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Predicting the occurrence of chemicals of emerging concern in surface water and sediment across the U.S. portion of the Great Lakes Basin

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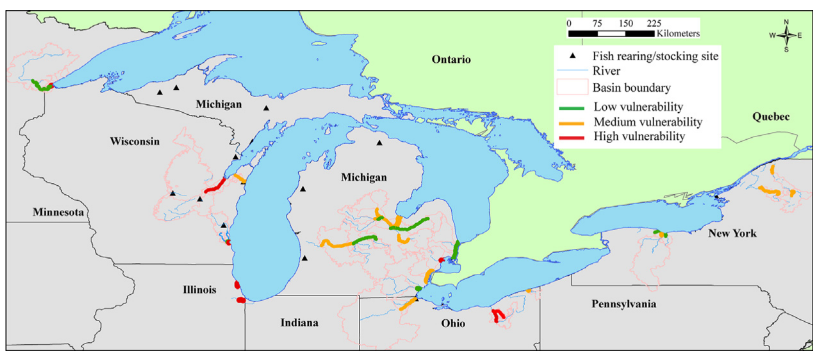
HIGHLIGHTS

- Boosted regression tree models predict the probability of CEC class occurrence.
- Upstream buffer land use/land cover variables are important components of all models.
- Upstream sources of contamination are important components of surface water models.
- CEC class models are unique combinations of predictor variables.
- CEC class models for water and sediment are dissimilar.

GRAPHICAL ABSTRACT



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ABSTRACT

Chemicals of emerging concern (CECs) are introduced into the aquatic environment via various sources, posing a potential risk to aquatic organisms. Previous studies have identified relationships between the presence of CECs in water and broad-scale watershed characteristics. However, relationships between the presence of CECs and source-related watershed characteristics have not been explored across the Great Lakes basin. Boosted regression tree (BRT) analyses were used to develop predictive models of CEC occurrence in water and sediment throughout 24 U.S. tributaries to the Great Lakes. Models were based on the distribution of both broad-scale and source-related watershed characteristics. Twenty-one upstream watershed characteristics, including land cover, number of permitted point sources, and distance to point sources were used to develop models predicting the probability of CEC occurrence in surface water and bottom sediment. Total accuracy of BRT models ranged from 66% to 94% for both matrices. All 21 watershed characteristics were important predictor variables in at least one surface-water model; twenty were important in at least one bottom-sediment model. Among the model variables, developed land use and distance to point sources were important predictors of the presence of CEC classes in both water and sediment. Although limitations exist, BRT models are one tool available for assessing vulnerability of fisheries and aquatic resources to CEC occurrences.

1. Introduction

Over the past twenty years, national reconnaissance studies in the United States have confirmed the presence of anthropogenic organic

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contaminants in the environment, including many compounds commonly referred to as chemicals of emerging concern (CEC; Kolpin et al., 2002, Focazio et al., 2008, Furlong et al., 2017, Bernot et al., 2016, Glassmeyer et al., 2017). Commonly reported CECs include hormones, pharmaceuticals, plasticizers, alkylphenols, current-use pesticides, fragrances, and flame retardants. Chemicals from these classes have been detected in several types of discharge and surface waters including urban stormwater runoff (Kolpin et al., 2004; Fairbairn et al., 2018), agricultural runoff (Gray et al., 2017) streams and rivers (Kolpin et al., 2002; Lee et al., 2010; Bradley et al., 2017), and lakes (Writer et al., 2010; Ferrey et al., 2015). More recent studies have detected CECs in source drinking water (Furlong et al., 2017; Glassmeyer et al., 2017) and in shallow groundwater influenced by on-site treatment systems (Erickson et al., 2014).

Despite more than two decades of research into the presence of CECs in various environmental media, many questions remain regarding the relationship between point or non-point sources of CECs and detections of CEC classes in the environment. Discharges from point sources and associated waste-water collection systems [e.g. wastewater treatment plants (WWTPs), combined sewer overflows (CSOs), etc.] have been characterized for CEC composition. For example, WWTP effluents tend to have strong chemical signatures indicative of household products such as pharmaceuticals, personal care products, and surfactants (Kolpin et al., 2002; Buerge et al., 2006; Kuster et al., 2008; Ginebreda et al., 2010; Barber et al., 2015). CSO discharges may contain CECs in common with WWTP effluents (e.g., Kostich et al., 2014), but they can also integrate chemicals from the urban landscape. For example, industrial chemicals or polycyclic aromatic hydrocarbons (PAHs; Phillips and Chalmers, 2009) often occur as part of an urban stormwater signature that includes diffuse runoff as well as permitted stormwater outfalls.

In addition to point sources, non-point sources including agricultural runoff and diffuse urban stormwater runoff are potentially important contributors of CECs to the environment. Chemicals such as pesticides, hormones, and veterinary-use pharmaceuticals may be introduced into surface waters via overland runoff (Fairbairn et al., 2016) or through stormwater collection systems (e.g. Fairbairn et al., 2018). Although the specific influence of either point or non-point sources on surface waters may be obvious locally, distinct chemical signatures observed in close proximity to sources can be mixed as they are transported downstream, eventually creating complex mixtures of CECs in receiving waters (e.g. Phillips et al., 2012).

The Laurentian Great Lakes (hereafter 'Great Lakes') represent the largest freshwater supply in the world, providing direct and indirect ecosystem services to over 35 million people in the United States and Canada (NOAA-GLERL, 2018). In addition, the Great Lakes provide habitat for important aquatic organisms such as fish and freshwater mussels along their >16,000 km of shoreline, 240,000 km^2 of surface area, and in the numerous tributary mouths along their coast (NOAA-GLERL, 2018). Based on several assessments or surveys of the Great Lakes, large areas of the lakes are under the influence of moderate to high levels of environmental stressors (Smith et al., 2015). The cumulative stress is highest in areas of the lakes associated with high levels of human activity including urban and agricultural land uses (Allan et al., 2013). These high stress profiles are partially attributed to the enhanced transport by tributaries of chemical loading, including many CECs, from multiple point and non-point sources. Considering that CECs have been observed to adversely affect resident fish at low (nanograms per liter; ng/L) concentrations (e.g., Kidd et al., 2007 for fathead minnows) or at ambient concentrations (e.g., Thomas et al., 2017 for resident sunfish), understanding the factors that influence the presence of CECs in Great Lakes tributaries and river mouths is of primary importance for conserving fish populations and possibly other aquatic organisms.

A previous statistical analysis of the data used in this study was conducted to characterize the occurrence of CECs in water and sediments from U.S. tributaries to the Great Lakes. Results from a two-way cluster analysis identified patterns in chemical distributions dependent on the

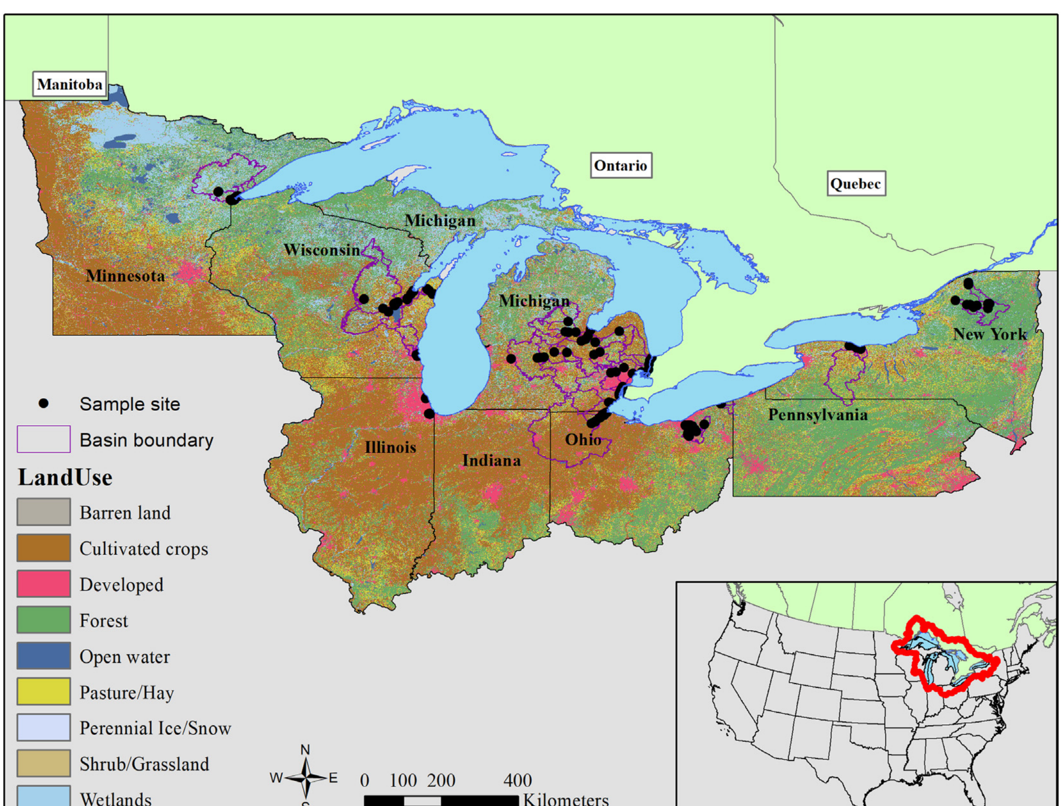

Fig. 1. Map showing location of sampling locations listed in Table S1.