Environment International 67 (2014) 1–11

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

High levels of perfluoroalkyl acids in sport fish species downstream of a firefighting training facility at Hamilton International Airport, Ontario, Canada

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ARTICLE INFO

Article history:
Received 11 October 2013
Accepted 11 February 2014
Available online 13 March 2014

Keywords:
Perfluorooctane sulfonic acid (PFOS)
Aqueous film-forming foam (AFFF)
Sport fish
Human fish consumers
Fish consumption advisories

ABSTRACT

A recent study reported elevated concentrations of perfluorooctane sulfonic acid (PFOS) and other perfluoroalkyl acids (PFAAs) in surface water, snapping turtles, and amphipods in Lake Niapenco, downstream of Hamilton International Airport, Ontario, Canada. Here, our goals were to 1) determine the extent of PFAA contamination in sport fish species collected downstream of the airport, 2) explore if the airport could be a potential source, and 3) compare fish PFOS concentrations to consumption advisory benchmarks. The PFOS levels in several sport fish collected from the three locations closest to the airport (<40 km) were among the highest previously published in the peer-reviewed literature and also tended to exceed consumption benchmarks. The only other fish that had comparable concentrations were collected in a region affected by inputs from a major fluorinated chemical production facility. In contrast, PFOS concentrations in the two most downstream locations (>70 km) were comparable to or below the average concentrations in fish as observed in the literature and were generally below the benchmarks. With regards to perfluorocarboxylates (PFCAs), there was no significant decrease in concentrations in fish with distance from the airport and levels were comparable to or below the average concentrations observed in the literature, suggesting that the airport is not a significant source of PFCAs in these fish species. PFOS-based aqueous film-forming foam (AFFF) was used at a firefighting training facility at the airport in the 1980s to mid-1990s. Taken together, our results provide evidence that the historical use of AFFF at the airport has resulted in fish PFOS concentrations that exceed the 95th percentile concentration of values reported in the literature to date.

1. Introduction

Since the 1950s, perfluoroalkyl acids (PFAAs) and their precursors have found numerous applications as polymers and surfactants due to their chemical and thermal stability and because they contain both hydrophobic and lipophobic properties (Buck et al., 2011; Paul et al., 2009; Prevedouros et al., 2006). One such use is as an additive in aqueous film-forming foam (AFFF) used to extinguish large-scale fuel fires such as those produced by aircraft (Moody and Field, 2000). AFFF was developed by the 3M company and the US Naval Research Laboratory in the mid-1960s (Scheffey and Hanauska, 2002). Perfluorooctane sulfonic acid (PFOS) was an active ingredient utilized in the 3M AFFF formulation until the phase-out of PFOS and related chemistry in 2000–2002 (Moody and Field, 2000; Paul et al., 2009; Place and Field, 2012). Current AFFF formulations are based on telomer fluorosurfactants that do not contain PFOS (Fire Fighting Foam Coalition; Place and Field, 2012; Scheffey and Hanauska, 2002; Seow, 2013). However, as a result of their persistence (Cheng et al., 2008), PFAAs are expected to remain in the environment long after their release (Awad et al., 2011; Butt et al., 2010; Houde et al., 2011). Although PFOS-based AFFF has not been imported into Canada since 2002, approximately 300 tons of AFFF (representing approximately 3 tons of PFOS) are estimated to remain present in stockpiles (Environment Canada, 2006).

Several studies have implicated AFFF as a source of PFAAs to the environment. Elevated PFOS concentrations in AFFF-affected areas have been observed in fish (Awad et al., 2011; Karrman et al., 2011; Moody et al., 2002; Schlummer, 2008), surface water (Awad et al., 2011; Moody et al., 2002; Schlummer, 2008), sediment (Awad et al., 2011; Karrman et al., 2011), soil (Houtz et al., 2013; Karrman et al., 2011; Weber et al., 2010), groundwater (Backe et al., 2013; Moody et al., 2003; Weber et al., 2010) including samples from wells used for

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http://dx.doi.org/10.1016/j.envint.2014.02.005
0160-4120/

drinking water (Weis et al., 2012), fruits and vegetables collected from home gardens (Weis et al., 2012), and human blood plasma (Weis et al., 2012). At sites where replacement AFFF formulations have been used following the 2000–2002 3M phase-out of PFOS and related chemistries, fluorinated chemicals other than PFOS were found in environmental media (Moe et al., 2012; Oakes et al., 2010). The non-occupational exposure of PFAAs to humans is generally thought to be dominated by dietary intake (D'Hollander et al., 2010; EFSA, 2008; Fromme et al., 2009; Trudel et al., 2008; Vestergren and Cousins, 2009). Recent studies have implicated fish consumption from contaminated water bodies as a major source of intake to anglers (Haug et al., 2010; Holzer et al., 2011; Lindstrom et al., 2011). As such, it is important to determine how PFAA concentrations in sport fish are impacted by AFFF releases.

In 2010, as part of a routine monitoring study, de Solla et al. (2012) discovered highly elevated concentrations of PFOS and other PFAAs in surface water, snapping turtle (*Chelydra serpentine*) plasma, and amphipods (*Gammarus* or *Hyalella*) in Lake Niapenco (also known as Binbrook Reservoir), in southern Ontario, Canada. An in-depth follow-up study found PFOS concentrations of 122 ± 23.5 ng/L (arithmetic mean ± standard deviation), 2377 ± 1460 ng/g wet weight, and 1125 ± 637 ng/g wet weight in water, snapping turtle plasma, and amphipods, respectively. These concentrations were elevated compared to reference locations, and for the snapping turtle, higher than any PFOS concentrations previously reported in the literature (de Solla et al., 2012). The spatial trends observed by de Solla et al. (2012) implicated AFFF contamination at the John C. Munro Hamilton International Airport, which drains into the Welland River upstream of Lake Niapenco, as the likely source of the PFAAs. Firefighting training was conducted at the airport between 1985 and 1994, with 15,000 L of PFOS-based foam sprayed each year during this time period (exp Services Inc., 2011; Minor, 2012; O'Reilly, 2011; Van Dongen, 2012). Although a landfill, which closed in 1980, is located downstream of the airport, there are no waste water treatment plant discharges to this lake and there is virtually no industry nearby.

The objective of this study was to examine the extent of PFAA contamination in Lake Niapenco and Welland River sport fish species and to explore a link between PFOS accumulation in fish and possible upstream sources such as the Hamilton International Airport. We also compare fish concentrations of PFOS to the fish consumption advisory benchmarks used by the Ontario Ministry of the Environment (OMOE), Ontario, Canada (OMOE, 2013). Overall, this study is important in realizing the extent of impact that a point source of AFFF has on PFOS concentrations observed in sport fish species.

2. Materials and methods

2.1. Fish collection

Sport fish species were collected by OMOE Sport Fish Contaminant Monitoring staff between 2009 and 2012 from within six sampling blocks (including Lake Niapenco) along the Welland River (Fig. 1). The fish species collected included Black Crappie (*Pomoxis nigromaculatus*), Bluegill (*Lepomis macrochirus*), Brown Bullhead (*Ameiurus nebulosus*), Channel Catfish (*Ictalurus punctatus*), Common Carp (*Cyprinus carpio*), Freshwater Drum (*Aplodinotus grunniens*), Largemouth Bass (*Micropterus salmoides*), Northern Pike (*Esox lucius*), Pumpkinseed (*Lepomis gibbosus*), Rock Bass (*Ambloplites rupestris*), Smallmouth Bass (*Micropterus dolomieu*), Walleye (*Sander vitreus*), White Crappie (*Pomoxis annularis*), White Sucker (*Catostomus commersonii*), and Yellow Perch (*Perca flavescens*) (Tables S-1 and S-2). The number of samples collected for each fish species/block/year are provided in Tables S-1 and S-2. The fish were collected using an electrofishing boat, trap nets, gill nets, and seine nets. Upon collection, the length, weight, and sex of each fish were recorded, and a skinless, boneless filet of the dorsal muscle was removed and stored frozen at −20 °C until analysis. The dorsal muscle is most commonly consumed by humans and is the portion recommended for human consumption by the OMOE Guide to Eating Ontario Sport Fish (OMOE, 2013) to minimize contaminant exposures. The preparation and analysis of

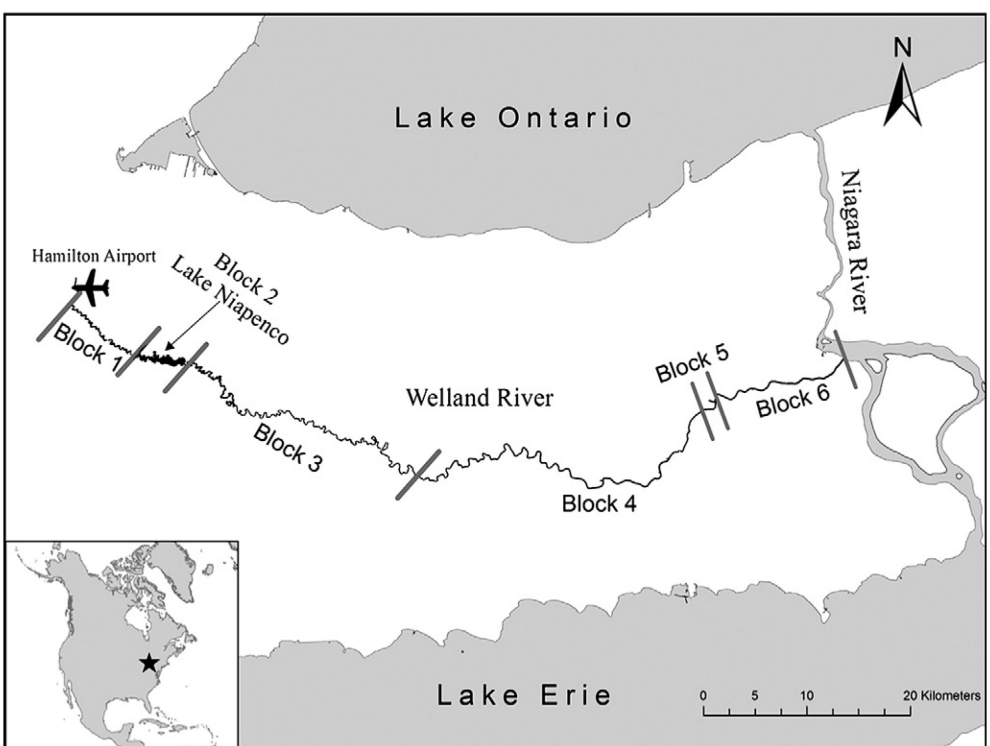

Fig. 1. Map illustrating the locations of the six sampling blocks in the Welland River, Ontario.