Trace organic contaminant (TOrC) mixtures in Minnesota littoral zones: Effects of on-site wastewater treatment system (OWTS) proximity and biological impact

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HIGHLIGHTS
- OWTSs are a potential diffuse source of TOrCs in Minnesota Lakes.
- TOrC concentrations increase with residential proximity.
- Increased endpoints of endocrine disruption in fish near residences.
- Targeted analysis may limit understanding of mixture bioactivity.

ABSTRACT
On-site wastewater treatment systems (OWTSs) are an international wastewater management strategy for rural and semi-rural communities without access to centralized sewage treatment. These systems are a suspected source of trace organic contaminants (TOrCs) that may be responsible for endocrine disrupting effects to resident fish species in Minnesota Lakes. This study assessed localized porewater concentrations of TOrCs in near-shore environments across five Minnesota Lakes. Sampling sites were designated as either likely (HOME) or unlikely (REF) to receive OWTS discharges based on their proximity to shoreline households. Sampling sites also served as sunfish spawning habitats concurrently studied for biological impacts to resident adult males. Two-group hypothesis tests demonstrated significantly (p = .02) higher total TOrC concentrations in HOME (Mean = 841 ng/L) versus REF (Mean = 222 ng/L) sites. HOME sites also contained a wider suite of TOrC detections relative to REF sites. The distance to the nearest household (most proximal distance; MPD) negatively correlated (r = −0.62) with total TOrC concentrations. However, 2,4-D and DEET were major contributors to these total concentrations, suggesting that anthropogenic influence from households may not be exclusively attributed to OWTS discharges. Further, TOrC presence and elevated nitrogen concentrations in REF site porewater suggest additional, non-household TOrC discharges to these lakes. Significantly higher
Trace organic contaminants (TORCs) represent many emerging contaminants prioritized in current environmental monitoring efforts. TORCs encompass pharmaceuticals, herbicides, pesticides, hormones, steroids, personal care products, cleaning agents, and food preservatives detected at low (ng/L) concentrations throughout the environment (Koutrakis et al., 2010; Kolpin et al., 2002; Lapworth et al., 2012). Many of these chemicals are not currently regulated despite the association of several TORCs with adverse biological impacts, particularly endocrine disrupting effects (Ortiz de García et al., 2014). Endocrine disrupting chemicals mimic or inhibit normal androgen or estrogen receptor function, resulting in abnormal masculinization or feminization of affected species, respectively (Diamanti-Kandarakis et al., 2009; Söfker et al., 2015). The endocrine disruption capabilities of individual TORCs are defined using laboratory exposure experiments through biochemical, histological, and behavioral endpoints (Blair et al., 2000; Elliott et al., 2014; Han et al., 2010; Oropesa et al., 2016). Still, the endocrine activity of environmental TORC mixtures are poorly understood, particularly in light of the likely co-occurrence of unknown TORCs (with unknown biological activities) and the potential for co-contaminants to enhance biological impacts (McCarty and Borgert, 2006).

Minnesota littoral zones are a prime field environment for studying diffuse sources of TORCs and subsequent effects to aquatic life. Previous assessments of nutrient loadings in the United States (US) have led to the consensus that diffuse sources, such as agricultural runoff, groundwater infiltration, and atmospheric deposition, are responsible for most water quality degradation (U.S. Environmental Protection Agency, 1996). The lack of discrete inputs within proximity of surveyed Minnesota water resources indicates diffuse sources are also responsible for widespread TORC occurrence in these waters (Erickson et al., 2014; Ferrey et al., 2015; Writer et al., 2010). In addition, many lakes in the state (90% of those surveyed by Writer et al.) also contain adult male fish with elevated vitellogenin concentrations, a biomarker of fish feminization (Writer et al., 2010). The spatial and seasonal heterogeneity of TORC presence, both across lakes and within the same lake, impedes alignment of current chemical and biological observations (Baker et al., 2014). More strategic sampling methods, specifically sampling porewater in littoral zones (near-shore environments with depth < 5 m) during the spring and summer months should enable better characterization of biologically active TORC mixtures that affect Minnesota fish species. Littoral zones serve as spawning habitats for fish species, such as the commonly studied bluegill sunfish Lepomis macrochirus. Spawning season is a critical time of TORC exposure for larvae and the adult male sunfish that guard them (Becker, 1983). On-site wastewater treatment systems (OWTSs) are one of the proposed diffuse sources affecting the health of these fish (Baker et al., 2014; Writer et al., 2010). Analysis of sediment porewater in these locations advantageously provides insight into the TORC concentrations of inflowing, potentially OWTS-impacted shallow groundwater and relevant exposure concentrations to fish interacting with lake sediments while spawning.

OWTSs are a documented diffuse source of wastewater-derived contaminants in groundwater, drinking water wells, and surface waters around the world (Gago-Ferrero et al., 2017; Godfrey et al., 2007; Phillips et al., 2015; Schäfer et al., 2014; Subedi et al., 2015). This method of treatment typically serves rural and semi-rural populations without access to centralized sewage, around 25% of the population in the United States and 20% of Minnesotans (U.S. Environmental Protection Agency, 2014; West, 2008). Removal of nutrients, suspended solids, and pathogens is achieved by percolating pre-treated wastewater through unsaturated native soils (Crites and Tchobanoglous, 1998; Stanford et al., 2010). TORCs readily sorbed or biotransformed in these subsurface conditions are also effectively attenuated, even though their removal is not considered in OWTS design (Conn et al., 2006, 2010; Teerlink et al., 2012b). Nevertheless, certain TORCs remain recalcitrant to modern wastewater treatment technologies (Du et al., 2014; Wode et al., 2015). For this reason, several TORCs, such as carbamazepine, are now designated as environmental domestic wastewater indicators (Kahle et al., 2009). Furthermore, the ability of OWTSs to effectively treat heterogeneous inputs of TORCs at their small sewershed scale is highly variable (Teerlink et al., 2012a). Out-of-compliance systems, attributed to either improper installation or maintenance, allow insufficiently treated wastewater to reach the water table and enter shallow groundwater flow paths (Bremner and Harter, 2012; Yates, 1985). In addition, cesspool and leach pit OWTS designs have a decreased ability to remove TORCs compared to a conventional two-stage system (Schäfer et al., 2017). Transport through the subsurface poses an environmental health risk, and can lead to diarrhea outbreaks in children consuming water from OWTS-impacted drinking wells (Borchardt et al., 2003). An estimated 21% of OWTSs in Minnesota are operated out of compliance (Robinson and Schultz, 2015), but subsurface clay moraines with low hydraulic conductivity may also compromise soil driven treatment in the region (Engelking and Kovacevic, 2016). While advanced treatment options, such as aerated biofilters, are available, current OWTS regulation does not require their implementation (Jantrania and Gross, 2006). In light of the potential for OWTSs to serve as sources of TORCs to Minnesota waters, it seems prudent to evaluate their occurrence in relation to OWTSs and their potential biological impacts before additional steps are taken to reduce these potential impacts.

The objectives of this study were to characterize targeted TORC mixtures in littoral zones affected by discharges from OWTSs and evaluate potential associations between these TORCs and biological impacts to adult fish. We hypothesized that locations more proximal to shoreline households would have more TORC detections at higher concentrations and these locations would contain sunfish with elevated biomarkers of endocrine disruption. The following research questions were addressed: (1) what TORC mixtures are present at near-shore environments in Minnesota Lakes, (2) are there significant compositional differences between sites likely impacted by OWTSs versus those which likely are not, (3) how are localized environmental TORC mixtures related to biological responses in fish species? To address these questions, targeted aqueous analysis of porewater grab samples from spawning habitats in five Minnesota Lakes were compared to endpoints of biological impact in captured adult male sunfish.

2. Materials and methods

2.1. Site selection

Five lakes were selected for this study: Cedar Lake (Wright County, MN), Franklin Lake (Otter Tail County, MN), Lake Mary (Wright County, MN), Pearl Lake (Stearns County, MN), and Sullivan Lake (Wright County, MN).
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