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Polyfluoroalkyl substance exposure in the Mid-Ohio River Valley, 1991–2012☆

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ABSTRACT

Background: Industrial discharges of perfluorooctanoic acid (PFOA) to the Ohio River, contaminating water systems near Parkersburg, WV, were previously associated with nearby residents' serum PFOA concentrations above US general population medians. Ohio River PFOA concentrations downstream are elevated, suggesting Mid-Ohio River Valley residents are exposed through drinking water.
Objectives: Quantify PFOA and 10 other per- and polyfluoroalkyl substances (PFAS) in Mid-Ohio River Valley resident sera collected between 1991 and 2013 and determine whether the Ohio River and Ohio River Aquifer are exposure sources.
Methods: We measured eleven PFAS in 1608 sera from 931 participants. Serum PFOA concentration and water source associations were assessed using linear mixed-effects models. We estimated between-sample serum PFOA using one-compartment pharmacokinetics for participants with multiple samples.
Results: In serum samples collected as early as 1991, PFOA (median = 7.6 ng/mL) was detected in 99.9% of sera; 47% had concentrations greater than US population 95th percentiles. Five other PFAS were detected in greater than 82% of samples; median other PFAS concentrations were similar to the US general population. Serum PFOA was significantly associated with water source, sampling year, age at sampling, tap water consumption, pregnancy, gravidity and breastfeeding. Serum PFOA was 40–60% lower with granular activated carbon (GAC) use. Repeated measurements and pharmacokinetics suggest serum PFOA peaked 2000–2006 for participants using water without GAC treatment; where GAC was used, serum PFOA concentrations decreased from 1991 to 2012.
Conclusions: Mid-Ohio River Valley residents appear to have PFOA, but not other PFAS, serum concentrations above US population levels. Drinking water from the Ohio River and Ohio River Aquifer, primarily contaminated by industrial discharges 209–666 km upstream, is likely the primary exposure source. GAC treatment of drinking water mitigates, but does not eliminate, PFOA exposure.

1. Introduction

Perfluoroalkyl and polyfluoroalkyl substances (PFAS) are industrial chemicals used for stain-, stick- and water-resistant coatings, food contact papers, firefighting foams, metal plating, semiconductors and photographics/photolithographics (Buck et al., 2011; Lau et al., 2007). Perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS) are the most widely studied PFAS. 2610-21400 tons of carboxyl PFAS were emitted to the environment 1951–2015; up to 6420 more will be emitted by 2030 (Wang et al., 2014). US PFAS manufacturers reduced PFOA emissions 91–100% by 2014 (United States Environmental Protection Agency, 2015a), but production continues internationally.

Since 1999–2000, US population serum PFAS concentrations, measured by the National Health and Nutrition Examination Survey (NHANES), have steadily decreased; median 2011–2012 concentrations were 2.08 ng/mL (PFOA) and 6.53 ng/mL (PFOS) (CDC, 2015). Food, drinking water and house dust are major exposure sources (D'Hollander et al., 2010). Children are also exposed during

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pregnancy and through breastfeeding (Liu et al., 2011). Industrial PFAS discharges can contaminate municipal source waters (Holzer et al., 2008), and contaminated water may be the most important environmental PFAS exposure source (Ericson et al., 2008).

Mid-Ohio River Valley (Huntington, WV to Louisville, KY, Fig. 1) residents may be exposed to PFAS, especially PFOA, through contaminated drinking water from the Ohio River or Ohio River Aquifer. Serum PFOA concentrations above NHANES medians were reported in the C-8 Health Project cohort in Parkersburg, WV (Emmett et al., 2006), and two Cincinnati cohorts, the Breast Cancer and the Environment Research Project (BCERP) Puberty Study (Pinney et al., 2014), and the Health Outcomes and Measures of the Environment (HOME) study (Kato et al., 2014; Braun et al., 2016). Serum PFOA concentrations were significantly associated with water utility (Pinney et al., 2014) or Ohio River Aquifer use (Emmett et al., 2006). The Ohio River Aquifer in the C-8 study area was contaminated by industrial PFOA discharges to the Ohio River (Paustenbach et al., 2007). September 2009 downstream Ohio River PFOA concentrations were 9.2–19.1 ng/L; historical PFOA concentrations were higher (Paustenbach et al., 2007). PFAS exposure may be reduced by granular activated carbon filtration (GAC). Some Ohio River Valley utilities use GAC, and GAC has reduced PFOA concentrations in other municipal water systems (Rahman et al., 2014). GAC use was associated with lower resident serum PFOA concentrations, although some were still above US general population medians (Pinney et al., 2014; Bartell et al., 2010).

To assess exposure to PFAS in Mid-Ohio River Valley residents, we measured the concentrations of 11 different PFAS in sera collected between 1991 and 2013. PFOA concentrations between serum measurements were estimated for participants with multiple serum samples using pharmacokinetic models. We evaluated PFOA and water source associations using mixed-effects statistical models to determine whether the Ohio River and Ohio River Aquifer are significant PFOA exposure sources for residents of the Mid-Ohio River Valley.

2. Materials and methods

2.1. Study population

This analysis included participants from the Fernald Community Cohort (FCC), Ohio River Valley (ORV) Study and BCERP Puberty Study Cincinnati site cohort. FCC adult participants were recruited from residents living less than 8 km from a uranium plant 32 km northwest of Cincinnati, primarily between 1990 and 1994; follow-up continued through 2008. Eligibility, design and participant characteristics were described previously (Wones et al., 2009; FCC website). Cohort members were eligible for inclusion in this study if they lived in zip codes bordering the Ohio River from Gallipolis, OH to southeast Indiana sometime between 1980 and 2008 (Fig. 1). An additional 20 members of the remaining FCC cohort were included because they had nuclear family members living between Parkersburg, WV and Cincinnati, OH during those years. A total of 450 FCC cohort members were contacted in 2011–2014 for additional residential history and beverage consumption data collection.

We recruited ORV participants between 2009 and 2012 from medical practices in Huntington, Portsmouth, OH, and Cincinnati suburbs; some family members of BCERP participants were

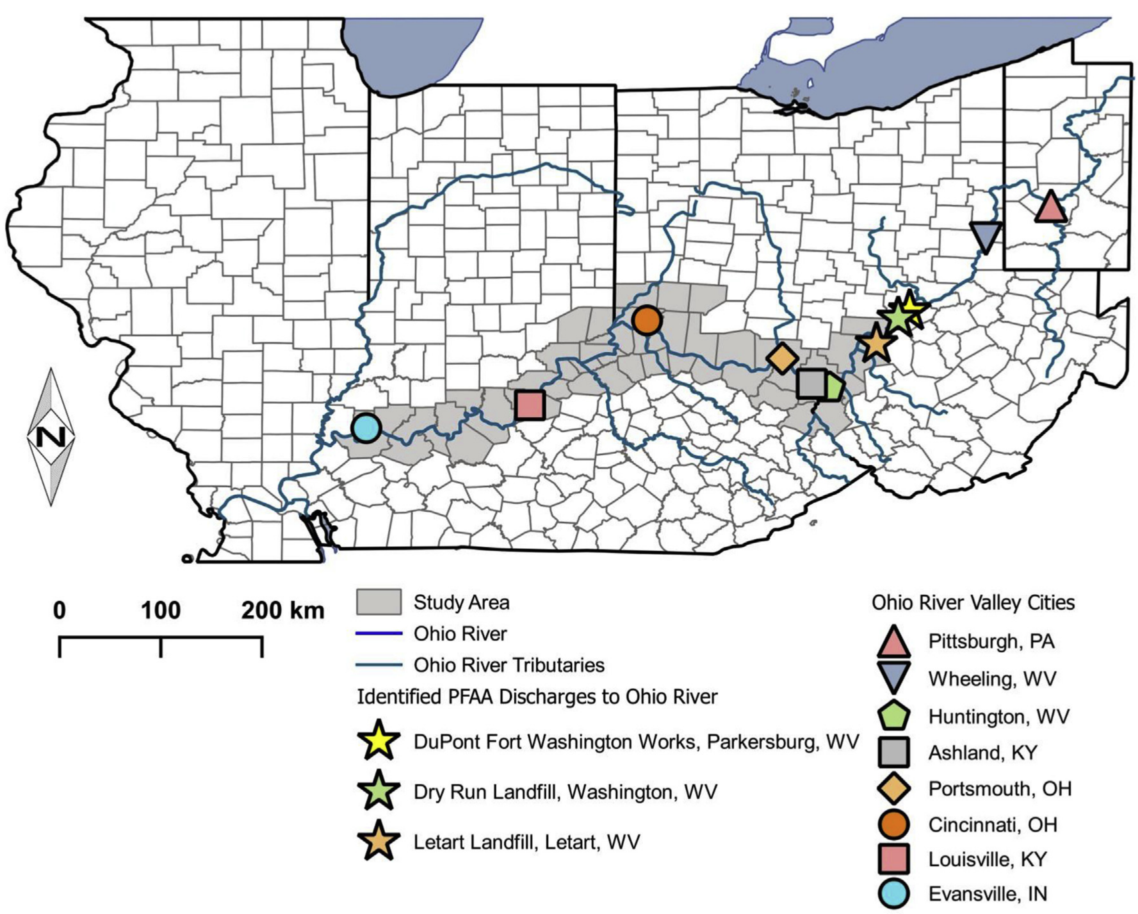

Fig. 1. The Ohio River Valley.