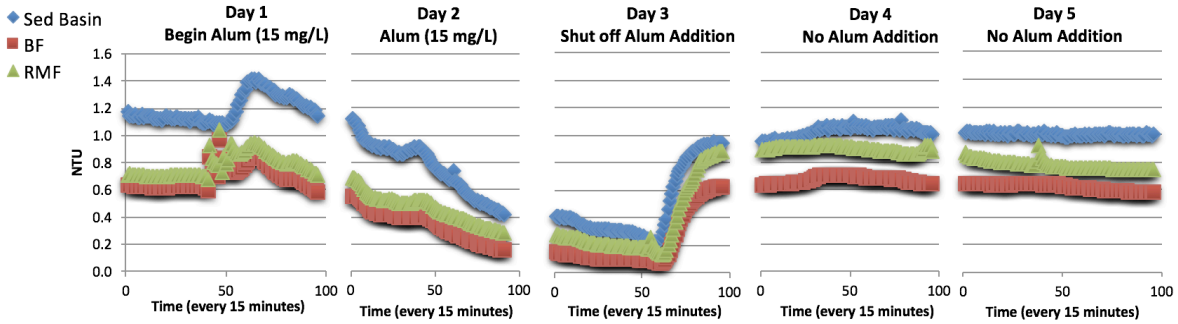

Figure C6. Biofilter (BF) and a conventional rapid media filter (RMF) response to intermittent coagulant operations.

			Turbidity (NTU)		
HRL (m/hr)	**EBCT (min)**	**Alum Dose (mg/L)**	**Sed. Basin**	**Biofilter**	**RMF**
2	30	15	0.2	0.025	0.061
2	30	0	1.0	0.60	0.80
6	10	15	0.25	0.09	0.10
6	10	0	1.2	0.75	0.80

Table C1. Sedimentation basin (Sed. Basin), biofilter (BF) and a conventional rapid media filter (RMF) response to intermittent coagulant operations at different hydraulic loading rates (average of triplicate runs).

C.4 Extracellular Polymeric Substances (EPS) Analysis

Extracellular polymeric substances (EPS) are compounds secreted by microbes that help establish the functional and structural integrity of biomass communities, but do not provide any degradation of the organic matter. EPS, via polysaccharides, were measured before and after

backwashing for two coagulant doses and two hydraulic loading rates (HRL). EPS was higher in every scenario before backwash and decreased after backwash at each filter depth, as seen in Figure C7. This trend was more pronounced at the top of the filter (a factor of 2) and less pronounced throughout the bed of the filter. The average reduction of EPS after backwash was 1.92 µg/g, 0.87 µg/g, 0.87 µg/g and 1.11 µg/g for the top, 5 minute EBCT, 15 minute EBCT and 30 minute EBCT respectively. EPS was lowest at the higher HLR compared to the lower HLR. Comparing different coagulant doses, EPS was lower at the coagulant dose of 3 mg/L alum compared to dose of 15 mg/L alum. EPS decreased with filter depth, which is expected as biomass concentration and activity decrease with filter depth. Backwashing had a large effect on EPS at the top and bottom of the filter, but had less effect on EPS at the middle of the filter.

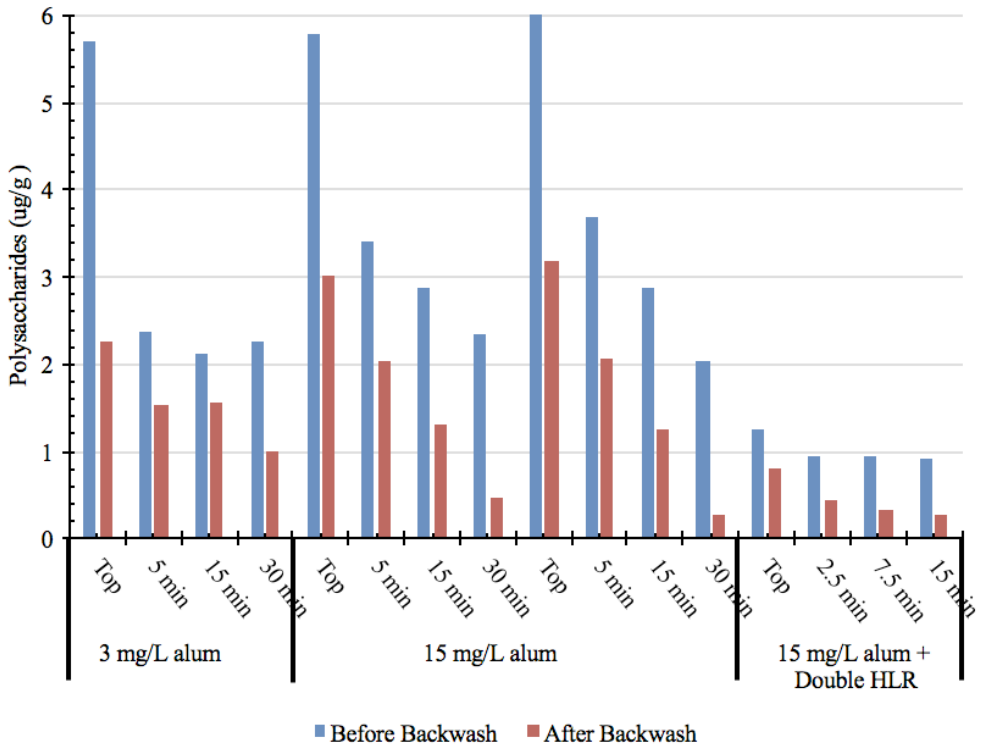

Figure C7. Extra polymeric substances (EPS) before and after backwash for 3 different aluminum sulfate doses and two different hydraulic loading rates (HLR)

Appendix D
Environmentally Sustainable Scenarios for Drinking Water Biological Filtration

D1. Water Quality Regulations

Table D1. Total organic carbon (TOC) percent removal requirements, as a function of source water TOC and alkalinity, as defined by the enhanced coagulation requirement in the Stage 1 DBP Rule (McGuire et al. 2002, U.S. EPA 1998).

Source Water TOC (mg/L C)	Source Water Alkalinity (mg/L $CaCO_3$)		
	<60	60 to 120	>120
>2 to 4	35%	25%	15%
>4 to 8	45%	35%	25%
>8	50%	40%	30%

Table D2. Distribution by source water quality (enhanced coagulation requirement) bin of the water treatment plants that are required to remove TOC (McGuire et al. 2002). This accounts for about 60% of U.S. water plants since about 40% of have source water TOC equal to or below 2 mg/L and are not required to remove TOC. n/a is not available due to lack of sufficient data.

Source Water TOC (mg/L C)	Source Water Alkalinity (mg/L $CaCO_3$)		
	<60	60 to 120	>120
>2 to 4	26%	27%	16%
>4 to 8	15%	10%	5%
>8	2%	n/a	n/a

D2. Life Cycle Inventory and Impact Assessment Categories

Table D3. Life cycle unit process data and descriptions. Data were from the Ecoinvent v3 database (Swiss Centre for Life Cycle Inventories 2014) except for unit process data on anthracite coal, which was from US-EI 2.2 database (Earthshift 2014). The relative amount of US electricity produced by each electrical grid is stated under application (percent contribution) (U.S. EIA 2016); n/a is not available.

Description	Calc. Section #	Unit Process Name	Application
Alum	D3.1	Aluminium sulfate, powder {RoW}| production | Alloc Def, U	Coagulation
Anthracite	D4.1	Anthracite coal, at mine NREL/RNA U	Filter media
Caustic Soda	D5	Sodium hydroxide, without water, in 50% solution state {RoW}| chlor-alkali electrolysis, membrane cell | Alloc Def, U	pH adjustment
Chlorine	(Jones et al. 2017)	Sodium hypochlorite, without water, in 15% solution state {RoW}| sodium hypochlorite production, product in 15% solution state | Alloc Def, U	Disinfection (free chlorine from NaOCl)
Concrete	(Jones et al. 2017)	Concrete, 20MPa {RoW}| concrete production 20MPa, RNA only | Alloc Def, U	Chlorine contact basin
Electricity	D4.2, (Jones et al. 2017)	Electricity, medium voltage {ASCC}| market for | Alloc Def, U	ASCC grid (n/a)
		Electricity, medium voltage {FRCC}| market for | Alloc Def, U	FRCC grid (6% US electricity)
		Electricity, medium voltage {NPCC, US only}| market for | Alloc Def, U	NPCC grid (7% US electricity)
		Electricity, medium voltage {MRO, US only}| market for | Alloc Def, U	MRO grid (17% US electricity)
		Electricity, medium voltage {RFC}| market for | Alloc Def, U	RFC grid (20% US electricity)
		Electricity, medium voltage {SERC}| market for | Alloc Def, U	SERC grid (17% US electricity)
		Electricity, medium voltage {SPP}| market for | Alloc Def, U	SPP grid (6% US electricity)
		Electricity, medium voltage {TRE}| market for | Alloc Def, U	TRE grid (9% US electricity)

		Electricity, medium voltage {WECC, US only}| market for | Alloc Def, U	WECC grid (18% US electricity)
		Electricity, medium voltage {HICC}| market for | Alloc Def, U	HICC grid (n/a)
Hauling	D4.3	Trasport freight, lorry 3.5-7.5 metric ton, EURO6 {RoW}| transport, freight, lorry 3.5-7.5 metric ton, EURO6 | Alloc Def, U	Solids and chemical hauling
Reinforcing Steel	(Jones et al. 2017)	Reinforcing steel {RoW}| market for | Alloc Def, U	Chlorine contact basin
Sand	D4.1	Sand {GLO}| market for | Alloc Def, U	Sand (filter media)
Soft Plastic	(Jones et al. 2017)	Polyethylene, high density, granulate {RoW}| production | Alloc Def, U	Plastic cylindrical tank (contact basin)
Stainless Steel	D4.1, (Jones et al. 2017)	Steel, chromium steel 18/8, hot rolled {RoW}| production | Alloc Def, U	Filter housing, steel baffles
Tap Water	(Jones et al. 2017)	Tap water {RoW}| tap water production, conventional treatment | Alloc Def, U	Dilution water (chlorine solution)

Table D4. Material, energy, and chemical quantities for the 2 filtration alternatives. Values are for the entire functional unit (i.e., treatment of 2,700 m^3/day over 40 years) and are the outputs from the expected uncertainty parameters. Unit process details are in Table D3. Chlorine mass is kg dry 15% sodium hypochlorite solution.

Inventory Unit Process (Units)	**Filtration Alternatives**	
	Conventional Filtration	Biofiltration
Alum (kg)	1.31 E+06	7.16 E+05
Anthracite (kg)	5.44 E+03	5.44 E+03
Backwash Energy (kWh)	3.77 E+04	3.77 E+04
Baffle Steel (kg)	9.19 E+03	1.44 E+04
Caustic (kg)	5.29 E+05	3.98 E+05
Chemicals Hauling (tkm)	3.92 E+04	2.46 E+04
Chlorine (kg)	5.57 E+04	5.57 E+04
Chlorine Dose (mg free Cl_2/L)	1.4	1.4
Chlorine Pump Energy (kWh)	2.49 E+02	2.49 E+02
Polyethylene Chlorine Storage Tank (kg)	3.73 E+01	3.73 E+01
Concrete (m^3)	4.52 E+01	6.83 E+01
Contactor Pump Energy (kWh)	1.02 E+00	3.86 E-01
Stainless Steel Filter Housing (kg)	2.97 E+04	2.97 E+04
Filter Operational Energy (kWh)	5.52 E+05	5.52 E+05

Rebar (kg)	1.91 E+02	3.10 E+02
Sand (kg)	6.81 E+03	6.81 E+03
Solids Hauling (tkm)	2.63 E+04	1.44 E+04

Table D5. TRACI Environmental Impact Category descriptions (Bare 2012).

Impact Category	Unit	Description
Ozone depletion	kg CFC-11 eq	Ozone provides protection from radiation. Emissions of substances known as chlorofluorocarbons (CFCs) reduce stratospheric ozone levels.
Carcinogenics	CTUh	Substances are chemicals of concern that can cause cancer in humans
Non Carcinogenics	CTUh	Substances are chemicals of concern that are toxic to humans but do not cause cancer.
Respiratory effects	kg PM2.5 eq	Fine particulate matter and precursors to particulates ambient in the air can be inhaled by a human and cause respiratory illnesses or even death
Eutrophication	kg N eq	Excess of nutrients in a body of water results in dense growth of plants and algae and a reduction of oxygen.
Acidification	kg SO2 eq	A decrease in the pH of water because of the uptake of CO2 and SOx
Smog	kg O3 eq	Ground level ozone can cause respiratory illnesses and ecosystem damages. Ozone is created with the presence of nitrogen oxides (NOx) and volatile organic compounds (VOCs)
Fossil fuel depletion	MJ surplus	Non-site specific use of fossil fuels.
Global warming	kg CO2 eq	The raising of the Earth's atmospheric temperature due to the increase in CO2 and other greenhouse gas emissions. Global warming has many additional adverse climate and health effects
Ecotoxicity	CTUe	Substances are chemicals of concern that are toxic to the ecosystem.

D3. TOC Removal Design Calculations

D3.1 Coagulation

Alum coagulation is effected most by pH and specific ultraviolet absorbance (SUVA), both of which are effected by alkalinity and TOC. Also, alum lowers the water's pH and alkalinity. Due to these complex interactions, the following approach was used to determine the alum dose needed for a specific percent TOC removal target. An alum dose and TOC removal table (Table D6) was generated for every source water scenario to determine the proper alum dose for coagulation using 6 main steps. First, the source water quality was defined in terms of TOC, alkalinity, pH, SUVA, and temperature. Second, a comprehensive range of possible alum doses was generated (from 0 to 75 mg/L, in 1 mg/L increments). Third, the pH of the coagulated water was calculated by iteratively solving Eq. D1 from the U.S. EPA's Water Treatment Plant Model v2 (U.S. EPA 2001). Fourth, the coagulated water TOC was calculated using Eq. D2 with values

for the coagulated water pH and alkalinity input as well as variables from the Edwards Model (Edwards, 1997). Fifth, the SUVA of coagulated water was determined (Eq. D3). Sixth, the percent TOC removal was calculated based on the source water TOC and final TOC. Table D6 shows example alum doses and the corresponding percent TOC removals for an example source water. Overall, the required alum dose for a specified source water quality and TOC removal target was found from these tables. The final, selected alum dose was the smallest dose associated with any of the follow situations, as long as that value was above the minimum allowable value (18 mg/L alum, uncertainty parameter, (Table D11): (i) percent TOC removal target, (ii) 2 mg/L coagulated water TOC, or (iii) 2 L/mg/m coagulated water SUVA. Table D7 shows the alum doses needed for all of these conditions for an example source water.

Table D6. Example alum dose and TOC removal table. Values were calculated for the national average source water scenario (77 mg/L $CaCO_3$, 3.2 mg/L TOC, 15 °C).

Alum Dose (mg/L)	Coagulated Water pH	Coagulated Water TOC (mg/L)	Coagulated Water SUVA	TOC Removal (%)
0	7.50	3.20	3.13	0%
1	7.46	3.18	2.77	0.7%
2	7.42	3.16	2.70	1.4%
…	**…**	**…**	**…**	**…**
33	6.8	2.38	2.60	25%

Table D7. The four alum dose options for the national average source water scenario (77 mg/L $CaCO_3$, 3.2 mg/L TOC, 15 °C). The smallest, above the minimum allowable alum dose, was chosen as the modeled alum dose (green shade).

Purpose	Alum Dose (mg/L)	Coagulated Water TOC (mg/L)	Coagulated Water SUVA	TOC Removal (%)
Turbidity removal (minimum allowable dose)	18	2.76	2.50	14%
SUVA (≤ 2 L/mg/m)	DNO*	n/a	n/a	n/a
TOC (≤ 2 mg/L)	52	2.00	2.73	38%
%TOC removal	**33**	2.38	2.59	25%

*DNO = does not occur. SUVA did not drop below 2 mg/L for this source water quality.

$$(\alpha_1 + (2^*\alpha_2))^*[Ct_{CO3}] + \left[\frac{k_w}{[H^+]}\right] - [H^+] = [\alpha_1{}^*C_{TCO3}] + 2^*[\alpha_2{}^*C_{TCO3}] + \left[\frac{k_w}{[H^+]}\right] - [H^+] - 6^*[Alum] \quad \textbf{Eq. D1a}$$

$$\alpha_1 = \frac{k_{1CO3} * [H^+]}{[H^+]^2 + k_{1CO3} * [H^+] + k_{1CO3} * k_{2CO3}} \quad \textbf{Eq. D1b}$$

$$\alpha_2 = \frac{k_{1CO3} * k_{2CO3}}{[H^+]^2 + k_{1CO3} * [H^+] + k_{1CO3} * k_{2CO3}} \quad \textbf{Eq. D1c}$$

$$C_{TCO3} = \frac{[Alk] + [H] - [OH]}{\alpha_1 + (2 * \alpha_2)} \quad \textbf{Eq. D1d}$$

$$k_{1CO3} = \exp\left\{\left[\left(\frac{7700 \frac{J}{mole}}{8.314 \frac{J}{K*mole}}\right)\left(\frac{1}{298.15\ K} - \frac{1}{T}\right)\right] - 14.5\right\} \quad \textbf{Eq. D1e}$$

$$k_{2CO3} = \exp\left\{\left[\left(\frac{14900 \frac{J}{mole}}{8.314 \frac{J}{K*mole}}\right)\left(\frac{1}{298.15\ K} - \frac{1}{T}\right)\right] - 14.5\right\} \quad \textbf{Eq. D1f}$$

Where:

$[OH^-]$ = Concentration of hydroxide (M)
$[H^+]$ = Concentration of hydrogen (M)
$[CO_3^{2-}]$ = Concentration of carbonate (M)
$[HCO_3^-]$ = Concentration of bicarbonate (M)
[Alum] = Concentration of alum added (M)
α_1 = Water chemistry equilibrium value for the second hydrogen state (Eq. D1b)
α_2 = Water chemistry equilibrium value for the third hydrogen state (Eq. D1c)
C_{TCO3} = Total concentration of all carbonate species (M)
k_{1CO3} = Carbonate equilibrium constant for second hydrogen (Stumm & Morgan 1981)
k_{2CO3} = Carbonate equilibrium constant for second hydrogen (Stumm & Morgan 1981)
k_w = Water equilibrium constant (4.52E-15)
[Alk] = Concentration of influent alkalinity eq/L
T = Influent water temperature (K)

$$\mathbf{DOC_i} = (\mathbf{1}\text{-}(\mathbf{SUVA}^*\mathbf{K_1})\text{-}\mathbf{K_2})^*\mathbf{TOC} \quad \textbf{Eq. D2a}$$

$$\mathbf{Al^{3+}} = \frac{\mathbf{Alum}^*\left(\frac{\mathbf{2\ mmol\ Al^{3+}}}{\mathbf{1\ mmol\ Alum}}\right)}{\left(\frac{\mathbf{594.4\ mg\ Alum}}{\mathbf{1\ mmol\ Alum}}\right)} \quad \textbf{Eq. D2b}$$

$$\mathbf{a} = (\mathbf{x_1}^*\mathbf{pH^3} + \mathbf{x_2}^*\mathbf{pH^2} + \mathbf{x_3}^*\mathbf{pH}) \quad \textbf{Eq. D2c}$$

$$\frac{\mathbf{DOC_i}\text{-}[\mathbf{C}]_{\mathbf{eq}}}{\mathbf{Al^{3+}}} = \frac{\mathbf{a}^*\mathbf{b}^*[\mathbf{C}]_{\mathbf{eq}}}{\mathbf{1} + \mathbf{b}^*[\mathbf{C}]_{\mathbf{eq}}} \quad \textbf{Eq. D2d}$$

$$[\mathbf{C}]_{\mathbf{eq}} = \frac{\sqrt{\left(\mathbf{b^{2*}(DOC_i}\text{-}\mathbf{a}^*\mathbf{Al^{3+}})^2 + (\mathbf{2b}^*(\mathbf{DOC_i} + \mathbf{a}^*\mathbf{Al^{3+}}) + \mathbf{1})\right)} + \left(\boldsymbol{b}^*(\mathbf{DOC_i}\text{-}\mathbf{a}^*\mathbf{Al^{3+}})\right)\text{-}\mathbf{1}}{\mathbf{2b}} \quad \textbf{Eq. D2e}$$

$$\mathbf{TOC_{coagulated}} = [\mathbf{C}]_{\mathbf{eq}} + (\mathbf{TOC}\text{-}\mathbf{DOC_i}) \quad \textbf{Eq. D2f}$$

$$\mathbf{TOC\ Removal} = \frac{\mathbf{TOC}\text{-}\mathbf{TOC_{coagulated}}}{\mathbf{TOC}} \quad \textbf{Eq. D2g}$$

Where:

DOC_i = Sorbable DOC of coagulation influent water (mg/L)

SUVA = Specific ultraviolet absorbance of influent water (L/mg/m)

K_1 = Constant: (-0.075) (Edwards 1997)

K_2 = Constant: (0.56) (Edwards 1997)

TOC = Influent TOC (mg/L)

Alum = Alum dose added (mg/L)

Al^{3+} = Aluminum ions present (mM)

a = Maximum TOC sorption per mM of Al^{3+} added

x_1 = Constant: (284) (Edwards, 1997)

x_2 = Constant: (-74.2) (Edwards 1997)

x_3 = Constant: (4.91) (Edwards 1997)

$[C]_{eq}$ = Amount of sorbable TOC remaining after coagulation (mg/L)

b = Constant: (0.147) (Edwards 1997)

TOC = Influent TOC (mg/L)

$TOC_{coagulated}$ = Coagulated water TOC concentration (mg/L)

TOC Removal = Amount of TOC removed from source water (%)

$$\mathbf{SUVA_{coagulated}} = \frac{(\mathbf{5.716}^*(\mathbf{UVA})^{\mathbf{1.0894}}*(\mathbf{Al^{3+}})^{\mathbf{0.306}}*(\mathbf{pH})^{\mathbf{-0.9513}})}{\mathbf{TOC_{coagulated}}} * \frac{\mathbf{100cm}}{\mathbf{m}} \quad \textbf{Eq. D3}$$

Where:

$SUVA_{coagulated}$ = Specific ultraviolet absorbance of coagulated water (L/mg/m)

UVA = ultraviolet absorbance at 254 nm of influent water (1/cm)

D3.2 Biological TOC Removal

Table D8 shows experimental data from the literature (Terry & Summers 2017) on percent TOC removal using biofiltration, for biofiltration systems that match the treatment process configuration in this LCA (e.g., without pre-ozonation).

Table D8. Experimental data from the published literature that shows biofiltration TOC removal efficacy at different temperatures. Table data was adapted from Terry and Summers (2017).

Temperature	Biofilter percent TOC removal		
	Min	Max	Median
10 °C	2%	14%	10%
15 °C	5%	20%	12%
20 °C	10%	20%	15%

D4. Filter Design Calculations

Major materials and energy requirements to operate each filter were accounted for over the functional unit timeframe (40 years). A dual media filter of anthracite over sand was chosen for the rapid media filter design due to its prevalence in practice.

D4.1 Filter Materials

For each filter, the filter area was calculated using Eq. D4. The mass of media was calculated using Eq. D5. These equations assumed values for hydraulic loading rate and media depth, respectively, which were uncertainty parameters based on typical values for each type of filter (Table D11) (Kawamura, 2000). The total filter depth included the (packed) media depth and freeboard (0.3 m) (Kawamura, 2000). Filter volume was calculated using this total depth and Eq. D6. Then, the mass of steel needed for the filter housing was calculated using Eq. D7; this equation assumed a square cross section and typical steel thickness (Colorado Department of Public Health and Environment and Department of Civil and Environmental Engineering Colorado 2014), which was an uncertainty parameter (Table D11).

$$\mathbf{A_T = \frac{Q}{HLR}} \qquad \textbf{Eq. D4}$$

Where:

A_T = Total filter area requirement (m^2)
Q = Plant capacity flow rate (m3/hr)
HLR = Filter design hydraulic loading rate (m/hr)

$$\mathbf{M_{media} = A_T * D_{media} * \rho_{media}} \qquad \textbf{Eq. D5}$$

Where:

M_{media} = Mass of filter media (kg)
D_{media} = Media Depth (m) (Table D11)
ρ_{media} = Media density (kg/m^3): (1,500 kg/m^3 sand, and 800 kg/m^3 anthracite) (Urfer et al., 1997)

$$\mathbf{V_{Filter} = A_T * \left(D_{media} + H_{fb} + D_{expansion}\right)} \qquad \textbf{Eq. D6}$$

Where:

V_{Filter} = Required filter volume (m^3)
H_{fb} = freeboard (m)
$D_{expansion}$ = Backwash filter expansion depth (m): Assumed 50% bed expansion (Miltner et al., 1995)

$$\mathbf{M_{steel} = \left\{\left(\left(\sqrt{\frac{V_{filter}}{D_{total}}}\right) * t_b\right) + \left(4t_w^{\,2} + 4\left(\sqrt{\frac{V_{filter}}{D_{total}}}\right) * t_w\right) * \left(D_{total} + t_B\right)\right\} * \rho_{steel}} \qquad \textbf{Eq. D7}$$

Where:

D_{total} = Sum of media depth and filter head requirement (m)
t_b = Thickness of filter base (m)
t_w = Thickness of filter walls (m)

ρ_{steel} = Density of steel (kg/m^3): (7,500 kg/m^3) ("The Engineering ToolBox," n.d.)

D4.2 Filter Energy Requirements

Pumping energy (for operation and backwash) was determined using Eq. D8. Filter operational head loss uncertainty was accounted for because media depth and water height above media were uncertainty parameters (Table D11) (Kawamura 2000). Typical values were used to estimate the backwash flowrate and pressure (Howe et al. 2012) and ultimately to determine head loss during backwashing; both were uncertainty parameters (Table D11). Other than the 10 minutes of backwash every day, constant filtration was assumed.

$$\mathbf{P} = \frac{(\mathbf{Q}^*\boldsymbol{\rho}^*\mathbf{g}^*\mathbf{H})}{\left(\mathbf{1000}\frac{\mathbf{W}}{\mathbf{kW}}^*\boldsymbol{\eta}\right)} \qquad \textbf{Eq. D8}$$

Where:

P = Power (kW)
Q = Flow rate (m^3/s): water treatment plant flow rate or backwash flow rate
ρ = Density of liquid solution (kg/m^3): 1000 kg/m^3 for water
g = Gravity (9.81 m/s^2)
H = Head loss (m): filter operational head loss or backwash pressure
η = Efficiency (60%)

D4.3 Solids and Chemical Hauling Requirements

The hauling requirements, in tonne kilometers, were determined for solid coagulation waste and all chemicals (Eq. D9). The masses of alum, caustic soda, and chlorine were based on their treatment doses. The hauling distance was assumed to be the same for all chemicals and was an uncertainty parameter (Table D11). The solid waste generated from coagulation and sedimentation was conservatively estimated as the alum mass plus the mass of TOC removed. This waste was hauled to a landfill; the distance was an uncertainty parameter (Table D11).

$$\mathbf{tkm} = \mathbf{M_T}^*\mathbf{L_T} \qquad \textbf{Eq. D9}$$

Where:

tkm = tonne kilometers (tkm)
M_T = Mass of chemicals or solids (tonne)
L_T = Transport Distance (km)

D5. pH Adjustment

The final water's pH was raised to 8.2 at the end of the plant with caustic soda (sodium hydroxide). Similar to the alum dose calculations, the caustic soda dose was determined by generating a caustic soda dose and final pH table (Table D9) for every source water scenario using three main steps. First, the pH after chlorination was calculated using Eq. D10. Second, a comprehensive range of possible caustic doses was generated (from 0 to 2,000 mg/L in 0.1 mg/L increments until 15 mg/L, then 1.0 mg/L increments until 50 mg/L, then 5.0 mg/L increments until 300 mg/L, and then 100 mg/L increments until 2,000 mg/L). Third, the pH of the final adjusted water was calculated by iteratively solving Eq. D11 based on the U.S. EPA's Water Treatment Plant Model v2 (U.S. EPA, 2001). Overall, the required caustic dose for the pH adjustment needed was found from the table. Figure D2 shows an example of how pH changed throughout the treatment train and displays the input and output pH at the point of each chemical

addition. Table D9 shows example caustic doses and the corresponding final water pH (when the starting pH was 7.5).

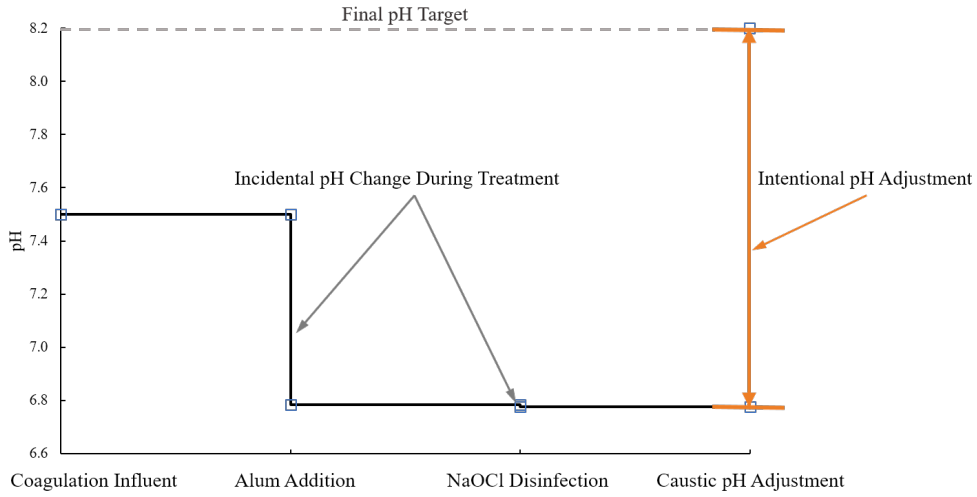

Figure D2. Example pH changes throughout the treatment process. Values were calculated for an example source water (77 mg/L $CaCO_3$, 3.2 mg/L TOC, 7.6 pH, 3.1 SUVA, 15 °C).

Table D9. Example caustic dose and final pH tale. Values were calculated for an example source water representing national averages (77 mg/L $CaCO_3$, 3.2 mg/L TOC, 7.6 pH, 3.1 SUVA, 15 °C).

Caustic Added (mg/L)	Final pH
0	6.78
0.25	6.79
0.5	6.80
0.75	6.81
…	…
13.5	8.2

$$\left((\alpha_1+2)*\alpha_2*\left[CO_3^{2-}\right]\right)+[OH^-]-[H^+]=[HCO_3^-]+2*\left[CO_3^{2-}\right]+[OH^-]-[H^+]-\left(\frac{[Ct_{OCl^-}]}{1+\frac{[H^+]}{k_{ocl^-}}}\right) \quad \textbf{Eq. D10a}$$

$$[Ct_{OCl^-}]=[Ct_{NaOCl}]=\frac{Cl_2\ Dose}{1000\frac{mg}{g}*\left(\frac{(2*35)g\ Cl_2}{1\ mol\ Cl_2}\right)*\left(\frac{1\ mol\ Cl_2}{2\ mol\ NaOCl}\right)} \quad \textbf{Eq. D10b}$$

$$k_{OCl^-}=\exp\left\{\left[\left(\frac{13800\frac{J}{mol}}{8.314\frac{J}{K*mol}}\right)\left(\frac{1}{298.15\ K}-\frac{1}{T}\right)\right]-17.5\right\} \quad \textbf{Eq. D10c}$$

$$\left((\alpha_1+2)*\alpha_2*\left[CO_3^{2-}\right]\right)+[OH^-]-[H^+]=[HCO_3^-]+2*\left[CO_3^{2-}\right]+[OH^-]-[H^+]+[Caustic] \quad \textbf{Eq. D11}$$

Where:

$[OH^-]$ = Concentration of hydroxide (M)
$[H^+]$ = Concentration of hydrogen (M): Known target
$[CO_3^{2-}]$ = Concentration of carbonate (M)
$[HCO_3^-]$ = Concentration of bicarbonate (M)
$[Ct_{NaOCl}]$ = Concentration of sodium hypochlorite added (M) (Eq. D10ab)
$[Ct_{OCl^-}]$ = Concentration of hypochlorite chlorine (M) (Eq. D10ab)
k_{ocl^-} = Hypochlorite equilibrium constant (Stumm & Morgan 1981)
α_1 = Water chemistry equilibrium value for the second hydrogen state (Eq. D1b)
α_2 = Water chemistry equilibrium value for the third hydrogen state (Eq. D1c)
[Caustic] = Amount of caustic added (M)
T = Influent water temperature (K)
$[Cl_2$ Dose] = Required residual chlorine dose (mg Cl_2/L): Using Jones et. al. (Jones et al. 2017)

D6. Unit Cost Estimates

The primary difference between biofiltration and conventional filtration was chemical use, so a simple chemical cost analysis was completed. Yearly chemical costs for each treatment train was based on each chemicals unit cost and the modeled chemical doses. Sodium hypochlorite and caustic soda quantities were adjusted as needed to and from non-diluted solutions (100%) and diluted solutions (e.g., 12%). Table D10 shows the expected unit costs of the three main chemicals (U.S. EPA 2005) and the doses needed for biofiltration and conventional filtration, calculated for typical scenario.

Table D10. Chemical cost data. Values were calculated for an example source water representing national averages (77 mg/L $CaCO_3$, 3.2 mg/L TOC, 7.6 pH, 3.1 SUVA, 15 °C).

Chemical	**Unit Cost**(U.S. EPA, 2005) **($/ton)**	**Biofiltration (ton/year)**	**Conventional Filtration (ton/year)**
Alum	300	20	36
Chlorine	1,100	1.5	1.5
Caustic	350	11	15

D7. Uncertainty and Sensitivity Analysis

Table D11. The low (L) and high (H) values of each uncertainty parameter. The base case (B) value represents the most likely or most typical value expected based on the range. All parameters were characterized with a uniform probability distribution except "Minimum Allowable Alum Dose (mg/L)" and "15°C Biofilter TOC percent removal."

#	Uncertainty Parameter	Low Value	High Value	Base Case	Basis and Citations
1	Minimum Allowable Alum Dose (mg/L)	11	30	18	25th (L), 75th (H), and 50th (B) percentiles of U.S. values (Randtke et al., 1994)
2	15°C Biofilter TOC percent removal	5%	20%	12%	(Terry & Summers 2017)
3	Steel Tank Thickness (m)	0.14	0.55	0.27	L/H (Colorado Department of Public

					Health and Environment and Department of Civil and Environmental Engineering Colorado, 2014), B=Average
4	Steel Life Expectancy (yr)	30	60	45	L/H (U.S. EPA, 2003), B (U.S. EPA, 2004)
5	Backwash Flowrate ($m^3/h/m^2$)	30	60	50	L/H (Howe et al., 2012), B=expert judgment
6	Backwash Pressure (m)	8	10	9	L/H (Howe et al., 2012), B=average
7	Water Height Above Media (m)	1.5	2.5	2	L/H (Kawamura, 2000), B=average
8	Media Lifetime (yr)	15	25	20	expert judgment
9	Hydraulic Loading Rate (m/h)	10	25	15	L/H (Kawamura, 2000), B=expert judgment
10	Hydraulic Loading Rate Ratio (Biofiltration/Conventional Filtration)	0.5	1.0	0.75	Expert judgment
11	Anthracite Depth (m)	0.405	0.5	0.45	B= 0.45 (Kawamura, 2000), L/H = ±10% of B
12	Sand Depth (m)	0.27	0.33	0.3	B= 0.3 (Kawamura, 2000), L/H = ±10% of B
13	Chlorine Storage Tank Lifetime (yr)	30	35	30	L/H (U.S. EPA, 2003), B(U.S. EPA, 2004)
14	Chlorine Delivery Rate (trips/week)	0.5	2	1	L=every other week, H=twice a week, B=weekly
15	Chlorine Pump Head (m)	1.22	70.3	70.3	L/H (Neptune Chemical Pump Company, 2010), B=conservative
16	Concrete Basin Base Thickness (m)	0.30	0.61	0.46	L/H (ADOT, 2006), B=average
17	Concrete Basin Wall Thickness (m)	0.23	0.46	0.46	L/H (ADOT, 2006), B=same as base thickness
18	Baffling Factor for Tank with 2 Baffles	0.3	0.5	0.4	L/H (Colorado Department of Public

					Health and Environment and Department of Civil and Environmental Engineering Colorado, 2014), B=average
19	Baffle Thickness (cm)	3.8	4.5	4.5	L (Meurer Research, 2016), H= (Taylor et al., 2015)(Fig. 1), B=conservative
20	Steel Baffle Life Expectancy (yr)	30	60	45	L/H (U.S. EPA, 2003), B(U.S. EPA, 2004)
21	Concrete Life Expectancy (yr)	30	60	30	L/H (U.S. EPA, 2003), B (U.S. EPA, 2004)
22	Chlorine Storage Tank Thickness (cm)	1.3	5.1	3.2	L/H (Colorado Department of Public Health and Environment and Department of Civil and Environmental Engineering Colorado, 2014), B=average
23	Landfill Hauling Distance (km)	20	100	20	L (Thompson et al. 2016)
24	Chemical Hauling Distance (km)	20	100	20	L (Thompson et al. 2016)

Table D12. Alum dose data of U.S. water treatment plants (Randtke et al., 1994), which was used to represent plants dosing solely for turbidity.

Chemical	Quartile Dosage (n=193)				
	0^{th}	25^{th}	50^{th}	75^{th}	100^{th}
Alum (mg/L)	0.8	11	18	30	150

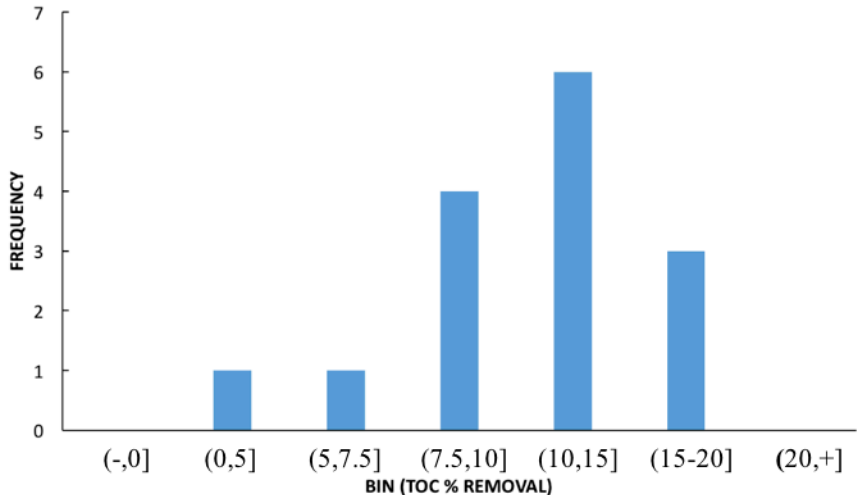

Figure D3. TOC percent removal data from biofiltration literature (Terry & Summers, 2017).

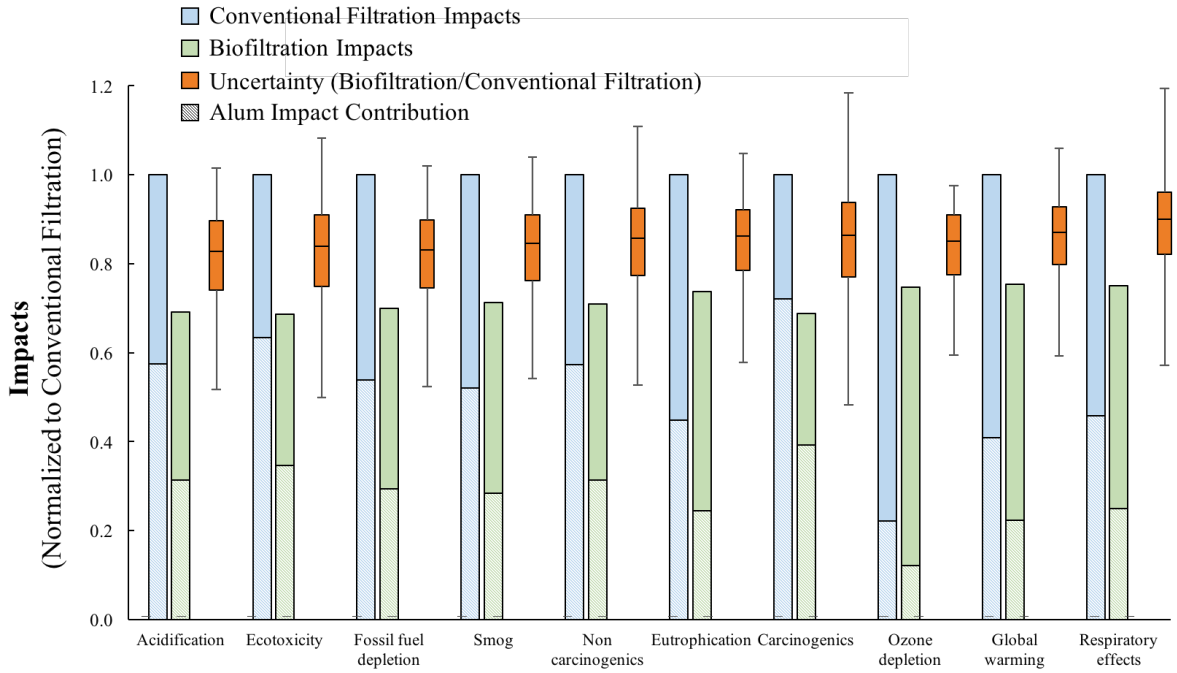

Figure D4. Same as Figure 6.2 in the main paper, except Biological TOC removal percent and minimum allowable alum dosing for turbidity were given uniform distributions instead of triangular distributions. The distribution selections had no notable changes in results.

D8. Results

D8.1 Average Source Water Scenario Process Contribution

a)

b)

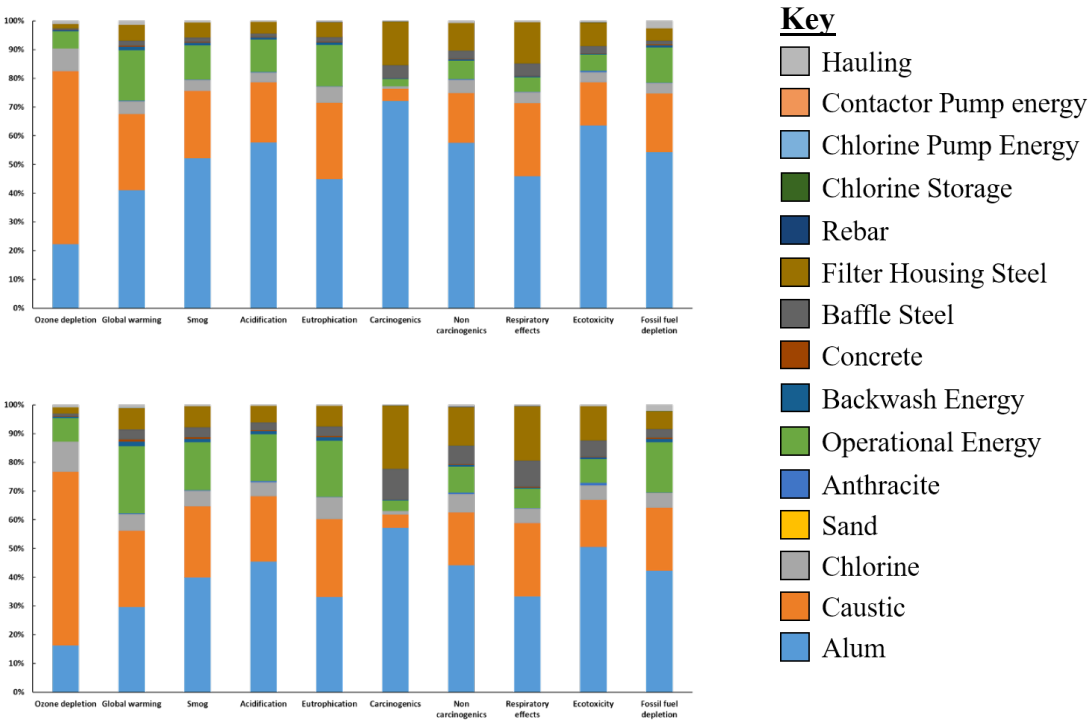

Figure D5. Impact breakdown for (a) conventional filtration and (b) biofiltration for the typical source water scenario (from Figure 6.2).

D8.2 Source Water Scenarios

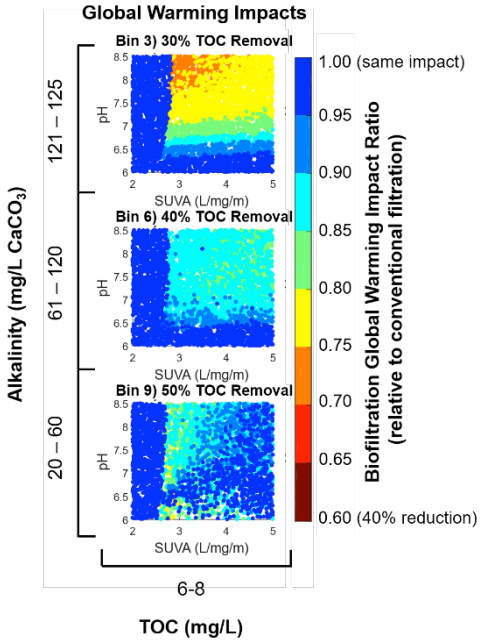

Figure D6. High TOC range (8 to 10 mg/L TOC) bins (excluded from Figure 6.3).

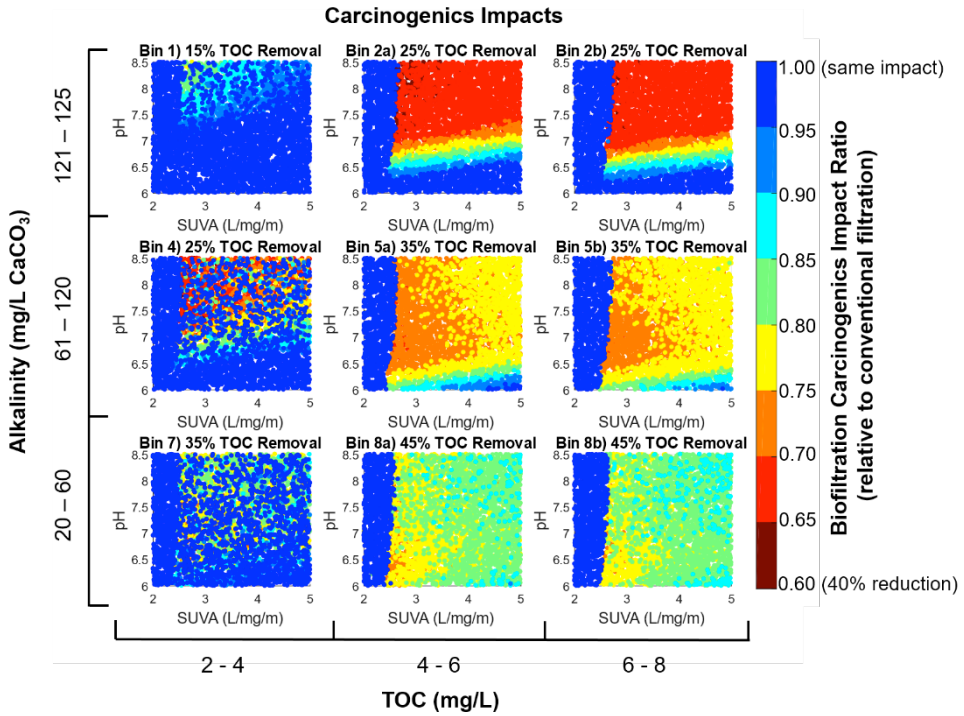

Figure D7. Biofiltration carcinogenics impact ratio, relative to conventional filtration. Note that caustic contributes less to carcinogenics than to global warming, which is why alum trends were more dominant and clear in the carcinogenics impacts. Global warming was most different when compared to the carcinogenics impact category, which had a larger contribution from alum and steel production and lower contribution from caustic soda production than the other categories (Figure D4).

D8.3 Caustic Dose

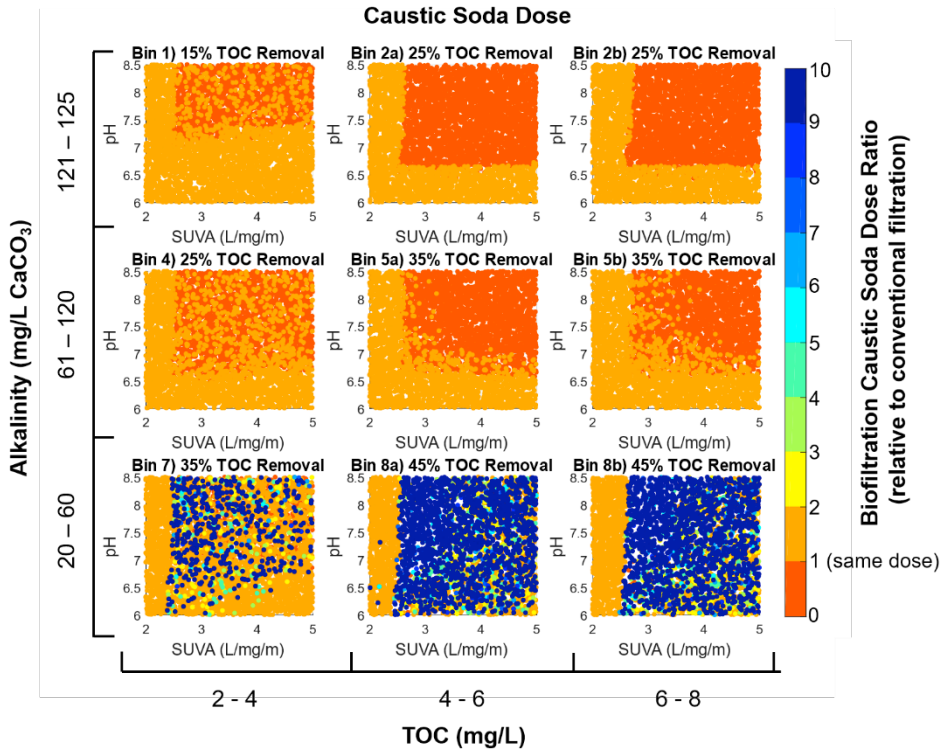

Figure D8. Biofiltration caustic soda dose compared to conventional filtration caustic dose for all 15°C scenarios.

D8.4 pH Trend Analysis

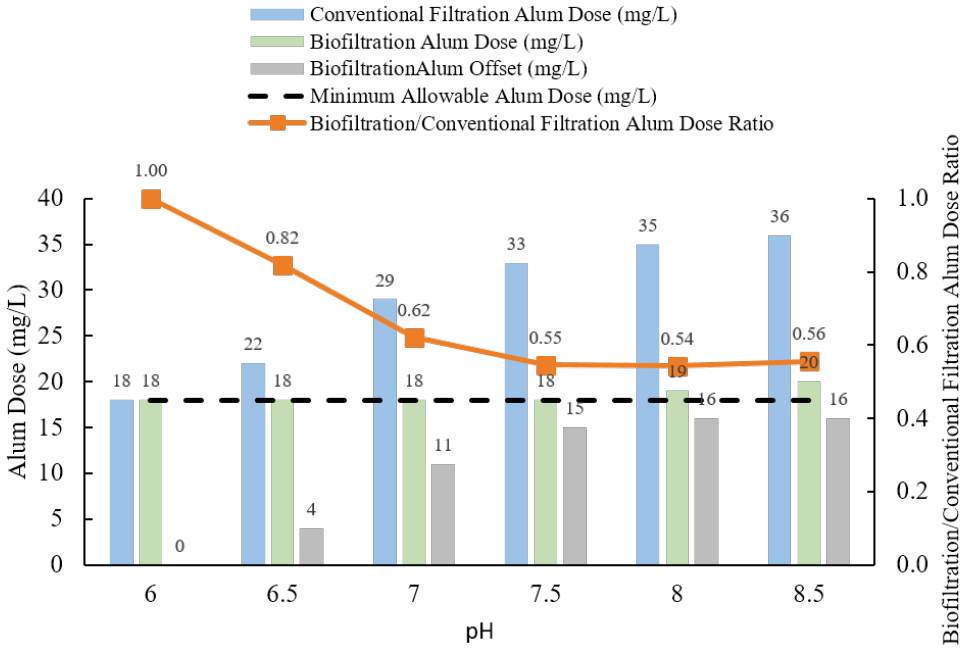

Figure D9. Alum dose and pH trends for the following water quality scenario: alkalinity of 80 mg/L $CaCO_3$, SUVA of 2.75 L/m/mg, temperature of 20°C, TOC of 5 mg/L, and 45% TOC removal required.

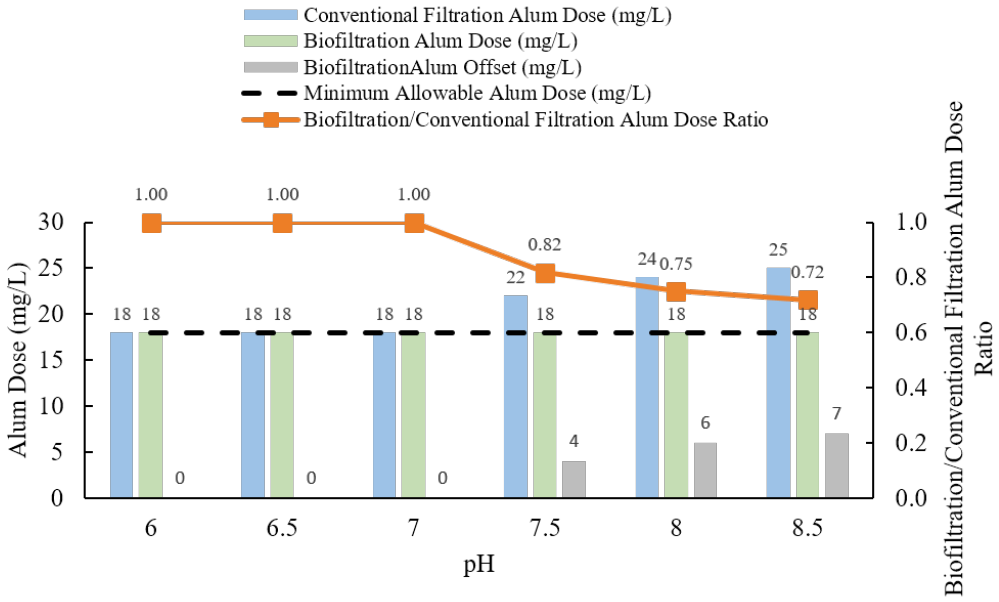

Figure D10. Alum dose and pH trends for the following water quality scenario: alkalinity of 121 mg/L $CaCO_3$, SUVA of 2.75 L/m/mg, temperature of 20°C, TOC of 5 mg/L, and 45% TOC removal required.

D8.5 Minimum Allowable Alum Dose Sensitivity

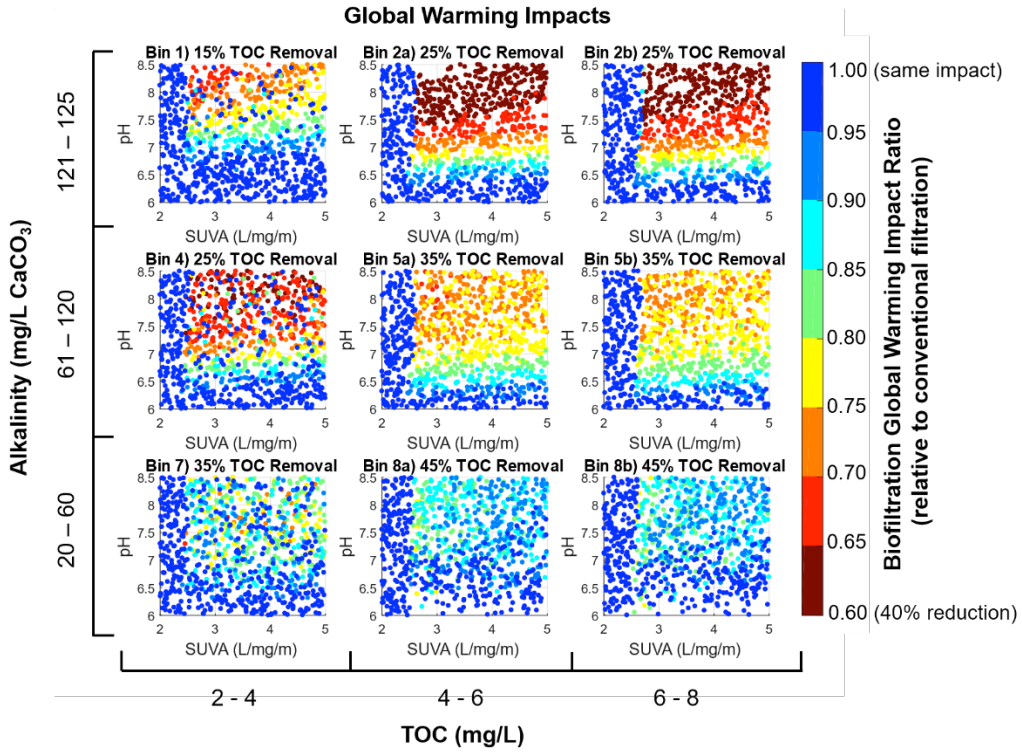

Figure D11. Figure 6.3 from main paper with an 11 mg/L minimum allowable alum dose.

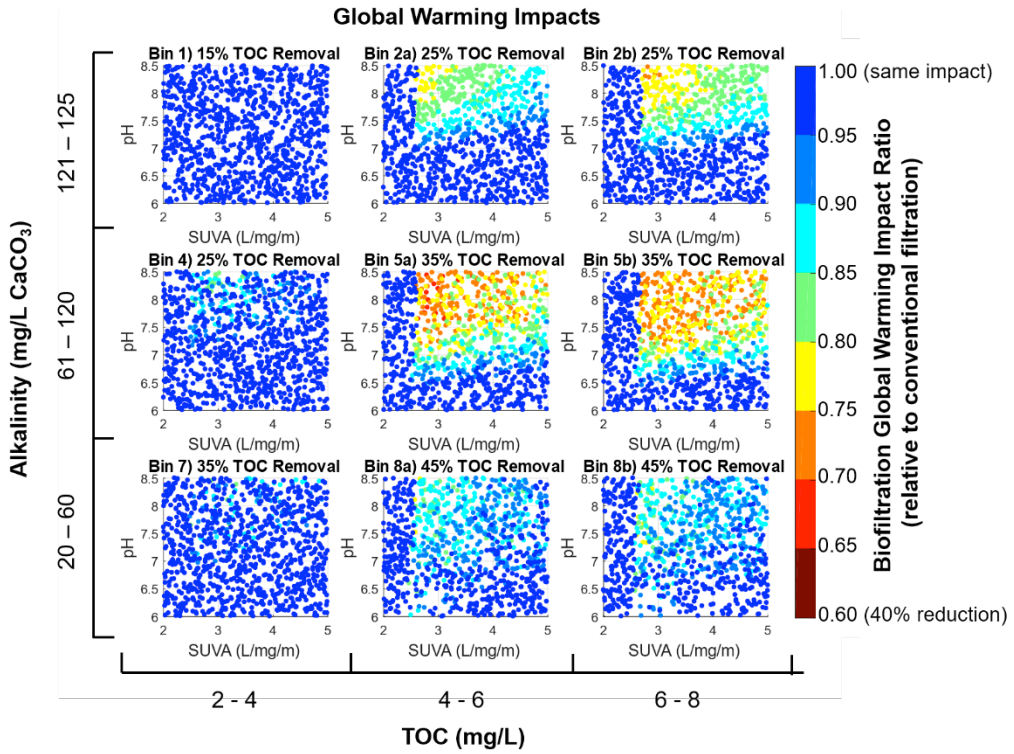

Figure D12. Figure 6.3 from main paper with a 30 mg/L minimum allowable alum dose.

D8.6 Biofilter Temperature and Percent TOC Removal Sensitivity

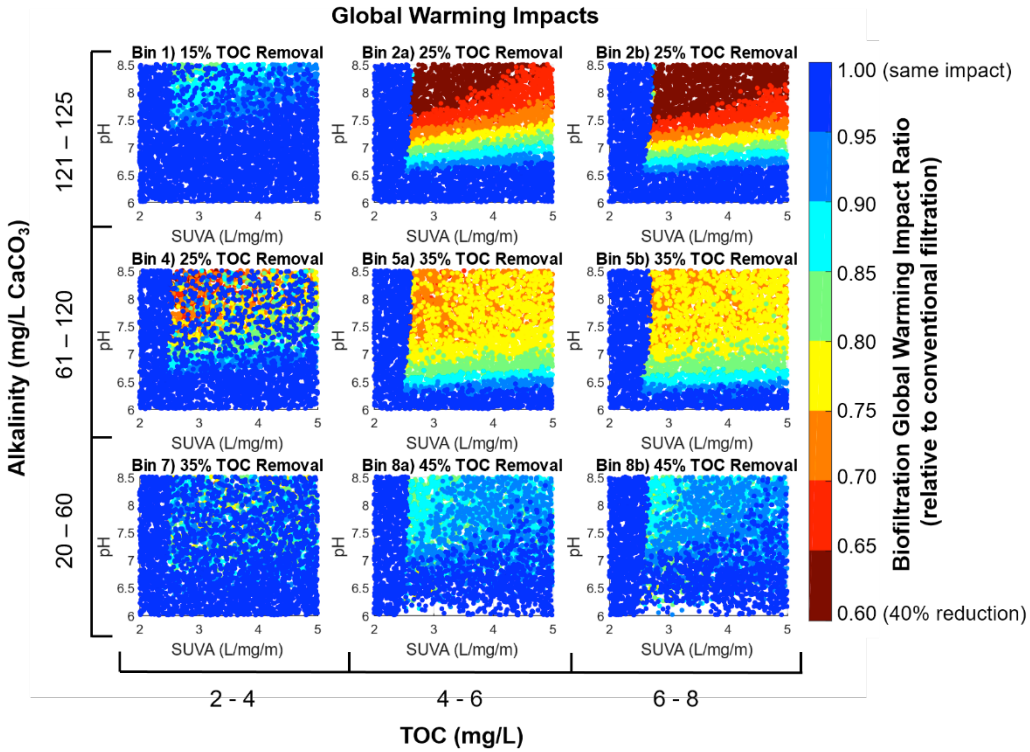

Figure D13. 15% biofilter TOC removal, corresponding to performance expected at 20°C or above.

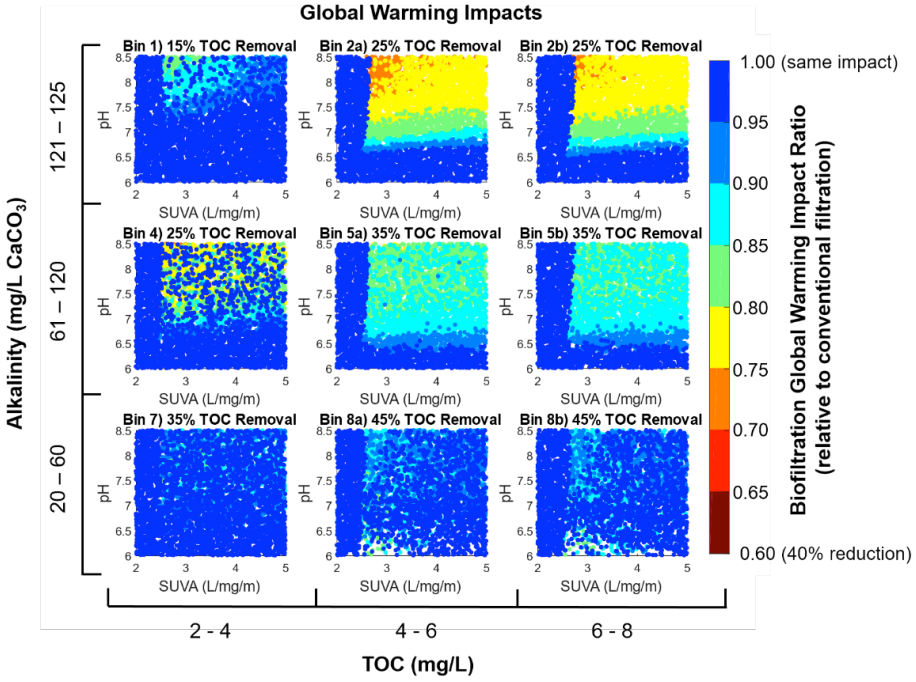

Figure D14. 10% biofilter TOC removal, corresponding to performance expected at or below 10°C.