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Predicting trace organic compound attenuation with spectroscopic parameters in powdered activated carbon processes

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HIGHLIGHTS

- Powdered activated carbon (PAC) efficiency predicted by UV absorption and fluorescence.
- PAC is highly effective for attenuation of trace organic compounds in wastewater.
- Chick-Watson CT model was useful for trace organic compound attenuation prediction.
- UV/FL removal were well correlated with trace organic compound attenuation.
- UV/FL removal has potential for on-line monitoring and process control strategies.

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ABSTRACT

The removal of trace organic compounds (TOrCs) is of growing interest in water research and society. Powdered activated carbon (PAC) has been proven to be an effective method of removal for TOrCs in water, with the degree of effectiveness depending on dosage, contact time, and activated carbon type. In this study, the attenuation of TOrCs in three different secondary wastewater effluents using four PAC materials was studied in order to elucidate the effectiveness and efficacy of PAC for TOrC removal. With the notable exception of hydrochlorothiazide, all 14 TOrC indicators tested in this study exhibited a positive correlation of removal rate with their log D_{ow} values, demonstrating that the main adsorption mechanism was hydrophobic interaction. As a predictive model, the modified Chick-Watson model, often used for the prediction of microorganism inactivation by disinfectants, was applied. The applied model exhibited good predictive power for TOrC attenuation by PAC in wastewater. In addition, surrogate models based upon spectroscopic measurements including UV absorbance at 254 nm and total fluorescence were applied to predict TOrC removal by PAC. The surrogate model was found to provide an excellent prediction of TOrC attenuation for all combinations of water quality and PAC type included in this study. The success of spectrometric parameters as surrogates in predicting TOrC attenuation by PAC are particularly useful because of their potential application in real-time on-line sensor monitoring and process control at full-scale water treatment plants, which could lead to significantly reduced operator response times and PAC operational optimization.

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1. Introduction

Trace organic compounds (TOrCs) comprising of pharmaceuticals, personal-care products, industrial constituents, and others are commonly detected in the aquatic environment and drinking water sources (Focazio et al., 2008; Benotti et al., 2009; Scott et al., 2014). Wastewater discharge has been identified as a primary source of entry due to inefficient removal of TOrCs by conventional water treatment processes (Ternes et al., 2002; Stackelberg et al., 2007; Anumol et al., 2013; Kostich et al., 2014). While some of these compounds have been implicated in causing adverse effects in wildlife, toxicological data for the vast number of TOrCs is currently unavailable and mixture effects on humans and the environment are greatly lacking (Sumpter, 2009; Snyder, 2014). With the increased focus on implementing potable water reuse schemes around the world, there is a greater possibility of these wastewater

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derived contaminants entering drinking water sources. Hence, it is prudent to monitor key indicator TOrCs and determine suitable treatment and remediation strategies for broader classes of organic contaminants.

Advanced treatment technologies have been implemented in water reuse systems as an effective means to attenuate TOrCs. Advanced oxidation processes (AOPs) like ozone and UV with addition of hydrogen peroxide and other oxidants has proven to remove many TOrCs present in municipal wastewater (Wert et al., 2009; Wols et al., 2013; Park et al., 2015). However, studies also have shown that these AOPs result in transformation products, some of which are more toxic than the precursor compounds (Rizzo, 2011; Jia et al., 2015). Adsorption processes like granular activated carbon (GAC) and powdered activated carbon (PAC) are capable of removing significant amounts of TOrCs from water without forming byproducts (Snyder et al., 2006; Redding et al., 2009; Altmann et al., 2014). The removal efficacy of TOrCs by adsorption processes depends on type of adsorptive material, the contact time, water quality, and the specific molecular properties of the TOrC species evaluated. GAC adsorption is an efficient and commonly used treatment process; however, PAC has some advantages, particularly lower capital cost and greater flexibility in dosing (i.e., seasonal application). PAC materials also typically have smaller particles sizes than GAC and hence have faster kinetics of adsorption (Nowotny et al., 2007).

With several thousands of chemicals being released into the environment every day and toxicological data unavailable for a lot of them, it is not feasible to monitor each one individually. The use of indicator compounds that mimic the behavior of wider groups of chemicals is now prevalent (Merel et al., 2015). However, measuring TOrCs still requires sophisticated analytical equipment, highly trained technicians, and often requires days to weeks to receive results. Hence the use of surrogate parameters that can accurately predict the removal of TOrCs in a simple, fast, and cost-effective manner is highly desirable. Spectrophotometric parameters like UV absorbance and fluorescence have been shown to accurately predict formation of disinfection byproducts and removal of TOrCs through various disinfection and oxidation processes used during water treatment (Roccaro and Vagliasindi, 2010; Gerrity et al., 2012; Sgroi et al., 2016). Further, UV absorbance at 254 nm wavelength (UV_{254}) has been successfully demonstrated as an efficient surrogate in predicting TOrC removal by PAC and GAC processes (Zietzschmann et al., 2014; Anumol et al., 2015a). The use of spectrophotometric parameters and other bulk organic parameters as surrogates allows the potential for real-time, on-line monitoring of water treatment plant efficiency while also predicting specific TOrC attenuation through different treatment processes (Roccaro et al., 2015; Yu et al., 2015).

The present study investigated the efficiency of PAC in removal of TOrCs from three different wastewater treatment plants and developed a simple model to predict TOrC removal by PAC irrespective of water quality. The application of UV_{254} absorbance and fluorescence as surrogates for TOrC removal prediction by PAC was demonstrated. To the authors' knowledge, this is the first study to investigate total fluorescence as a potential surrogate parameter in predicting TOrC removal in a PAC process.

2. Materials and methods

2.1. Sample collection and adsorption test

Secondary effluent samples from three different wastewater treatment plants (WWTP) were collected and filtered using glass fiber filters (0.7 μm pore size, Whatman GF/F). Four powdered activated carbons (PAC) were used in this study namely Cabot 20BF and HDB (Boston, MA, USA), and Calgon PWA and WPH (Pittsburgh, PA, USA). The combination of various PAC doses (0, 5, 20, 50 and 100 mg/L) and contact times (0, 5, 15 and 30 min) were applied for the batch adsorption experiment with each activated carbon. A 1.5 L WWTP secondary effluent sample was placed in a jar and mixed at 300 rpm using Phipps & Bird PB900 programmable jar tester after dosing the activated carbon. At the desired contact time, 20 mL samples were collected and filtered using a syringe filter with 0.45 μm pore size (PVDF Millex-HV, Billerica, MA, USA). The filtered water samples were placed in amber glass scintillation vials and refrigerated at 4 °C until chemical analyses.

2.2. Analysis

Dissolved organic carbon (DOC) was measured using a Shimadzu TOC analyzer (Japan). All DOC samples were filtered before analysis using 0.45 μM syringe filters (PVDF Millex-HV, Billerica, MA, USA) and acidified with hydrochloric acid (37%, ACS grade, Sigma-Aldrich) to a pH below 1.5. UV absorbance and fluorescence spectra were measured using a Horiba Aqualog fluorometer (Kyoto, Japan). UV absorbance was scanned from the wavelength of 200 nm to 800 nm with 4 nm intervals and UV absorbance at 254 nm was extracted from the scanned spectra. Fluorescence intensity was measured by scanning the excitation wavelength from 200 nm to 450 nm with the interval of 5 nm and emission wavelength from 220 nm to 580 nm with the interval of 1 nm. Inner filter effect of fluorescence spectra was corrected using the scanned UV–visible spectra (Lakowicz, 2006). Fluorescence intensity was normalized by the area of Raman scattering peak of deionized water (Lawaetz and Stedmon, 2009). Fluorescence excitation emission matrices (F-EEMs) were produced for each sample as described in earlier research (Anumol et al., 2015a). Total fluorescence was calculated by integration of fluorescence spectra under the scanning excitation and emission wavelength range (Cheng et al., 2003b).

The fourteen TOrCs studied (Table 1) were analyzed using direct aqueous large volume injection (80 μL) onto an ultra-high performance liquid chromatography (LC) system coupled to a tandem mass spectrometer (MS/MS). Isotope-dilution was used for sample quantification by adding a known mass of stable isotopically-labeled versions of each substance to each sample prior to analysis (Vanderford and Snyder, 2006). An Agilent 1260 binary LC pump equipped with a Pursuit XRs C-8 column (100 mm × 2.0 mm, 3 μm) was used for analyte separation. Mass spectrometry was performed with an Agilent 6490 MS system and an electrospray ionization source equipped with Agilent jet stream technology for enhanced sensitivity. Method optimization parameters, compound transitions and relevant QA/QC has been published in previous work (Anumol et al., 2015b). All data analysis and processing was done using the Agilent MassHunter software (Ver. 6.00).

3. Results and discussion

3.1. Attenuation of TOrCs by PAC

In general, the main adsorptive interaction between organic molecules and activated carbon is hydrophobic interaction (Snyder et al., 2006; Altmann et al., 2014). While octanol-water partition coefficients (i.e., Log K_{ow}) can be indicative of hydrophobicity for organic molecules including TOrCs, natural water and wastewater effluent have pH ranges that generally fall in a neutral range (6.5–8), thereby hydrophobicity can be greatly influenced by ionization of weak acids and bases. Hence, log D_{ow} (octanol-water partition coefficient at a given pH) was considered in order to examine the relation of removal rate of TOrCs with hydrophobicity

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