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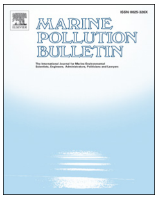

Bioaccumulation of perfluoroalkyl substances in exploited fish and crustaceans: Spatial trends across two estuarine systems

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ABSTRACT

Spatial patterns in perfluoroalkyl substances were quantified for exploited fish and crustaceans across two contrasting Australian estuaries (Port Stephens and Hunter River). Perfluorooctane sulfonate (PFOS) was detected in 77% of composites from Port Stephens and 100% of composites from Hunter River. Most species from Port Stephens showed a clear trend with distance to source. In contrast, only a subset of species showed this trend in the Hunter River, potentially due to species movement patterns and differing hydrology. Spatial modelling showed that PFOS concentrations were expected to exceed the relevant trigger value up to ~13,500 m from the main point source for Port Stephens and ~9000 m for the Hunter River. These results represent the first major investigation of bioaccumulation of PFASs in exploited species in Australian estuaries, and highlight various factors that can contribute to spatial patterns in bioaccumulation.

1. Introduction

Per- and poly-fluorinated alkyl substances (PFASs) are a group of synthetic chemicals produced since the mid-twentieth century. PFASs have been used extensively in a wide range of industrial and domestic products including textiles, food packaging, mist suppressants, pesticides, polishes, electronic components and firefighting foams (Ahrens, 2011; Norden, 2013). Aqueous film forming foams (AFFF), used for controlling standing hydrocarbon fires, have typically incorporated PFASs and PFAS precursors. Although AFFF production has only been a relatively small proportion of the total production of PFASs (Ahrens, 2011), AFFF use has resulted in significant point-sources of contamination, particularly where firefighting training activities have occurred (Bräunig et al., 2017; Gewurtz et al., 2014). Because of the relatively high mobility of some PFASs in water, and the fact they are highly persistent, these point sources often result in contamination of nearby waterways via groundwater or surface drainage (Gaylard, 2016; Gewurtz et al., 2014). PFAS emissions into waterways can also arise from diffuse sources such as wastewater treatment plants, landfill, and stormwater (Ahrens, 2011; Gaylard, 2016).

Bioaccumulation of PFASs including perfluorooctane sulfonate (PFOS) and some long-chain perfluorocarboxylic acids (PFCAs) by aquatic organisms has been demonstrated in both laboratory (Inoue et al., 2012; Martin et al., 2003a, 2003b) and field studies (Gewurtz et al., 2014; Hong et al., 2015; Kelly et al., 2009; Lescord et al., 2015). PFASs are classed as emerging contaminants as the toxicological and ecotoxicological implications of exposure are not yet fully understood. To date, evidence of human health and ecological effects related to PFAS exposure has been inconsistent. However, as a precautionary approach, substantial attention has been given to determining the level and spatial extent of PFAS contamination in waterways because of the potential risks. Considering that these chemicals will inevitably make their way into aquatic ecosystems, identifying the factors that affect PFAS concentrations in aquatic biota is necessary for quantifying and managing these risks.

Studies defining general patterns of PFAS contamination in aquatic ecosystems are rapidly increasing, and recent field studies have sought to identify general patterns of contamination, and factors that potentially contribute to these patterns. PFOS and long-chain PFCAs were generally shown to dominate PFAS concentrations in biota from estuaries and coastal areas of South Korea (Hong et al., 2015; Naile et al., 2010) and China (Yang et al., 2012). The South Korean data (Hong et al., 2015; Naile et al., 2010) suggest a possible decrease in PFOS concentrations over a five-year period, whereas short-chain PFASs

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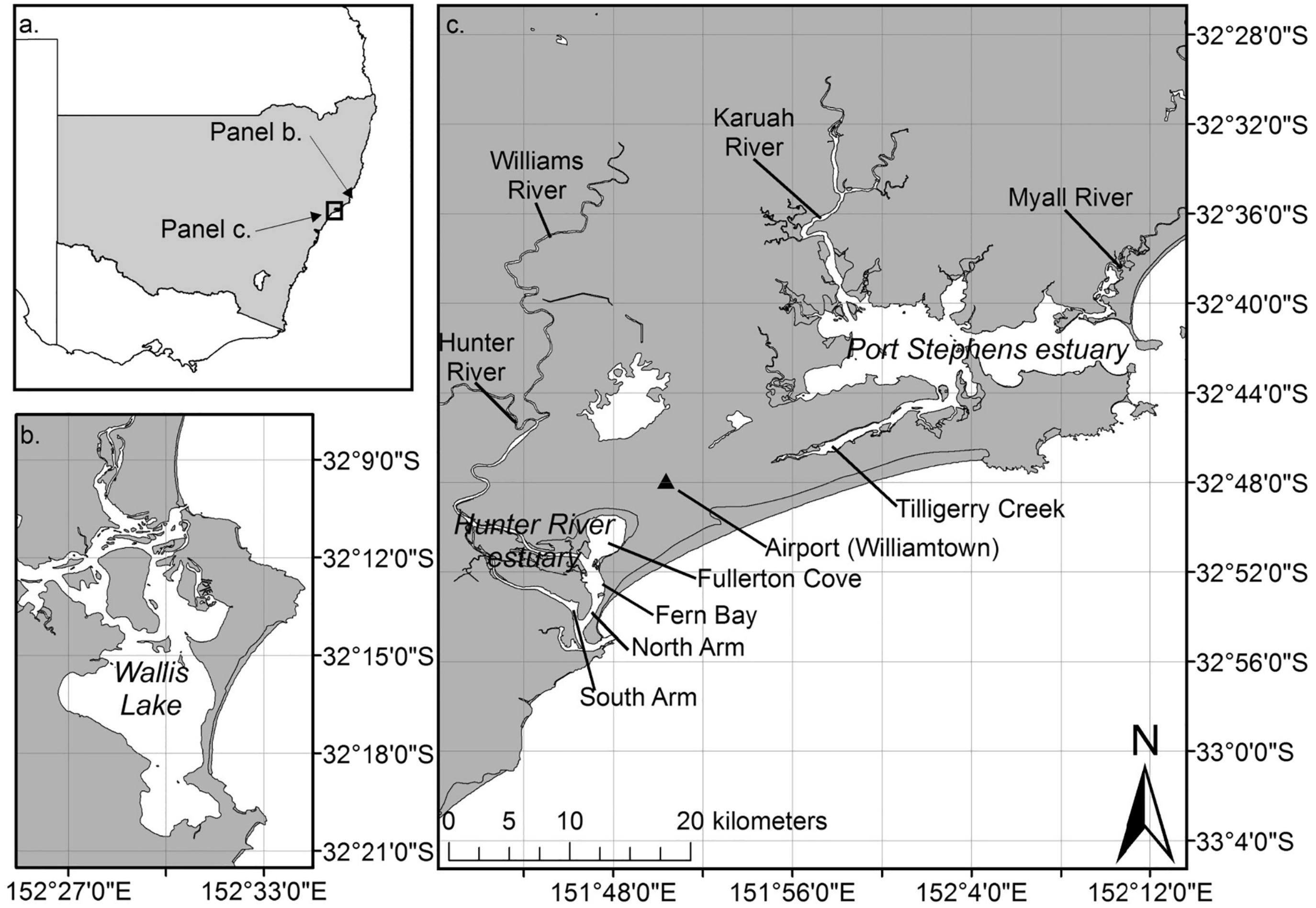

Fig. 1. Map showing locations and key features of estuaries sampled during study. Location of study estuaries in south-eastern Australia (a.; New South Wales shaded in gray); Wallis Lake (b.); Port Stephens and the Hunter River estuaries (c.; location of Williamtown RAAF Base and airport indicated as triangle). Gray shading (on b. and c.) represents land. More detail on Port Stephens and Hunter River estuary can be found in Figs. 2 and 3 respectively.

increased in concentration and PFOA remained relatively consistent. PFAS concentrations have been reported to be higher in biota from lakes close to a point source (e.g. an airport) than more remote lakes (Lescord et al., 2015) and in fish with benthic-based diet compared to pelagic-based (Lescord et al., 2015; Martin et al., 2004). While these studies provide some insight, differences in biology, proximity to sources and PFAS concentrations have not been well studied in the context of species-specific differences in bioaccumulation.

The Williamtown area in New South Wales, Australia, offers a useful site to test hypotheses about spatial patterns of PFAS concentrations in aquatic biota species. The Williamtown Royal Australian Airforce (RAAF) Base has conducted firefighting training with AFFF for a number of decades, resulting in contamination of groundwater and surface water coming from the RAAF base (AECOM, 2016). This has resulted in PFAS contamination of Tilligerry Creek (part of Port Stephens estuary, Fig. 1), located to the north-east of the RAAF base. The RAAF base is likely to be the main source of PFAS to Tilligerry Creek (and Port Stephens more broadly) from groundwater and surface water flow, and other major point sources of PFASs are thought to be lacking throughout the catchment. To the south of the RAAF base, contamination has spread to Fullerton Cove, which adjoins the lower Hunter River. While the RAAF base is likely to be the main point source to the estuary, there is considerable industrial and urban influence around the Hunter River, so it is possible that contamination may originate from other diffuse sources elsewhere in the catchment (including possible non-AFFF sources). In previous studies, PFOS was the primary PFAS observed in aquatic biota samples from both Tilligerry Creek and Fullerton Cove (Taylor and Johnson, 2016), which is consistent with findings elsewhere where AFFF was the dominant source (Ahrens and Bundschuh, 2014).

This study reports an extensive investigation of PFAS contamination in estuarine biota. Specifically, this study 1) quantifies species-specific contamination levels in the major species exploited by fisheries in the estuaries of south-eastern Australia; and 2) models spatial trends in contamination relative to the major PFAS point-source in two south-eastern Australian estuaries.

2. Methods

2.1. Study area

Port Stephens and Hunter River are two large estuaries in south-eastern Australia (Fig. 1). Port Stephens is classified as an immature, tide-dominated, drowned valley estuary, whereas the Hunter River is a mature, wave-dominated barrier estuary (Roy et al., 2001). Both estuaries support productive fisheries dominated by Sea Mullet (*Mugil cephalus*), Yellowfin Bream (*Acanthopagrus australis*), Sand Whiting (*Sillago ciliata*), Dusky Flathead (*Platycephalus fuscus*), Luderick (*Girella tricuspidata*), Common Silverbiddy (*Gerres subfasciatus*), Mud Crab (*Scylla serrata*), Blue Swimmer Crab (*Portunus armatus*, mainly Port Stephens) and School Prawn (*Metapenaeus macleayi*, mainly Hunter River), and also represent important nursery grounds for Eastern King Prawn (*Penaeus plebejus*) (Taylor et al., 2016). Collectively, these species normally comprise > 85% of the annual commercial harvest (by