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Conventional drinking water treatment and direct biofiltration for the removal of pharmaceuticals and artificial sweeteners: A pilot-scale approach

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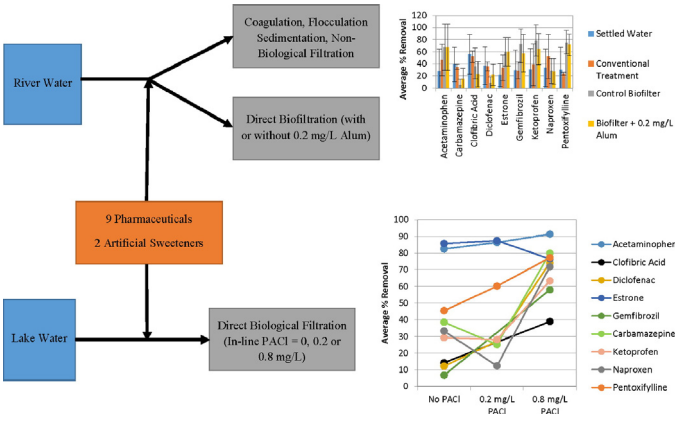
HIGHLIGHTS

- Pilot plants treating Otonabee River and Lake Ontario, Canada water were utilized.
- 9 pharmaceuticals and 2 artificial sweeteners were spiked into the pilot systems.
- Conventional treatment and direct biofiltration were examined for compound removal.
- Coagulation and/or biofiltration removed at least 7 of 9 pharmaceuticals by >50%.
- Sweetener removal increased with increasing concentrations of in-line PACl.

GRAPHICAL ABSTRACT



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ABSTRACT

The presence of endocrine disrupting compounds (EDCs), pharmaceutically active compounds (PhACs) and artificial sweeteners are of concern to water providers because they may be incompletely removed by wastewater treatment processes and they pose an unknown risk to consumers due to long-term consumption of low concentrations of these compounds. This study utilized pilot-scale conventional and biological drinking water treatment processes to assess the removal of nine PhACs and EDCs, and two artificial sweeteners. Conventional treatment (coagulation, flocculation, settling, non-biological dual-media filtration) was compared to biofilters with or without the addition of in-line coagulant (0.2–0.8 mg Al^{3+}/L; alum or PACl). A combination of biofiltration, with or without in-line alum, and conventional filtration was able to reduce 7 of the 9 PhACs and EDCs by more than 50% from river water while artificial sweeteners were inconsistently removed by conventional treatment or biofiltration. Increasing doses of PACl from 0 to 0.8 mg/L resulted in average removals of PhACs, EDCs increasing from 39 to 70% and artificial sweeteners removal increasing from ~15% to ~35% in lake water. These results suggest that a combination of biological, chemical and physical treatment can be applied to effectively reduce the concentration of EDCs, PhACs, and artificial sweeteners.

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1. Introduction

Anthropogenic contaminants of interest in drinking water include pharmaceutically active compounds (PhACs), endocrine disrupting compounds (EDCs) and artificial sweeteners that are poorly removed by wastewater treatment facilities (Ternes et al., 2004). EDCs interfere by mimicking or blocking natural hormones (USEPA, 2001) and PhACs are used to diagnose, treat, alter, or prevent illness (USEPA, 2012). These compounds are of interest because of their potential impacts to the natural environment, the unknown risk they pose to human consumers when consumed at trace levels for long periods of time (Safe, 2004; Schwab et al., 2005), and their ability to indicate wastewater contamination.

Artificial sweeteners are used to sweeten foods and beverages (Scheurer et al., 2010), and though they have been approved by government health organizations worldwide, concern exists because long-term health impacts are presently unknown (Mawhinney et al., 2011). Artificial sweeteners have been proposed as an indicator for wastewater in drinking water sources due to the fact that they are poorly removed by traditional waste and drinking water treatment processes and are only present in water sources impacted by anthropogenic activities (Torres et al., 2011; Oppenheimer et al., 2011).

EDCs, PhACs, and artificial sweeteners have been detected in source waters around the world and many studies have examined their removal using a range of water treatment processes. Ozonation has been demonstrated to be an effective method of reducing the concentration of EDCs and PhACs, but may create unknown degradation products (Westerhoff et al., 2005; Dodd et al., 2009). Coagulation has been examined for the removal of PhACs and EDCs; however removals are typically low (<30%) (Diemert and Andrews, 2013). Biologically active filtration is utilized in drinking water treatment facilities because of its ability to degrade organic compounds while providing effective physical removal (LeChevallier et al., 1992). Biofiltration is most commonly used in drinking water treatment to remove organic carbon and disinfection by-product precursors (Onstad et al., 2008; Chu et al., 2012). Recently, biofiltration has been shown to remove EDCs, PhACs (Reungoat et al., 2011), and artificial sweeteners (Mawhinney et al., 2011) from municipal wastewater and drinking water (Zearly and Summers, 2012). Biofilters may also be enhanced with low doses of in-line coagulant to combine physical, chemical and biological processes to improve removal of large organic compounds without adversely impacting headloss (Azzeh et al., 2015).

This study examined the removal of 9 EDCs and PhACs, as well as the artificial sweeteners sucralose and acesulfame potassium (acesulfame-K), through pilot-scale conventional treatment (coagulation, flocculation, settling, non-biological filtration) and biofiltration (with or without coagulant enhancement). The objective of the study was to quantify the removal of these compounds from drinking water, and to determine which treatment processes are most effective.

2. Materials and methods

2.1. Compounds of interest

Compounds monitored in this study were selected due to their occurrence in the natural environment and to examine a range of physical and chemical properties including: hydrophobicity, solubility, molecular weight, and acidity (Supplemental information Table S1). Generally, smaller and more hydrophilic compounds (low molecular weight, low log K_{ow} and high solubility) have been reported to be more biodegradable (Kickham et al., 2012). Notably absent from this group of analytes are antibiotics, such as sulfamethazine and sulfamethoxazole, due to the negative impact they may pose on growth within biological filters. Antibiotics are designed to prevent the growth of bacteria, and exposing drinking water biofilters to antibiotics may impair their ability to degrade other organics. If the biofilm is too thin it may be unable to prevent the migration of antibiotics to active bacteria (Cochran et al., 2000), or increased growth rates within the biofilm due to warmer water temperatures may make the bacteria more susceptible to the impact of antibiotic exposure (Brown et al., 1988). Based on these factors, antibiotics were omitted from the list of assessed compounds to ensure that the biofilm was not negatively impacted for other studies being completed at these locations.

2.2. Source waters

Pilot-scale studies were completed at the Peterborough Water Treatment Plant (Otonabee River), Peterborough, Ontario, and the R. C. Harris Water Treatment Plant (Lake Ontario), Toronto, Ontario. Pilot plant influent water quality is shown in Supplemental information Table S2.

These two waters can be differentiated by higher temperatures and organic concentrations in the Otonabee River, while both locations have similar pH and raw water turbidity.

2.3. Pilot plant configurations

The pilot using Otonabee River water as its source was configured to examine biofiltration with or without in-line alum addition (0.2 mg Al^{3+}/L), as well as non-biological, conventional filtration (Fig. 1a). Two biofilters were operated with an empty bed contact time (EBCT) of 10 min to meet the requirements of a concurrent study, while the conventional filter was operated at 15 min to match full-scale operation. All filters contained 50 cm of anthracite (effective size d_{10} = 0.85 mm, uniformity coefficient UC = 1.8) over 50 cm of sand (d_{10} = 0.5 mm, UC = 1.8). The conventional treatment train consisted of alum coagulation (3.0–5.0 mg Al^{3+}/L to match full-scale), three-stage tapered flocculation, parallel plate settling, and non-biological filtration. One biofilter was operated without chemical addition and is referred to as a control. The second biofilter was pretreated with 0.2 mg Al^{3+}/L inline alum. The biofilters were backwashed with their own unchlorinated effluent while the conventional filter was backwashed with chlorinated water (~1 mg/L) from the full-scale clearwell. All filters were backwashed three times per week. Analytes of interest were spiked directly into an influent constant head tank and completely mixed prior to treatment.

A second pilot plant, illustrated in Fig. 1b, consisted of three filters utilizing ozonated Lake Ontario water (dose = 1 mg/L, contact time = 8 min, O_3:DOC = 1:2), and was used to examine the impact of polyaluminum hydroxychloride (PACl) dose on biofiltration. One of the filters acted as a control, with no chemical addition, while the other two filters received 0.2 and 0.8 mg Al^{3+}/L PACl, respectively. The biofilters consisted of 50–150 cm GAC over 15–50 cm of sand and were operated with a 16 min EBCT to match the typical full-scale flow rate. All media had been in operation for at least 4 years prior to sampling and was considered to be exhausted in terms of adsorption capacity. All filters were backwashed with their own unchlorinated effluent, at most once per week. EDCs and PhACs were dosed immediately following ozone quenching in order to isolate their removal within the filters and eliminate the impact of oxidation. At the Lake Ontario pilot, analytes were spiked into a constant head tank immediately prior to the filters.

To determine if any analyte losses were occurring as a result of contact with plumbing materials, a filter column was operated without media. Results showed that losses within the pilot plant were always <12%.

2.4. Analyte spiking procedure

EDCs and PhACs were dissolved in acetonitrile as per Diemert and Andrews (2013), and spiked into biofilter influent water to achieve a nominal concentration of 500 ng/L. Artificial sweeteners were dissolved in Milli-Q® water (1000 ng/L). Spiking began 7 days prior to sample