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Bioaccumulation of perfluorinated carboxylates and sulfonates and polychlorinated biphenyls in laboratory-cultured *Hexagenia* spp., *Lumbriculus variegatus* and *Pimephales promelas* from field-collected sediments

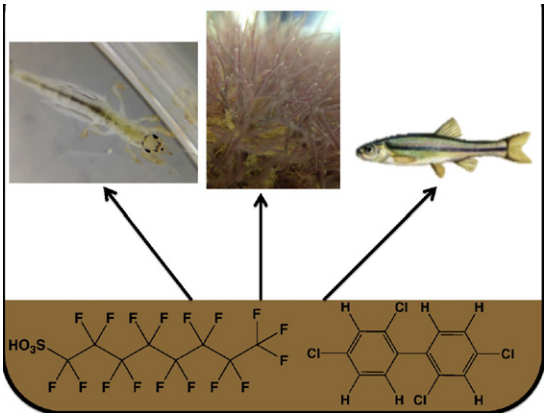
R.S. Prosser [a,*], K. Mahon [b], P.K. Sibley [a], D. Poirier [b], T. Watson-Leung [b]

[a] School of Environmental Sciences, University of Guelph, Guelph, Ontario, Canada
[b] Aquatic Toxicology Unit, Ontario Ministry of the Environment and Climate Change, Toronto, Ontario, Canada

HIGHLIGHTS

- BSAF values for total PCBs and PFOS greatest in *Hexagenia* spp.
- BSAF values for other PFASs not consistently greatest in *Hexagenia* spp.
- Trends in BSAF values for PFASs varied as a function of carbon chain length among species.
- Differences in exposure pathways likely explain variation in accumulation across species.

GRAPHICAL ABSTRACT



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ABSTRACT

Polychlorinated biphenyls (PCBs) and perfluorinated carboxylates and sulfonates (PFASs) are persistent pollutants in sediment that can potentially bioaccumulate in aquatic organisms. The current study investigates variation in the accumulation of PCBs and PFASs in laboratory-cultured *Hexagenia* spp., *Lumbriculus variegatus* and *Pimephales promelas* from contaminated field-collected sediment using 28-day tests. $BSAF^{lipid}$ (lipid-normalized biota-sediment accumulation factor) values for total concentration of PCBs were greater in *Hexagenia* spp. relative to *L. variegatus* and *P. promelas*. The distribution of congeners contributing to the total concentration of PCBs in tissue varied among the three species. Trichlorobiphenyl congeners composed the greatest proportion of the total concentration of PCBs in *L. variegatus* while tetra- and pentabiphenyl congeners dominated in *Hexagenia* spp. and *P. promelas*. Perfluorooctane sulfonate (PFOS) was present in all three species at concentrations greater than all other PFASs analyzed. *Hexagenia* spp. also produced the greatest $BSAF^{lipid}$ and $BSAF^{ww}$ (non-lipid-normalized biota-sediment accumulation factor) values for PFOS relative to the other two species. However, this was not the case for all PFASs. The trend of BSAF values and number of carbon atoms in the perfluoroalkyl chain of perfluorinated carboxylates varied among the three species but was similar for

* Corresponding author.
E-mail addresses: prosserr@uoguelph.ca, rsprosser@gmail.com (R.S. Prosser).

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perfluorinated sulfonates. Differences in the dominant pathways of exposure (e.g., water, sediment ingestion) likely explain a large proportion of the variation in accumulation observed across the three species.

1. Introduction

Polychlorinated biphenyls (PCBs) and perfluoroalkyl acids (PFASs) have been used in the production of a variety of commercial products. The largest use of PCBs was as a coolant or insulating fluid in transformers, capacitors, fluorescent light ballasts, and a variety of other electrical products. PFASs have been mainly used in the production of stain- and grease-proof coatings on food packaging and fabrics, polytetrafluoroethylene (Teflon®), and fire-fighting foams. However, PCBs and particular PFASs (e.g., PFOS) are considered persistent organic pollutants under the Stockholm Convention and participating countries are required to eliminate or reduce the release of these chemicals into the environment (Stockholm Convention, 2009). PCBs and PFOS have been found to be relatively persistent to degradation in the environment, as well as possess the capacity to accumulate in the tissue of exposed organisms, which can lead to trophic magnification (Ankley et al., 1992; Beyer and Biziuk, 2009; Borga et al., 2005; Geisy and Kannan, 2001, 2002; Giesy et al., 2010; Kannan et al., 2001a, 2001b, 2005; Muir et al., 2003).

The physicochemical properties that cause PCBs and PFASs to be susceptible to bioaccumulation also cause these classes of compounds to sorb to sediment when introduced to an aquatic system (Higgins and Luthy, 2006; Li et al., 2003). A relatively large number of studies have documented the contamination of sediment with these persistent pollutants (Helm et al., 2011; Muir et al., 1996; Myers et al., 2012; Painter et al., 2001; Yeung et al., 2013). When assessing the risk of these contaminants in sediment to an aquatic ecosystem it is important to consider the bioavailability of the contaminants. For this reason, standardized 28-day test protocols have been developed to assess the potential for a contaminant to accumulate in benthic oligochaetes from sediment (OECD, 2008; USEPA, 1994). A number of studies have examined the use of oligochaetes (e.g., *Lumbriculus variegatus*) and other laboratory-cultured benthic invertebrates and/or fish (e.g., *Pimephales promelas*) to assess the accumulation of PCBs in tissue from field-collected sediments (Ankley et al., 1992; Ciborowski et al., 1991; Drouillard et al., 1996; Pickard et al., 2001; Van Geest et al., 2011a, 2011b) and a small number of these studies have drawn comparisons between different species ability to accumulate PCBs (Ankley et al., 1992; Van Geest et al., 2011a, 2011b). However, an even smaller number of studies have investigated the uptake of PFASs into laboratory-cultured organisms from field-collected sediments and these studies have focused on two species, *L. variegatus* (Higgins et al., 2007; Lasier et al., 2011) or *Chironomus* spp. (Bertin et al., 2014; Xia et al., 2012). Variation in the accumulation of PFASs from contaminated field sediment across laboratory-cultured species has not been investigated.

The current study investigated variations in the accumulation of PCBs and various PFASs using sediment from a creek in Southern Ontario, Canada, which has historically had a number of suspected sources of these persistent organic pollutants located along its banks. These suspected sources have since stopped releasing PCBs and specific PFASs (specifically PFOS). However, there is a concern that historical inputs have contaminated the sediment along the creek with PCBs and various PFASs and these compounds are available for uptake by organisms inhabiting the creek, potentially increasing the risk of trophic magnification of these compounds. The Ontario Ministry of the Environment and Climate Change (MOECC) has developed and validated a standard test method to assess the bioavailability of traditionally assessed contaminants (e.g., PCBs, polycyclic aromatic hydrocarbons, dioxin-like PCBs, DDT and metabolites) in field-collected sediments to benthic invertebrates and fish (MOECC, 2011). However, this method has not been used by the MOECC to examine the bioavailability of PFASs to aquatic organisms from field-collected sediment. Therefore, the objectives of the current study were to 1) investigate the contamination of sediment with PCBs and PFASs along the creek, which may have experienced historical inputs from the use of fire-fighting foams at an airport and/or manufacturing that has used both classes of chemicals, and 2) investigate the bioavailability of PCBs and PFASs in field sediment to three species of organism (*Hexagenia* spp., *L. variegatus*, and *P. promelas*) and examine variability in the species' ability to accumulate these classes of chemicals. To the best of our knowledge, no studies have simultaneously compared the ability of these test species to accumulate PCBs and PFASs from contaminated field sediments. More specifically, no studies have examined the accumulation of PFASs in *Hexagenia* spp. and *P. promelas* (fathead minnows) from contaminated field-collected sediment. This is of particular importance as the use of *Hexagenia* spp. in sediment tests for toxicity and bioaccumulation is growing (Harwood et al., 2014; Watson-Leung et al., 2015). The data produced from the current study will strengthen the rationale for the use of these species in standardized tests of bioaccumulation from sediment. The results of this study will also provide insight on how data from these types of tests with these species can be used to assess the risk of sediment-borne contaminants to an aquatic system, and potentially the risk to human health when we consider the potential concentration of contaminants in fish.

2. Methods

2.1. Sediment collection

Sediment was collected according to the MOECC sediment assessment procedure outlined in Jaagumagi and Persaud (1993) and Environment Canada (1994) from five sites (sites A to E) along a creek in southwestern Ontario (Fig. 1). Sediment was removed with a shovel to a depth of ~10 cm and placed in a 10-L polyethylene bucket lined with a food-grade polyethylene bag for transport to and storage in the laboratory at 4 ± 2 °C. Sub-samples of collected sediment were taken and stored at −20 ± 2 °C before analysis of PFASs and PCBs. Upon arrival at the laboratory, all buckets of sediment collected from a site were combined and homogenized using a stainless steel mixer. The homogenized sediment was re-distributed to the 10-L polyethylene buckets lined with polyethylene bags for storage until bioaccumulation tests could be initiated. The physical and chemical properties of sediment

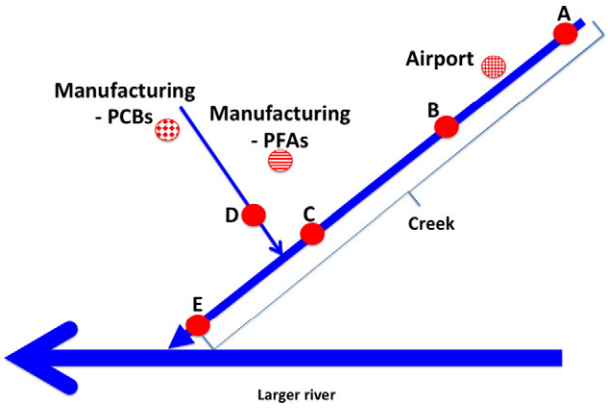

Fig. 1. Diagram identifying the location of sites at which sediment was sampled along a creek in Southern Ontario and sites of historical industrial use of PCBs and/or PFASs.