



Hunter & Hunter^{LLC}

640 Ardleigh Drive
Akron, OH 44303
T: 234.281.2528
F: 330.805.4518
hunterfirm.org

September 7, 2018

Sent via U.S. Certified Mail and electronically submitted via governor.ohio.gov

Governor John Kasich
Riffe Center, 30th Floor
77 South High St.
Columbus, OH 43215-6117

Re: Proposed Revisions to ORSANCO Pollution Control Standards

Dear Governor Kasich:

On behalf of FreshWater Accountability Project, I am writing you to request that you direct Commissioners Craig Butler, John M. Hoopingarner, and Stuart F. Bruny to vote against ORSANCO's current extreme proposal to essentially eliminate ORSANCO's Pollution Control Standards ("PCS").

FreshWater Accountability Project ("FreshWater") is a grassroots organization with members throughout Ohio that is focused on protecting the very resource that gives us life—our fresh water. FreshWater is extremely alarmed that at a time when the Ohio River continues to face ongoing and emerging threats, ORSANCO is proposing to abandon its critical role in setting, maintaining, reviewing, and revising PCS for the Ohio River. **ORSANCO's proposal is not supported by factual analysis and is nearly certain to result in increased levels of pollution and accumulation of toxins in the Ohio River—the drinking water source for approximately 5 million people.** The proposal would also result in an abrogation of ORSANCO's duties under its originating Compact.

A majority of ORSANCO Commissioners attempt to justify this drastic reduction by falsely stating the PCS are redundant to current protections under the Clean Water Act. Their assertion is not supported by facts, including the analysis undertaken by ORSANCO's own staff, which found there are 193 parameters for which ORSANCO has criteria that a state or federal agency does not, and that there are 251 parameters for which federal or state agency criteria is significantly less stringent than that criteria set by ORSANCO. **In other words, for 38% of the**

standards evaluated, ORSANCO has a *significantly more stringent* standard than applicable agency standards, and for 30% of the standards evaluated, ORSANCO *has* a standard where the agencies have none. ORSANCO did not even undertake the critical analysis of how the agency works in a cohesive manner to comprehensively address pollution in the Ohio River.

Just one example of the crucial role ORSANCO plays in protecting the Ohio River is ORSANCO's ban on mixing zones of bioaccumulative chemicals of concern (BCCs). There is no question that this ban decreases the amount of BCCs that can be permitted to enter the Ohio River. No equivalent ban exists through federal regulation and ORSANCO's own review of state regulations demonstrates that multiple signatory states have no such ban absent the ORSANCO PCS. **The proposed changes would, in turn, result in an increase of bioaccumulative chemicals entering the Ohio River.**

FreshWater further outlines its concerns with ORSANCO's proposal in its comments submitted to ORSANCO during the public review process. These are attached to this letter for your review.

ORSANCO held only one public hearing regarding the proposed PCS revision. **Comments provided at this well-attended hearing were *entirely in support of maintaining or expanding the PCS.*** In addition, 97% of comments received during the proposal's first round of review, and *all comments from municipal water suppliers, the general public, and environmental advocacy groups, supported maintaining or expanding the PCS.*

You have publicly stated that it is important to be "pro-environment." **FreshWater urges you to stand up for the Ohio River and the people who rely on it for their health and sustenance.** Accordingly, **FreshWater asks you to direct Commissioners Craig Butler, John M. Hoopingarner, and Stuart F. Bruny to vote against ORSANCO's current proposal to significantly reduce the PCS.** FreshWater further asks that you direct Commissioners Butler, Hoopingarner, and Bruny to support maintaining or expanding the PCS to ensure ORSANCO fulfills its duties under its originating Compact and works to reduce and abate pollution in the Ohio River.

Sincerely,



Megan M. Hunter, Esq.
Hunter & Hunter LLC
640 Ardleigh Dr.
Akron, OH 44303
Phone (234)281-2528
Email: megan@hunterfirm.org



Hunter & Hunter_{LLC}

640 Ardleigh Drive
Akron, OH 44303
T: 234.281.2528
F: 330.805.4518
hunterfirm.org

August 20, 2018

Via electronic mail (PCS@orsanco.org)

ORSANCO

Attn: PCS Comments

5735 Kellogg Avenue

Cincinnati, OH 45230

Re: 2018 Review of ORSANCO Pollution Control Standards

Dear Commissioners:

FreshWater Accountability Project (“FreshWater”) is alarmed that the Commission has moved forward with its extreme proposal to essentially eliminate ORSANCO’s critical role in setting, maintaining, reviewing, and revising Pollution Control Standards (“PCS”) for the Ohio River. **This decision is in direct opposition to ORSANCO’s duties under the Ohio River Valley Water Sanitation Compact (the “Compact”), is not supported by factual analysis, and is nearly certain to result in increased levels of pollution and accumulation of toxins in the Ohio River.**

FreshWater submitted comments during the first round of public review of proposed changes to the PCS on February 24, 2018. These are attached as Exhibit A and incorporated herein. FreshWater restates and affirms the claims made therein for purposes of this round of review.

I. ORSANCO HAS NO FACTUAL BASIS TO SUPPORT ITS PROPOSED REVISION TO THE PCS—WHICH RESULTS IN AN ABROGATION OF THE COMMISSION’S DUTIES UNDER THE COMPACT

To support its proposal to essentially eliminate the PCS, ORSANCO has argued the demonstrably false notion that the PCS are redundant to standards adopted under the Clean Water Act (“CWA”), through Federal regulations and Federally-approved State Implementation Plans (“SIPs”). **ORSANCO’s own analysis clearly refutes this premise.** Analysis by ORSANCO staff of 661 standards, attached as Exhibit B and incorporated herein, demonstrates that there are 193 parameters for which ORSANCO has criteria that a state or federal agency

does not, and that there are 251 parameters for which federal or state agency criteria is significantly less stringent than that criteria set by ORSANCO. **This analysis shows that for 38% of the standards evaluated, ORSANCO has a significantly more stringent standard than applicable agency standards, and for 30% of the standards evaluated, ORSANCO has a standard where the agencies have none.**

This analysis alone demonstrates that ORSANCO PCS are not redundant to CWA standards as established by Federal and state agencies. Despite signatory states' responsibility to take a cooperative approach through ORSANCO to control and abate pollution in the Ohio River,¹ the majority of Commissioners did not even find it necessary to evaluate the cooperative value ORSANCO was created to uniquely provide. Instead, ORSANCO provided cursory statements concluding that ORSANCO standards are met or exceeded under CWA standards, providing no evidence to support these conclusions, and in fact providing evidence that demonstrates the PCS are not duplicative. Without an analysis regarding ORSANCO's role in the comprehensive addressing of pollution problems impacting the Ohio River, ORSANCO has not completed the factual analysis required to justify the proposed rule change.

One example of the unique protections provided by ORSANCO's current PCS is ORSANCO's ban on mixing zones for bioaccumulative chemicals of concern ("BCC"), which includes any chemical that accumulates in aquatic organisms and has the potential to cause adverse effects as the result of that accumulation.² Chemicals explicitly named in the mixing zone ban include: Lindane, Hexachlorocyclohexane, alpha-Hexachlorocyclohexane, beta-Hexachlorocyclohexane, delta-Hexachlorocyclohexane, Hexachlorobutadiene, Photomirex, 1,2,4,5-Tetrachlorobenzene, Toxaphene, Pentachlorobenzene, 1,2,3,4-Tetrachlorobenzene, Mirex, Hexachlorobenzene, Chlordane, DDD, DDT, DDE, Octachlorostyrene, PCBs, 2,3,7,8-TCDD, Mercury, and Dieldrin.³

ORSANCO's BCC mixing zone ban is critical to preventing the accumulation of BCCs in the Ohio River because it decreases the amount of bioaccumulative toxins that can be permitted to enter the river. No equivalent ban exists through U.S. EPA agency regulation, and ORSANCO's review of state agency regulation demonstrates that multiple signatory states have no such ban absent the ORSANCO PCS.⁴

Thus, ORSANCO's primary justification for its proposed revision is not supported by factual analysis, and what factual analysis ORSANCO has conducted on the matter suggests the PCS provide hundreds of numeric standards and policies that are not replicated anywhere by CWA standards and policies implemented by state and Federal law.

¹ See Preamble, Article I, VI, VIII, OHIO RIVER VALLEY WATER SANITATION COMPACT (June 30, 1948).

² Ohio River Valley Water Sanitation Commission, Pollution Control Standards (2015), Chapter 4: Mixing Zone Designation, F.4.

³ Ohio River Valley Water Sanitation Commission, Pollution Control Standards (2015), Chapter 4: Mixing Zone Designation, F.6.

⁴ See Implementation of ORSANCO Pollution Control Standards, "Alternative 2" Expanded: "Clean Water Act Alternative to ORSANCO PCS Rule."

II. THE MAGNITUDE OF THE PROPOSED CHANGES AND THE OVERWHELMING PUBLIC OUTCRY AGAINST THE PROPOSAL WARRANT ADDITIONAL HEARINGS IF THE PROPOSAL IS GOING TO BE PURSUED

FreshWater wishes to remind the Commissioners that the Ohio River serves as the drinking water source for approximately 5 million people and that the River continues to face threats by known and emerging contaminants that threaten public health. *See e.g.* Robert L. Herrick, et al. *Polyfluoroalkyl substance exposure in the Mid-Ohio River Valley, 1991-2012*, 228 ENVIRONMENTAL POLLUTION 50 (2017) (concluding “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.⁵) (attached and incorporated herein as Exhibit C). ORSANCO’s proposed revisions to the PCS would result in the Commission abandoning its duty to control and abate pollution in this important drinking water source using the comprehensive cooperative approach that can only be achieved by this unique interstate agency.

ORSANCO has held *one* public hearing regarding the proposed revision to the PCS, which occurred July 26, 2018 at the Cincinnati Airport in Erlanger, Kentucky. *One* public hearing on a matter that will impact the drinking water source for 5 million people in six different states. **Comments provided at this well-attended hearing were uniformly in support of maintaining or expanding the PCS.**⁶ Likewise, 97% of comments received during the proposal’s first round of review, and *all* comments received from municipal water suppliers, the general public, and environmental advocacy groups, supported maintaining or expanding the PCS.⁷

ORSANCO’s proposal would remove hundreds of standards for which no equivalent standard exists in state or Federal law, and it would significantly reduce hundreds of standards. A change in drinking water source protection of this magnitude should include public hearings throughout the affected area. **If ORSANCO does not abandon this proposal, as it should, ORSANCO should hold public hearings along the Ohio River in order to provide impacted users the opportunity to voice their concerns. These hearings should, at minimum, cover both the upper and lower basins of the Ohio River.** Likewise, the public comment period should be re-noticed and extended to conclude after ORSANCO has held these public hearings.

⁵ (Emphasis added).

⁶ Transcript, Ohio River Valley Water Sanitation Commission Public Hearing, Holiday Inn, Cincinnati Airport, 1717 Airport Exchange Boulevard, Erlanger, Kentucky July 26, 2018, *available at* <http://www.orsanco.org/wp-content/uploads/2018/08/Public-Hearing-Transcript-July-26-2018.pdf>

⁷ During the first round of public comment, only comments submitted by or on behalf of polluting industries supported the current proposal (which made up only 3% of comments received during the first round of review). **There was no testimony presented at the public hearing in support of the current proposal.**

CONCLUSION

For the reasons stated herein, FreshWater urges ORSANCO to abandon its proposed changes to its Pollution Control Standards. ORSANCO should maintain the PCS—which are essential to maintaining and advancing the goals of the Compact and to protecting the Ohio River.

Respectfully submitted,

A handwritten signature in black ink, appearing to read 'Megan M. Hunter', written in a cursive style.

Megan M. Hunter, Esq.
Hunter & Hunter LLC
640 Ardleigh Dr.
Akron, OH 44303
Phone (234)281-2528
Email: megan@hunterfirm.org

Exhibit A



Hunter & Hunter^{LLC}

640 Ardleigh Drive
Akron, OH 44303
T: 234.281.2528
F: 330.805.4518
hunterfirm.org

February 24, 2018

Via electronic mail (PCS@orsanco.org)

ORSANCO

Attn: PCS Comments

5735 Kellogg Avenue

Cincinnati, OH 45230

Re: 2018 Review of ORANSCO Pollution Control Standards

Dear Commissioners:

On behalf of FreshWater Accountability Project, we are submitting comments on the radical changes Ohio River Valley Water Sanitation Commission (“ORSANCO”) is considering regarding its Pollution Control Standards (“PCS”). FreshWater Accountability Project (“FreshWater”) is a nonprofit organization with a mission to preserve and protect freshwater supplies through education and community action, and is dedicated to promoting health and wellbeing by protecting the environment. FreshWater Accountability Project has members located throughout the state of Ohio, including members along the Ohio River and throughout the Ohio River Basin.

Introduction

During this year’s review of its Pollution Control Standards, ORSANCO is debating the extreme measure of essentially eliminating the Pollution Control Standards. ORSANCO has presented the public with five proposed alternatives to its current approach to its Pollution Control Standards. Two of these (Alternatives #1 and #2) eliminate the Pollution Control Standards, either directly or in effect. In its request for public comment, ORSANCO states that Alternative #2 is preferred by the majority of the Commissioners. In addition to brief descriptions of each of the five alternatives, the

request for public comments included supplemental documents providing more detail on Alternative #2 and a mock-up of revised standards based on Alternative #2. ORANSCO provided no comparable detailed documentation or mock-up for the other alternatives.

For reasons explained more fully in the following sections of this comment, FreshWater is deeply concerned that ORSANCO is considering abrogating its responsibilities under the Ohio River Valley Water Sanitation Compact (the “Compact”) in this way.

FreshWater commends the Minority Report of Commissioners and wishes to echo and affirm the statements made therein. FreshWater also supports the comments submitted by the Water Users Advisory Committee, the Watershed Organizations Advisory Committee, the Public Information Advisory Committee, and the majority of comments offered by the Technical Committee. Lastly, FreshWater cautions against letting comments submitted by the Chemical Industry Advisory Committee and the Power Industry Advisory Committee dictate the future of standards critical for the protection and restoration of the Ohio River.

I. Ceasing to Maintain Pollution Control Standards is an Abrogation of ORSANCO’s Responsibilities Under the Compact

The expanded Analysis of Alternative #2 provided in the request for public comment relies heavily on the idea that ORSANCO’s pollution control standards are not necessary to carry out ORSANCO’s responsibilities under the Compact because states and the U.S. EPA are satisfying the requirements of this Compact. However, the premise behind Alternative #2 is more accurately articulated as ORSANCO abrogating its responsibilities under the Compact at a time when the Ohio River continues to need the clarity and support ORSANCO’s enforceable Pollution Control Standards provide.

A. The Existence of Federal and State Regulations is not Sufficient to Meet the Duties of ORSANCO under the Compact

While the Ohio River has improved in water quality since the making of the Compact, it remains the most contaminated river in the United States, and it provides drinking water for 5 million people. The preamble of the Compact states as follows:

The control of future pollution and the abatement of existing pollution in the waters of [the Ohio River] basin are of prime importance to the people thereof, and can best

be accomplished through the cooperation of the States situated therein, by and through a joint or common agency

This statement focuses on the need for Ohio River states to form an entity in order to best govern the Ohio River as a whole. It does not state that the federal government is the best entity to address the needs of the Ohio River. It also does not propose that states, independently, in implementing federally devised criteria, are in the best position to address the pollution facing the Ohio River. The fact that the Compact predates the existence of the Clean Water Act does not somehow make the needs and purposes it identifies less real.

The current leader of the United States' executive branch is an individual who vowed to eliminate the U.S. EPA once he took office, and since that time his administration has systematically worked to eliminate or reduce environmental protections. It is reprehensible that, in a time marked by dogged environmental deregulation by the federal government, a majority of ORSANCO Commissioners would argue that federal environmental regulations are adequate to meet ORSANCO's responsibilities. This is even more troublesome given the current political commitment to natural gas and petrochemical production expansion in the Ohio River basin at both the state and federal level.

The signatory states in and of themselves lack the regulatory authority necessary to carry out the stated goals of the compact. These states also differ dramatically in their approach to environmental regulation, including their respective implementations of the Clean Water Act through their approved state implementation plans. These states also differ dramatically in how well they regulate current and emerging threats to the Ohio River.

By way of example, a majority of FreshWater members reside in Ohio, where they live under a weak regulatory framework that daily fails to protect them from the pollution stemming from natural gas development and its associated waste disposal operations. Under Ohio law, the Ohio Department of Natural Resources ("ODNR") has sole and exclusive jurisdiction over most aspects of natural gas development and oil and gas waste disposal, a power it wields with little regard or opportunity for public oversight. The agency also lacks the environmental expertise and resources to monitor, assess, and address environmental problems rampant in this industry. Notably, ODNR is also not the agency in Ohio responsible for implementing the Clean Water Act.

Ohio's regulation of oil and gas waste disposal is one pervasive example of the problems associated with the state's weak regulatory framework for the natural gas industry that poses a threat to the health of the Ohio River. Wastes associated with hydraulic fracturing are known to be radioactive to varying degrees. Ohio accepts large amounts of these wastes, originating in Ohio and in other ORSANCO signatory states, for processing and disposal. These wastes typically consist of a mix of solids, liquids, and slurries that are transported to surface facilities where the waste is further processed, disposed of, or transferred to landfills for ultimate disposal. The processing at such facilities often includes the practice of "downblending," where radioactive waste is blended with other substances for disposal in landfills. Leachate from these landfills then goes to POTWs or is directly discharged into waterways under NPDES permits that lack standards for Ra-226 and Ra-228 or other radionuclides. Studies have found radioactivity to be a problem in leachate from landfills accepting oil and gas waste. Disposal of waste associated with oil and gas extraction into streams has also led to significant radioactivity accumulation near discharge sites.

Despite the immense associated environmental concerns, including known risks regarding radioactivity, oil and gas waste facilities in Ohio are regulated ad hoc through Chief's Orders issued by ODNR without guiding regulations in place. Environmental violations by the facilities are also addressed ad hoc through Chief's Orders issued by ODNR. A review of Chief's Orders for all such facilities in the state of Ohio demonstrates that these facilities are regularly violating the environmental provisions of their orders with little repercussion, and certainly without Ohio implementing any prescriptive regulations to protect the state's water resources from this type of waste. Notably, the majority of Ohio's oil and gas waste facilities are located within the Ohio River basin, with an increasing amount of them in close proximity to the River itself.

Ohio also permits the spreading of liquid waste ("brine") from oil and gas operations on roads within the Ohio River basin as a de-icer and for dust control with little processing prior to disposal in this manner. This is done without adequate testing for radioactivity prior to the substance being released on roadways. What minimal testing has been done on the brine being spread on Ohio roads shows the substance is laden with chemicals known to be harmful to human health and the environment, including benzene, toluene, ethylbenzene, xylene, and other Volatile Organic Compounds, as well as numerous metals. Disposing of oil and gas waste in this way is an example of yet another under-

regulated pathway through which the Ohio River is experiencing an onslaught of additional contaminants from the natural gas industry.

The issue of Ohio's oil and gas waste disposal regulations are but one example of the way states are failing the public and the Ohio River by not issuing adequate regulations. Such examples also exist for other signatory states including, at least, Kentucky, West Virginia, and Pennsylvania, as well as for industries other than oil and gas waste disposal. This inadequate regulation in no way satisfies the goals of the Compact or ORSANCO's duties under the Compact. As such, Alternatives #1 and #2, or any alternative eliminating or reducing ORSANCO's role in setting, maintaining, implementing, enforcing, and revising the Pollution Control Standards is an abrogation of ORSANCO's responsibilities under the Compact.

B. ORSANCO's Pollution Control Standards Include Criteria for Parameters Not Addressed in Federal and State Standards and Act to Implement State and Federal Standards in a coherent manner to protect the Ohio River as a Whole

As stated in the Commission's Minority Report, ORSANCO's Pollution Control Standards include criteria for 188 parameters for which the states or U.S. EPA have no such criteria. This alone should defeat any notion that ORSANCO standards are redundant to standards established by the U.S. EPA and signatory states' through their approved implementation programs under the federal Clean Water Act. Furthermore, and as also stated in the Commission's Minority Report, "the presence of an individual parameter in an approved state program under the Clean Water Act, or the presence of a mixing zone or antidegradation policy in an approved state program, does not assure that *implementation* of that standard or policy is being undertaken in a manner that is coherent among the states in the basin, or is sufficiently protective of the Ohio River downstream of the state in question."

These points are particularly salient when considering the impact of ORSANCO's criteria on the drinking water of 5 million people. In its comments on the PCS Review, the Water Users Advisory Committee, gives several examples of how ORSANCO's Pollution Control Standards protect drinking water in ways that state and federal regulations have not. One example is ORSANCO's ammonia criterion that was specifically designed to protect the public's drinking water from ammonia levels in source water that would compromise utilities' disinfection processes. ORSANCO's implementation of this criterion gave the public adequate protection for its sourcewater

to ensure drinking water was not compromised. Other ORSANCO criteria essential for protecting public drinking water, that are not otherwise adequately addressed in state or federal criteria, include E. coli; Fecal Coliform; temperature; combined radium-226 and radium-228; gross total alpha; total gross beta; total gross strontium-90; and silver.

These ORSANCO criteria are directly relevant to the risks posed by the continued and proposed growth of industries associated with natural gas extraction, production, transport, and waste disposal, including petrochemical production in the Ohio River basin. Much of this development exists or is proposed along, on, and even under the Ohio River itself. Radium-226 and radium-228 are known constituents of concern in flowback and wastes from the Marcellus and Utica shale. Despite this fact, there is simply no set of regulations at the state or federal level providing comprehensive protection from radioactive contaminants entering public drinking source water.

Headlines across the U.S. have made the public more aware that public drinking water systems are not miracle workers, that source water matters. Drinking water utilities can only do so much to address the contamination of their source water, and what they can do has real costs, costs that are often borne by the individuals, families, businesses, and institutions paying the water bills. ORSANCO has a critical role to play in drinking source water protection, a role that is currently not being filled by state or federal regulations. Because of its basin-wide jurisdiction, ORSANCO is uniquely positioned to address these grave concerns facing the Ohio River and the people who rely on it. ORSANCO should be using this unique position to carry out its duties under the Compact instead of proposing to neglect its responsibility in the face of immediate threats to the river's health.

C. The three-year review requirement provides the agility necessary to address current and developing threats to the Ohio River based on the most up-to-date science

The three-year review requirement for the Pollution Control Standards allows ORSANCO to adopt and implement criteria needed to protect the Ohio River and address immediate concerns more rapidly than may be possible for federal and state agencies. This agility is critical to respond to the actual needs of the River. The three-year review requirement is not superfluous, but rather one of the qualities that gives ORSANCO the potential to provide for the effective regulation of real threats when federal and state agencies lack the agility to respond in a timely fashion.

D. The make-up of ORSANCO works to ensure the Pollution Control Standards are set based on the needs of the Ohio River instead of the interests of Polluters

ORSANCO's unique jurisdiction and specific commitment to a water body should work to insulate ORSANCO from both local and national politics in a way that allows the Commission to set the standards that the Ohio River needs, not the standards purchased by moneyed interests.

Unfortunately, that the majority of Commissioner's desire to strip ORSANCO of its role in maintaining, administering, and periodically updating its Pollution Control Standards suggests that a large part of the Commission is not immune from such lobbying efforts by polluters. Indeed, the undue weight given to comments made by the Chemical Industry Advisory Committee and the Power Industry Advisory Committee indicate that the Commission is at risk of prioritizing industrial bottom lines over the drinking water source of over 5 million people and a valuable ecological system.

Comments submitted by the Chemical Industry Advisory Committee suggest ORSANCO is better suited to assessing water quality and engaging in spill detection and response. In other words, they suggest ORSANCO should continue the hard, resource-intensive work of understanding the river's problems, while rendering itself impotent to address those problems. This advisory group also states ORSANCO should be ready to clean up industries' messes, like the MCHM spill, when they occur and directly harm people's drinking water and ecological systems. This reading of ORSANCO's duties is in no way in alignment with the purpose and stated goals of the Compact.

Comments submitted by the Water Users Advisory Committee, the Watershed Organizations Advisory Committee, the Public Information Advisory Committee, the majority of comments offered by the Technical Committee, and multiple comments from the POTW Advisory Committee, all recognize the importance of ORSANCO's leadership role in setting, maintaining, and implementing its Pollution Control Standards. These groups represent the public in a way that the specificity of the industry groups do not. The majority of comments submitted by advisory committees state that it is undoubtedly in the public's interest for ORSANCO to continue its Pollution Control Standards, and that Alternative #2 is not acceptable.

ORSANCO should not relinquish its responsibilities under the Compact by reducing the Pollution Control Standards because industry has unpersuasively recommended they do so. Instead, ORSANCO should assume its role as a lead regulator for the Ohio River, just as it is asked to do by its originating document.

II. Of the Alternatives Presented for Consideration, Alternative #4 Best Fulfills ORSANCO's Responsibilities under the Compact

Alternative #4 addresses various concerns raised regarding effort duplication and regulatory inconsistencies by giving ORSANCO a leadership role in ensuring state regulations and implementation of standards are part of a coherent regulatory effort for the wellbeing of the Ohio River. This alternative reasonably addresses real concerns about the interaction of the Pollution Control Standards with state regulations and implementation, without abandoning ORSANCO's responsibilities as an entity uniquely charged with controlling and abating pollution of the Ohio River at large.

In addition, as stated by the Public Information Advisory Committee, Alternative #4 also increases ORSANCO's fundraising abilities. If a primary driver in eliminating the Pollution Control Standards is to better direct ORSANCO's limited resources, the elimination of the Pollution Control Standards will only work to further limit resources, giving ORSANCO even less ability to carry out its responsibilities under the Compact. ORSANCO (and, in turn, the Ohio River) is best served by ORSANCO assuming the leadership role presented in Alternative #4, which would make ORSANCO an entity worth funding.

It is also worth noting that the maintenance of existing expenditures of \$200,000 per year to support Technical and Management Staff in order to implement Alternative #4 is also the cost of having these good jobs within the Ohio River Basin and not simply some burdensome expenditure. Likewise, reductions in these budgets also result in reductions in quality employment.

III. ORSANCO's Call for Public Comment Did Not Provide Information Sufficient for Interested Parties to Evaluate the Alternatives Presented and Unreasonably Dissuaded Public Input

In its call for public comment, the information presented to the public on the proposed alternatives is skewed in favor of Alternative #2. Despite asking the public for input on

all aspects of the standards and on the five alternatives proposed for the future of the Pollution Control Standards, ORSANCO provides no expanded analysis for the other alternatives nor any mockup of the rules for these alternatives. Thus, the public is left with limited information on the other alternatives to use in making comments. Further, ORSANCO's statement regarding a majority of commissioners' preference for Alternative #2 serves to dissuade the public from meaningfully participating in the public comment process and demonstrates ORSANCO's willingness to serve industrial interests over carrying out its responsibilities under the Compact.

The notice contains little to no information on the cost of each alternative and the impact of each alternative on various grants available to ORSANCO. Likewise, the information available to the public provides no side by side analyses of the vast network of laws the public is being asked to compare. ORSANCO should make this information easily accessible to the public in order for the public to be able to more specifically comment on the alternatives presented.

Conclusion

The preamble of the Compact states:

[T]he growth in industrial activity within [the Ohio River basin], [has] resulted in recent years in an increasingly serious pollution of the waters and streams within the said drainage basin, constituting a grave menace to the health, welfare and recreational facilities of the people living in such basin, and occasioning great economic loss

While the Ohio River has improved in many ways since 1948, this statement remains very true. The Ohio River is still in need of a coordinated effort to protect its many uses, and ORANSCO is charged with leading that effort. FreshWater urges the Commission to embrace its unique position in governing the Ohio River and to work with states in a leadership role in protecting this invaluable fresh water resource. FreshWater urges ORSANCO to continue to maintain, implement, and revise its Pollution Control Standards. If one of the alternatives presented is to be selected, FreshWater strongly advocates for Alternative 4.

Respectfully Submitted,

A handwritten signature in black ink, appearing to read 'M. Hunter', written in a cursive style.

Megan M. Hunter (0096035)
Hunter & Hunter LLC
640 Ardleigh Dr.
Akron, OH 44303
Telephone: 234-281-2528
Facsimile: 330-805-4518
megan@hunterfirm.org

Attorney for Fresh Water Accountability Project

Exhibit B

Comparison of Minimum Water Quality Criteria vs ORSANCO's 122 Minimum Criteria (12/3/2015)

State/USEPA	Total Number of Parameters Regulated by the State Agency or Recommended by the USEPA	No. of Parameters for Which ORSANCO has criteria but the Agency does not	No. of Parameters for which the Agency has Criteria but ORSANCO does not	No. of Parameters for Which the Agencies' Criteria are Less Stringent Than ORSANCO (more than 10% difference)	No. of Parameters for Which the Agencies' and ORSANCO's Criteria are Equivalent (less than 10% Difference)	No. of Parameters for Which the Agencies' Criteria are More Stringent than ORSANCO (more than 10% difference)	Number of Standards Compared
Illinois	238	44	160	38	5	35	78
Indiana	90	50	18	53	10	9	72
Kentucky	137	13	28	10	94	5	109
Ohio	149	17	44	87	13	5	105
Pennsylvania	140	20	38	7	73	22	102
West Virginia	95	38	11	33	38	11	84
USEPA	154	11	43	23	17	67	111
Totals	1003	193	342	251	250	154	661

Key Assumptions/Factors Utilized in Preparation of Comparison

- 1) The most stringent criterion for any parameter is utilized for this comparison regardless of differences in the intended protected use.
- 2) The comparison removes the separation of standards by criteria type: eg. "Human Health" and "Aquatic Life".
- 3) Comparison shows where ORSANCO Pollution Control Standards require discharge permits on the Ohio River to be more stringent than the state would allow on other bodies of water.
- 4) Some parameters are not included in this comparison. These pollutants include Aesthetics, Ammonia, Bacteria, Boron, Color, Diocetylphthalate, Total Dissolved Gases, Hardness, Nitrosamines, Nutrients.
- 5) Parameters included in this summary are taken from the individual State Agency's Website and do not reflect individual State's inclusion of ORSANCO Standards through other means such as adoption of ORSANCO standards by reference, through individual permit issuance or other means.
- 6) There may be other assumptions/combinations/arrangements that staff is not aware of at this time.

Tests of ORSANCO PCS vs USEPA and Mainstem States Water Quality Standards

Human Health (Water and Organism)

Test	Pollutants between	Shared Pollutants	Identical Criterion	Different Criteria
ORSANCO vs USEPA	125	52	7	45
ORSANCO vs Illinois	65	40	3	37
ORSANCO vs Indiana	88	34	0	34
ORSANCO vs Kentucky	110	52	7	45
ORSANCO vs Ohio	109	52	0	52
ORSANCO vs Pennsylvania	115	51	47	4
ORSANCO vs West Virginia	104	39	13	26

Human Health (Water Only)

Test	Pollutants between	Shared Pollutants	Identical Criterion	Different Criteria
ORSANCO vs USEPA*	123	42	2	40
ORSANCO vs Illinois	72	10	0	10
ORSANCO vs Indiana	110	26	3	23
ORSANCO vs Kentucky	130	47	44	3
ORSANCO vs Ohio	155	47	7	40
ORSANCO vs Pennsylvania**	54	0	0	0
ORSANCO vs West Virginia*	98	29	1	28

* USEPA and West Virginia are "Organism Only"

** Pennsylvania does not have two types of human health criteria

Aquatic Life Chronic

Test	Pollutants between	Shared Pollutants	Identical Criterion	Different Criteria
ORSANCO vs USEPA	48	12	5	7
ORSANCO vs Illinois	221	0	0	0
ORSANCO vs Indiana	30	9	1	8
ORSANCO vs Kentucky	39	11	4	7
ORSANCO vs Ohio	14	0	0	0
ORSANCO vs Pennsylvania	108	11	4	7
ORSANCO vs West Virginia	32	11	5	6

Aquatic Life Acute

Test	Pollutants between	Shared Pollutants	Identical Criterion	Different Criteria
ORSANCO vs USEPA	38	11	3	8
ORSANCO vs Illinois	222	0	0	0
ORSANCO vs Indiana	32	9	0	9
ORSANCO vs Kentucky	34	11	3	8
ORSANCO vs Ohio	13	0	0	0
ORSANCO vs Pennsylvania	108	11	3	8
ORSANCO vs West Virginia	34	12	4	8

Notes:

1. Ohio Aquatic Life Criteria not available
2. Narrative Criteria (23 of 2314 total standards) not yet analyzed

Number of Criteria of Each Type

State	Agency	AL Acute	AL Chronic	HH Water and Organism	HH Water only	Organism only	Other, Organoleptic	Total
ORSANCO	ORSANCO	14	14	57	54		4	143
USEPA	USEPA	43	46	121		111	12	333
Illinois	ILEPA	215	213	50	29			507
Indiana	IDEM	28	26	65	82			201
Kentucky	KYDEP	32	36	105	123			296
Ohio	OH EPA			104	148			252
Pennsylvania	PADEP	106	105					320
West Virginia	WVDEP	40	32	92		78	20	262
								2314

Number of Pollutants Regulated

Agency	CAS#
ORSANCO	122
USEPA	154
ILEPA	238
IDEM	90
KYDEP	137
OH EPA	149
PADEP	140
WVDEP	95

Unassessed Pollutants

Pollutant	# of Agencies	Reason
Aesthetic Qualities	1	Narrative or seasonally various
Ammonia	3	Narrative or seasonally various
Bacteria	1	Narrative or seasonally various
Boron	1	Narrative or seasonally various
Color	1	Narrative or seasonally various
diethylphthalate	2	"NI"
Gases, Total Dissolved	1	Narrative or seasonally various
Hardness	1	Narrative or seasonally various
Nitrosamines	1	Narrative or seasonally various
Nutrients	1	Narrative or seasonally various
Oil and Grease	1	Narrative or seasonally various
Oxygen, Dissolved	1	Narrative or seasonally various
Pathogen and Indicators	2	Narrative or seasonally various
pH	2	Narrative or seasonally various
Solids, Dissolved/Salinity	1	Narrative or seasonally various
Solids, Suspended/Turbidity	1	Narrative or seasonally various
Tainting Substances	1	Narrative or seasonally various
Temperature	5	Narrative or seasonally various
Turbidity	6	Narrative or seasonally various

Exhibit C



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



Robert L. Herrick^a, Jeanette Buckholz^a, Frank M. Biro^b, Antonia M. Calafat^c, Xiaoyun Ye^c, Changchun Xie^a, Susan M. Pinney^{a,*}

^a University of Cincinnati College of Medicine, Department of Environmental Health, Cincinnati, OH, United States

^b Division of Adolescent Medicine, Cincinnati Children's Hospital Medical Center, Cincinnati, OH, United States

^c Division of Laboratory Sciences, National Center for Environmental Health, Centers for Disease Control and Prevention, Atlanta, GA, United States

ARTICLE INFO

Article history:

Received 11 January 2017

Received in revised form

25 April 2017

Accepted 29 April 2017

Keywords:

Biomonitoring

Drinking water contaminants

Granular activated carbon (GAC)

Per- and polyfluoroalkyl substances (PFAS)

Pharmacokinetic modeling

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.

© 2017 Elsevier Ltd. All rights reserved.

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 photographs/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

[☆] This paper has been recommended for acceptance by Prof. von Hippel Frank A.

* Corresponding author. 208 Kettering Laboratory, 160 Panzeca Way, Cincinnati, OH 45269-0056, United States.

E-mail address: pinneysm@ucmail.uc.edu (S.M. Pinney).

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

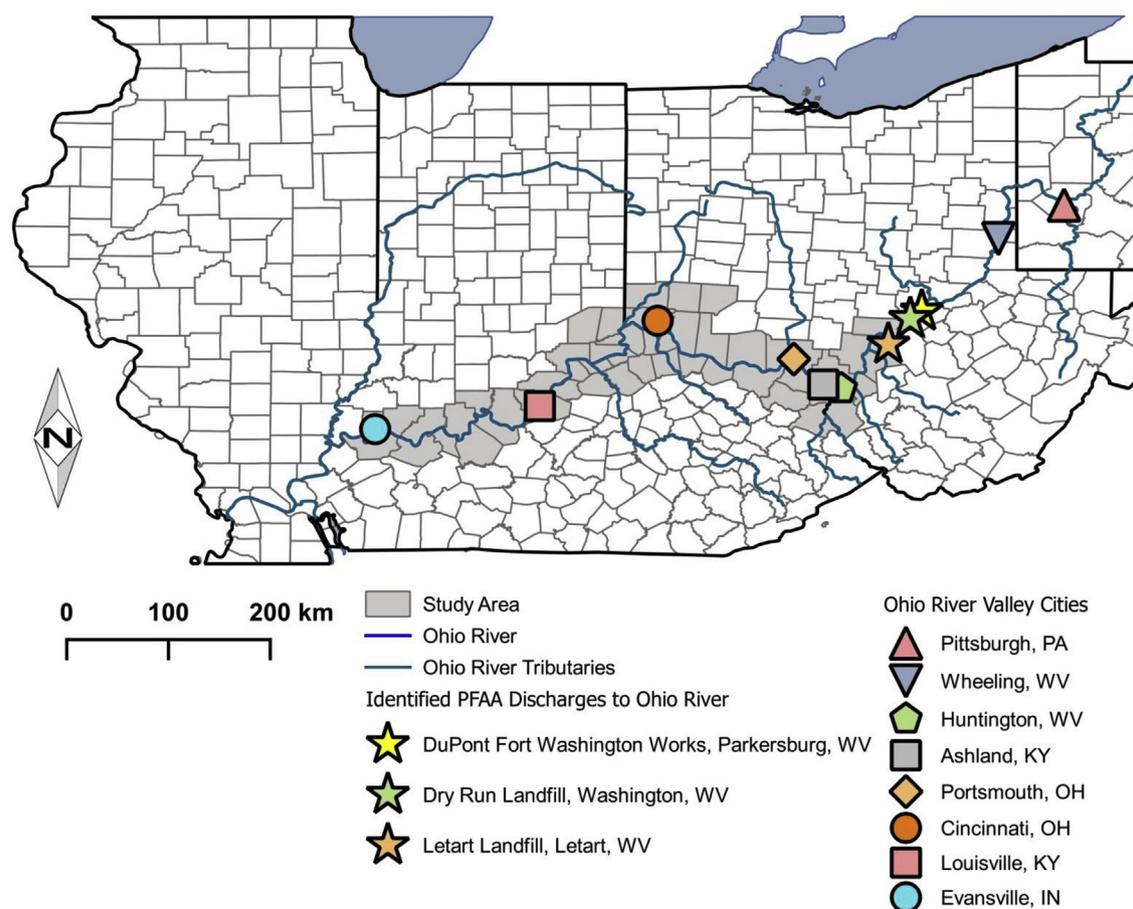


Fig. 1. The Ohio River Valley.

included. Participants were eligible if they had lived in the area for 2 or more years and were aged 2–85 years. ORV participants were not followed-up. BCERP participants were recruited as girls aged 6–8 years from Greater Cincinnati schools and Breast Cancer Registry of Greater Cincinnati participant families between 2004 and 2007. Follow-up continued until 2015. Study design and participant characteristics were previously described (Biro et al., 2010). Institutional review boards (IRB) at the University of Cincinnati and Cincinnati Children's Hospital Medical Center approved study protocols. Participant/parent consent and child assent were obtained. The Centers for Disease Control and Prevention (CDC) IRB approved the analysis of serum specimens with no personal identifiers for BCERP. For FCC and ORV, CDC laboratory involvement did not constitute engagement in human subjects research because samples were completely de-identified.

2.2. Serum samples and analysis

We obtained serum at baseline, second and/or final medical exam from FCC participants (1991–2008), at recruitment for ORV participants (2009–2012), and annually for BCERP participants (2005–2013) using protocols and materials provided by CDC. Sera were stored at -80°C until analysis. All available samples for FCC and ORV participants were analyzed. All BCERP participants with blood samples had baseline samples measured. Subsequent samples from BCERP participants were analyzed if available.

CDC staff measured serum PFAS concentrations at various times between 2005 and 2014 using published methods (Kato et al., 2011; Kuklenyik et al., 2005). CDC measured 2-(*N*-ethyl-perfluorooctane sulfonamido) acetic acid (Et-PFOSA-AcOH or EtFOSAA), 2-(*N*-methyl perfluorooctane sulfonamido) acetic acid (Me-PFOSA-AcOH or MeFOSAA), perfluorohexane sulfonic acid (PFHxS) and perfluorononanoic acid (PFNA) in all samples. CDC measured total PFOA and total PFOS in ORV, FCC and baseline BCERP samples. Linear (*n*-PFOA) and non-linear (branched) PFOA and linear (*n*-PFOS), methyl (Σm -PFOS) and dimethyl (Σm_2 -PFOS) PFOS isomer concentrations were reported separately for non-baseline BCERP samples (collected 2006–2013); we calculated total PFOA and PFOS concentrations as the sums of their respective linear and branched isomers. Perfluorodecanoic acid (PFDeA) was not measured in the first 82 BCERP samples. Perfluorobutane sulfonic acid (PFBS), perfluorododecanoic acid (PFDoA), perfluoroundecanoic acid (PFUA) and perfluorooctane sulfonamide (PFOA or FOSAA) were not measured in all samples because more than 80% of results were below limits of detection (LOD) in initial analyses.

2.3. Covariates

Questionnaires, including demographics, beverage consumption, residential, medical and reproductive histories, were administered annually to adult participants and parents of minors (BCERP and FCC), or at recruitment (ORV). FCC participants answered additional residential history and beverage consumption questions when recontacted. Participants used glassware or life-size illustrations to estimate beverage consumption. FCC participants received medical exams every two (1997–2008) or three years (1990–1996). We collected anthropometrics at recruitment for ORV participants. Physical exams were administered semi-annually to BCERP participants until 2010, then annually. Calibrated stadiometers and scales were used for anthropometrics. We obtained participants' residential histories from 1980 (for those born before 1980) or birth (for those born after) by interview (BCERP, ORV) or mailed questionnaire (FCC). We identified water utilities, sources, service areas, service dates and GAC use from 1980 to 2012 through utility and public record searches, resulting in 221 water source-

time zones, served by 114 utilities, utilizing 23 water sources (Pinney et al., 2014). Water sources, durations and GAC treatment were identified for each residential address of each participant.

2.4. Statistical analysis

Statistical analyses were conducted with SAS v9.2 (SAS Institute, Cary NC). PFAS concentrations were non-normal (skewness: 2.29–9.44) and log-transformed for analyses; LOD/ $\sqrt{2}$ was imputed for values below LOD (Hornung and Reed, 1990). We calculated sample-weighted Pearson correlations and descriptive statistics for PFAS and water sources, and then estimated serum PFOA-covariate associations using two linear mixed-effects models with random subject effects (PROC MIXED).

Model 1 categorized participants' water source at sample collection in eight groups: Ohio River in Huntington, Ashland/Ironton/Portsmouth, Northern Kentucky and Cincinnati, Ohio River Aquifer between Huntington-Cincinnati and Cincinnati-Louisville, the Great Miami Aquifer and all other water sources. Model 2 used participants' cumulative years the Ohio River, with and without GAC, and Ohio River Aquifer were municipal water sources. Sample collection year, sex, age at sample collection, daily tap water consumption, bottled water use, water filter use, pregnancy status, reproductive history and breastfeeding were covariates. Race/ethnicity was not included in analyses because 93% of non-Caucasians used the same water utility. Water source was always included in regression models; other covariates were retained if significant or caused a 15% or greater change in water source effect estimates (backwards/stepwise elimination). Linear regression analyses for other PFAS with 80% or more results above the LOD were conducted using significant terms from PFOA models as covariates, similar to Pinney et al. (2014). We approximated model r^2 as variance change between full/final and intercept-only models; intercept model random variance was constrained to that in full/final models.

2.5. Pharmacokinetics

Serum PFOA concentrations between samples were estimated using a one-compartment (serum) pharmacokinetic model for 508 participants from the FCC and BCERP cohorts with multiple samples (Thompson et al., 2010). Using this model, we examined two exposure scenarios. In Scenario 1, we assumed Ohio River PFOA concentrations from 1991 to 2013 were the same as those measured in September 2009 (Emery et al., 2010). In Scenario 2, we estimated annual PFOA Ohio River concentrations for 1991 to 2013 by linearly-interpolating between reported concentrations, assuming constant ratios between regions (Table S1). For both scenarios, we assumed aquifer-exchange or GAC treatment reduced PFOA concentrations by 50% and other source waters were uncontaminated. We used participant-reported tap water consumption for daily intake, and we assumed body weight changed linearly between measurements, to estimate PFOA dose from water utility PFOA concentration estimates. For both scenarios, non-water PFOA dose was estimated from NHANES median PFOA serum concentrations (CDC, 2015), assuming steady state. Because reported half-life estimates for PFOA vary widely, we conducted a sensitivity analysis; we calculated PFOA elimination constants using half-life estimates from 1.7 to 3.8 years, which cover estimates from the literature (Fu et al., 2016; Bartell et al., 2010; Brede et al., 2010; Olsen et al., 2007). We estimated model fit using Wilcoxon rank-sum tests on median difference, weighted kappa (κ) for PFOA quartile and interclass-correlations (ICCs), comparing predicted to subsequent measured serum PFOA concentration. Model fit statistics were calculated for all estimates, and separately by starting

PFOA concentration (above/below the mean), and by cohort (FCC vs BCERP).

3. Results

3.1. Descriptive characteristics

931/1201 participants provided data (Table 1). 1608 separate serum samples were analyzed. Serum was collected in Huntington 2009–2010, Portsmouth 2009–2011, and Greater Cincinnati, and regions between Portsmouth and Louisville, 1991–2013. Participants were 15% African-American, 2% Hispanic, 1% Asian and 82% Caucasian. FCC participants were exclusively Caucasian; ORV participants were 96% Caucasian. FCC and ORV cohorts had similar ages at sample collection, tap water consumption, proportions of women ever pregnant and breastfeeding, mean gravidity and children breastfed per mother. ORV children were breastfed longer than BCERP participants. We identified water sources for 95% of participants' addresses. 72.5% of participants used the Ohio River and 13.5% used the Ohio River Aquifer. Of those using the Ohio River, 37.2% always had GAC filtration, 44.7% never did, and 18.1% had GAC for some of the time their municipal utility used the Ohio River as a water source. Mean Ohio River water use was 7.5 years (range 0–32 years); mean Ohio River Aquifer water use was 1.2 years (range 0–32 years).

3.2. Serum PFAS concentrations

Unless indicated, serum PFOS and serum PFOA refer to total concentrations. PFHxS, PFOA, PFOS and PFNA were detected in greater than 99% of samples; Me-PFOA-AcOH and PFDeA were detected in more than 82% (Table S2; Fig. 2). PFBuS and PFDoA were never detected. PFOA and PFOS isomers were measured in 350/

1608 serum samples; *n*-PFOA was detected in 99.7% and branched PFOA in 44.6%. Mean serum PFOA was 97% linear, 3% branched. Mean serum PFOS was 71% *n*-PFOS, 28% Σm -PFOS, and 1% Σm_2 -PFOS.

Serum PFOA was highest in samples collected 1991–1993 (median = 13.8 ng/mL), and decreased for each sample year group to 2011–2013 (median = 4.3 ng/mL). Similar decreases were observed for *n*-PFOA (median = 5.6 ng/mL in 2007–2008; median = 2.6 ng/mL in 2011–2013). Median serum PFOS was highest 1994–1996 (32.2 ng/mL), decreasing to 6.3 ng/mL in 2011–2013; *n*-PFOS, Σm -PFOS and Σm_2 -PFOS concentrations decreased proportionately. For participants with multiple samples, median serum concentrations for measured PFAS decreased between samples (Table S3); median between-sample total PFOA decrease was 2.3 ng/mL (29%) and median total PFOS decrease was 4.9 ng/mL (37%) over a mean between-sample interval of 4.5 years.

Weighted PFOA-PFAS Pearson correlations (Table S4) were significant ($p > 0.05$) for all but PFOA-PFHxS and PFOA-PFUA. These correlations were moderate (PFOS) or weak (Et-PFOA-AcOH, Me-PFOA-AcOH, PFOSA, PFHxS, PFNA, PFDeA, PFUA); all were less than 0.5. Total and *n*-PFOA, total and *n*-PFOS, total and Σm -PFOS, and *n*-PFOS and Σm -PFOS were strongly correlated. Total PFOA-branched PFOA and *n*-PFOA-branched PFOA concentrations were weakly correlated.

3.3. Serum PFOA determinants

Water source was statistically significant in both PFOA regression models (Table 2). Final models explained 44–47% of serum PFOA variation (r^2 : 0.44–0.47). In Model 1, covariate-adjusted geometric mean (GM) serum PFOA was significantly higher for participants using Ohio River water in Huntington (8.9 ng/mL), Ashland, KY to Portsmouth (12.7 ng/mL), and Northern Kentucky

Table 1
Participant descriptive characteristics.

Study characteristic	FCC	ORV	BCERC	All three studies
Participants with samples	210 (22.6%)	366 (39.3%)	355 (38.1%)	931 (100.0%)
Serum samples	517 (32.2%)	366 (22.8%)	725 (45.1%)	1608 (100.0%)
Mean sample interval (years)	7.5 ± 4.7	–	2.1 ± 0.5	4.5 ± 4.2
Sample collection year				
1991–1993	139 (26.9%)	0 (0.0%)	0 (0.0%)	139 (8.6%)
1994–1996	189 (36.6%)	0 (0.0%)	0 (0.0%)	189 (11.8%)
1997–2000	40 (7.7%)	0 (0.0%)	0 (0.0%)	40 (2.5%)
2001–2006	3 (0.6%)	0 (0.0%)	229 (31.6%)	232 (14.4%)
2007–2008	146 (28.2%)	0 (0.0%)	320 (44.1%)	466 (29.0%)
2009–2010	0 (0.0%)	256 (69.9%)	153 (21.1%)	409 (25.4%)
2011–2013	0 (0.0%)	110 (30.1%)	23 (3.2%)	133 (8.3%)
Water source				
Mean Ohio River water years (without GAC)	0.49 ± 1.67	5.48 ± 9.32	4.02 ± 4.19	3.17 ± 5.72
Mean Ohio River water years (with GAC)	2.70 ± 5.03	1.76 ± 3.52	1.76 ± 3.52	4.30 ± 7.82
Mean Ohio River Aquifer water years	2.68 ± 5.62	0.04 ± 0.35	0.04 ± 0.35	1.24 ± 4.28
Age at sample (years)	46.8 ± 14.7	41.2 ± 16.2	9.0 ± 1.5	28.5 ± 21.1
Sex				
Female	322 (62.3%)	263 (71.9%)	725 (100.0%)	1310 (81.5%)
Male	195 (37.7%)	103 (28.1%)	0 (0.0%)	298 (18.5%)
Drank tap water	467 (90.3%)	338 (92.3%)	639 (88.1%)	1444 (89.8%)
Mean tap water consumption (L/day)	2.6 ± 1.7	2.6 ± 2.5	0.7 ± 0.6	1.24 ± 4.28
Drank bottled water	199 (38.5%)	308 (84.2%)	470 (64.8%)	977 (60.8%)
Used a water filter	114 (22.1%)	155 (42.3%)	331 (45.7%)	600 (37.3%)
Ever pregnant ^a	264 (82.0%)	206 (78.3%)	0 (0.0%)	470 (35.9%)
Pregnant at serum collection ^a	4 (1.2%)	15 (5.7%)	0 (0.0%)	19 (1.5%)
Mean gravidity ^a	3.1 ± 1.8	2.6 ± 1.7	–	2.8 ± 1.8
Ever breastfed a child ^a	133 (41.3%)	103 (39.2%)	0 (0.0%)	236 (18.0%)
Mean number of children breastfed ^a	1.1 ± 1.3	0.9 ± 1.1	–	1.0 ± 1.2
Breastfed as a child ^b	0 (0.0%)	13 (61.9%)	474 (65.4%)	478 (64.8%)
Mean breastfeeding duration (months) ^b	–	5.4 ± 6.8	3.7 ± 5.8	3.8 ± 5.8

^a Adult females only.

^b Children only.

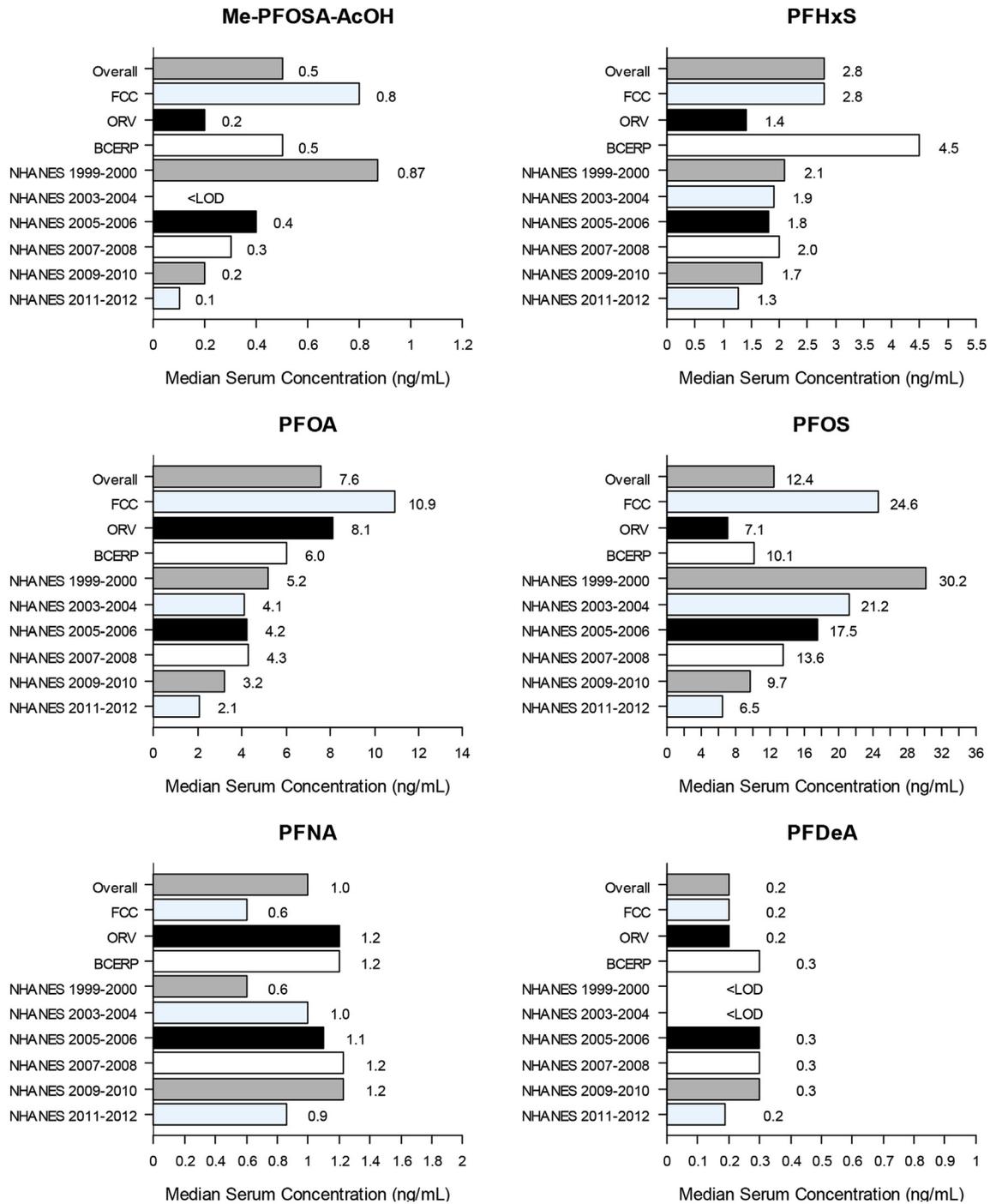


Fig. 2. Median Serum PFAS Concentrations. Notes: Only PFAS with >40% above LOD across all three cohorts shown. Total sample median used for NHANES data (CDC, 2015).

Cincinnati suburbs (10.4 ng/mL), but not Cincinnati itself (6.0 ng/mL), than non-Ohio River water sources (5.6 ng/mL). Serum PFOA was also significantly elevated for participants using the Ohio River Aquifer between Huntington and Cincinnati (GM = 8.0 ng/mL), but not between Cincinnati and Louisville (GM = 4.4 ng/mL). In Model 2, serum PFOA concentration significantly increased with each year the Ohio River or Ohio River aquifer was the municipal water source.

Serum PFOA declined with sample year (linear trend $p < 0.0001$) in both models. Covariate-adjusted GM serum PFOA was highest in samples drawn 1991–1993 (11.6/12.8 ng/mL) and lowest

2011–2013 (3.3/3.0 ng/mL) for Models 1 and 2, respectively. Water source and sample year interactions in Model 1 ($p < 0.0001$) revealed different temporal trends by region (Fig. 3). Serum PFOA was highest for participants using Cincinnati Ohio River water 1991–1993 (GM = 16.4 ng/mL) and declined subsequently; serum PFOA increased in participants using Northern Kentucky Ohio River water from 1991–1993 (GM = 13.5 ng/mL) to 2001–2006 (GM = 16.2 ng/mL) before declining. Serum PFOA in participants using the Ohio River Aquifer was constant 1991–2006 before declining 2007–2010. Serum PFOA for participants using the Great Miami Aquifer or other water sources was highest in 1991–1993

Table 2
Serum PFOA regression models.

	Model 1: Water Source at Sample				Model 2: Cumulative Water Years			
	Full model		Final model		Full model		Final model	
Total samples	1608		1608		1608		1608	
Samples in analysis	1472		1491		1431		1450	
r ²	0.47		0.45		0.45		0.44	
Model Parameters	β estimate	p-value	β estimate	p-value	β estimate	p-value	β estimate	p-value
Water source								
Ohio River (Huntington, GAC)	0.450	<0.0001	0.456	<0.0001				
Ohio River (Ashland/Ironton/Portsmouth)	0.830	<0.0001	0.816	<0.0001				
Ohio River Aquifer (Huntington-Cincinnati)	0.351	0.0001	0.356	<0.0001				
Ohio River (Northern Kentucky)	0.632	<0.0001	0.610	<0.0001				
Ohio River (Cincinnati, GAC)	0.075	0.2163	0.061	0.313				
Ohio River Aquifer (Cincinnati-Louisville)	-0.236	0.0081	-0.251	0.005				
Great Miami Aquifer	-0.041	0.5076	-0.052	0.400				
All other water sources	0.000	–	0.000	–				
Cumulative Ohio River water years (without GAC)					0.041	<0.0001	0.043	<0.0001
Cumulative Ohio River water years (with GAC)					0.015	<0.0001	0.016	<0.0001
Cumulative Ohio River Aquifer water years					0.015	0.001	0.017	0.000
Sample year								
1991–1993	1.277	<0.0001	1.254	<0.0001	1.439	<0.0001	1.436	<0.0001
1994–1996	1.156	<0.0001	1.148	<0.0001	1.320	<0.0001	1.327	<0.0001
1997–2000	1.069	<0.0001	1.057	<0.0001	1.260	<0.0001	1.258	<0.0001
2001–2006	1.041	<0.0001	1.037	<0.0001	1.165	<0.0001	1.158	<0.0001
2007–2008	0.648	<0.0001	0.641	<0.0001	0.717	<0.0001	0.707	<0.0001
2009–2010	0.338	<0.0001	0.336	<0.0001	0.415	<0.0001	0.412	<0.0001
2011–2013	0.000	–	0.000	–	0.000	–	0.000	–
Age at sample (years)	0.009	<0.0001	0.009	<0.0001	0.006	<0.0001	0.006	<0.0001
Sex								
Female	-0.042	0.509			-0.081	0.215		
Male	0.000	–			0.000	–		
Tap water consumption (L/day)	0.021	0.055	0.024	0.024	0.020	0.076	0.022	0.040
Drank bottled water								
Yes	0.061	0.077			0.064	0.074		
No	0.000	–			0.000	–		
Used a water filter								
Yes	0.040	0.238			0.029	0.406		
No	0.000	–			0.000	–		
Pregnant at serum collection								
Yes	-0.632	0.000	-0.493	0.001	-0.473	0.005	-0.395	0.008
No	0.000	–	0.000	–	0.000	–	0.000	–
Gravidity	-0.028	0.112	-0.037	0.021	-0.026	0.157	-0.041	0.011
Number of children breastfed	-0.078	0.007	-0.071	0.014	-0.081	0.006	-0.074	0.012

and declined throughout.

Pregnancy, breastfeeding and gravidity were significantly inversely associated with serum PFOA in both models. Serum PFOA significantly increased with age and tap water consumption. Serum PFOA was significantly greater in males than females in univariate models ($p < 0.0001$), but not multivariate analyses. Bottled water consumption and water filter use were non-significantly associated with serum PFOA.

We conducted linear regression analyses of other PFAS with more than 80% above LOD using both models. Water source was significantly associated with Me-PFOA-AcOH, PFHxS, total PFOS and PFNA, but not PFDeA (Table S5). Adjusted serum concentrations of Me-PFOA-AcOH, PFHxS, PFNA and PFDeA did not differ widely across regions (maximum difference 0.7 ng/mL) (Table S7). Total PFOS varied between Huntington, WV and other regions (maximum difference 4.3 ng/mL), but other adjusted between-region total PFOS differences were small. In Model 2 (Table S6), cumulative Ohio River water years without GAC was significantly associated with increasing total serum PFOS and PFNA, but not Me-PFOA-AcOH, PFHxS, or PFDeA. Cumulative Ohio River water years with GAC was associated with significantly decreasing serum PFHxS and total PFOS. Effect estimates for cumulative Ohio River water years with GAC were lower than those for cumulative Ohio River water years without GAC.

3.4. Pharmacokinetics

We estimated serum PFOA between measured concentrations for 508 FCC and BCERP study participants with multiple samples (Fig. S1). Overall, Scenario 2 and elimination constants based on a 3.8-year half-life best replicated measured serum concentrations (Table S8). When we stratified model fit by starting serum PFOA concentration (>12.9 ng/mL vs ≤ 12.9 ng/mL), Scenario 2 and a 3.8-year was the best fit for higher starting PFOA concentrations, while Scenario 1 and a 3.8-yr half-life was a better fit for lower starting PFOA concentrations (Table S9). Similar fit differences were present when stratified by cohort; Scenario 2 and a 3.8-year half-life better fit FCC participants, while Scenario 1 and a 3.8-yr half-life better fit BCERP participants' measured serum concentrations (Table S10). Model fit improved as estimated half-lives increased from 1.7 years to 3.8 years under both scenarios, and across all strata. Under Scenario 1, predicted serum PFOA was highest in 1991 and decreased throughout the study period. Under Scenario 2, predicted GM serum PFOA increased through the 1990s, peaking in 2000/2001 (Fig. S1).

4. Discussion

To our knowledge, this is the first study to report on serum

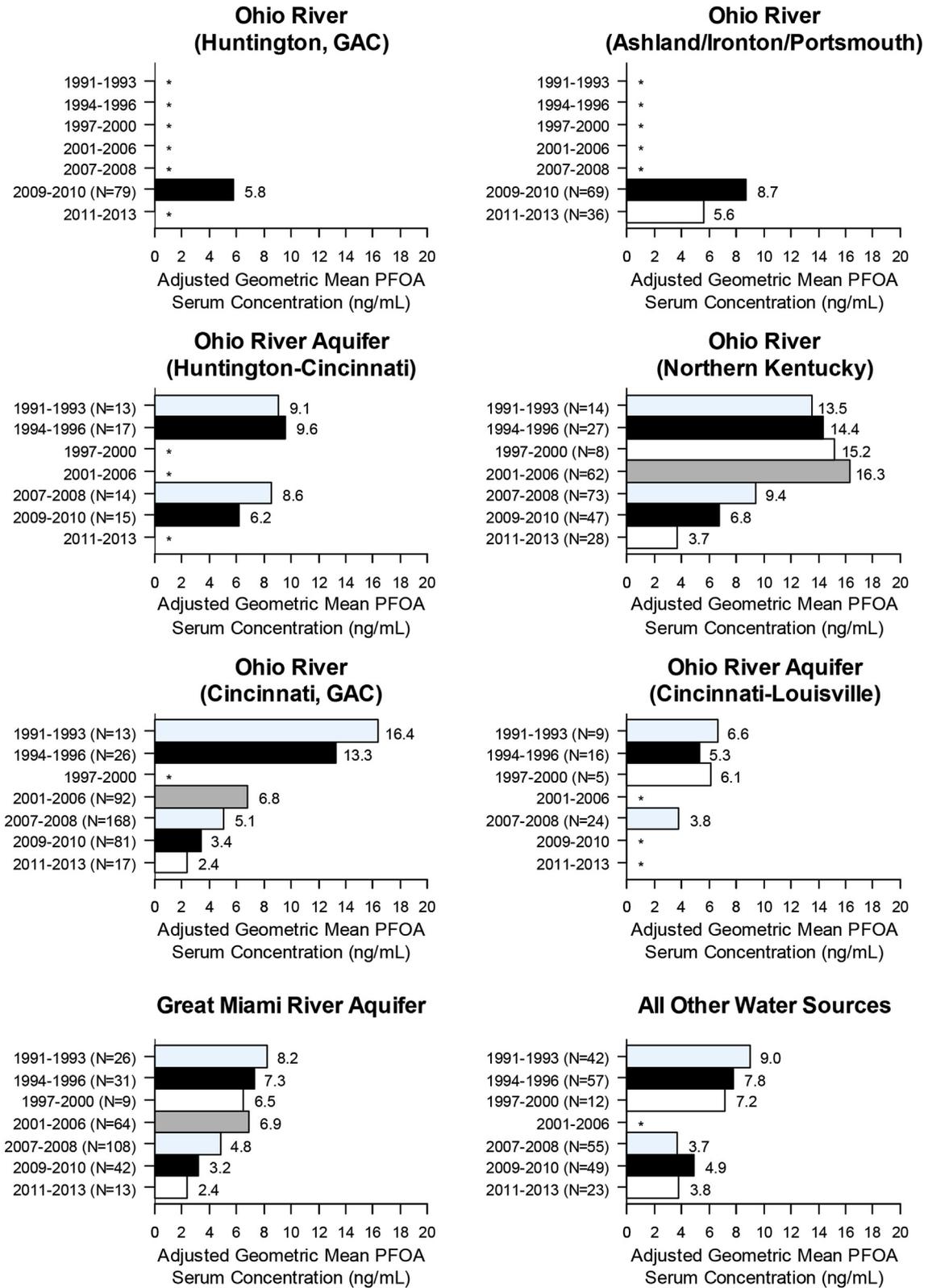


Fig. 3. Adjusted GM serum PFOA concentrations by water source and sample year. Notes: Adjusted for water source, sample year, age at sample, tap water consumption, pregnancy status, gravidity and number of children breastfed. * < 5 participants used this water source during these years.

concentrations of PFOA in samples obtained as early as 1991, with water supply source information for each study participant. However, earlier publications noted the presence of organic fluorinated

compounds in human plasma. Organic fluorinated compounds were noted in human plasma in blood bank samples collected from 106 persons living in five different US cities in a 1976 publication,

with speculation that they originated from an environmental source. The organic fluorine concentrations in plasma (1.35 μM or 0.025 ppm) were not related to the amount of fluoride in the water supply (Guy et al., 1976). The dominant peak in silica acid column chromatography showed a “nmr pattern consistent with a derivative of perfluorinated octanoic acid” (Taves et al., 1976). In 1981, Belisle (1981) reported on finding trace amounts of organic fluorine in 8 samples of human blood obtained from donors in a Chinese rural commune, very unlikely to have exposure to industrial sources of fluorochemicals, but lower than the amounts found in persons living in more urban areas. They also noted that the analytic method may have introduced the organic compounds.

Median serum PFOS (12.4 ng/mL), PFDeA (0.2 ng/mL), PFHxS (2.8 ng/mL), PFNA (1.0 ng/mL) and Me-PFOA-AcOH (0.5 ng/mL) concentrations in our participants were comparable to median NHANES concentrations. Median serum PFOA concentration (7.6 ng/mL) was greater than NHANES, and individual serum PFOA concentrations were greater than corresponding NHANES 95th percentiles for 47% of participants. Residents' PFOA serum concentrations were lower than in participants of the C-8 study in 2005–2006 (28.8 ng/mL) (Frisbee et al., 2009), comparable to those in the baseline (2005–2007) BCERP Cincinnati-site participant subset of this cohort we previously reported (6.4 ng/mL) (Pinney et al., 2014), and greater than HOME participants 2003–2006 (3.4–5.5 ng/mL) (Kato et al., 2014; Braun et al., 2016). These findings suggest Mid-Ohio River Valley residents were exposed to PFOA, but not the other PFAS we measured, above background levels. However, additional PFAS continue to be identified, and these may become more environmentally relevant as PFOA and PFOS are phased out and decrease in human serum (Newton et al., 2017); exposure to these PFAS is unknown in our participants.

While overall serum PFOA levels decreased with time in our population, we identified distinct temporal trends across regions; these may be explained by sourcewater and drinking water treatment technology differences. Residents using the Great Miami Aquifer and other water sources show a gradual decrease in PFOA similar to that identified in NHANES surveys (CDC, 2015). This suggests that PFOA contamination in these waters was minimal, and that their PFOA exposures decreased as PFOA levels in other sources, most likely food and house dusts, decreased. Both regions served by the Ohio River Aquifer show a relatively flat trend in serum PFOA in the 1990s, before decreasing in the mid-to-late 2000s. This suggests that the natural filtration of the aquifer reduced drinking water concentrations enough that residents reached steady-state serum concentrations, but that overall exposures were higher than the US general population, and that an aquifer's natural filtration is less effective at removing PFOA than other treatment technologies. The most striking difference in temporal trend is between Greater Cincinnati and Northern Kentucky. Because both utilities use the Ohio River as a water source at the same point along the river, though Northern Kentucky also draws on the Licking River, we believe drinking water treatment explains this difference. Cincinnati began using GAC in 1992 (Westerhoff et al., 2009), while Northern Kentucky did not until June 2012, after all samples from Northern Kentucky residents were collected. GAC treatment may explain why Cincinnati participants' serum PFOA concentration decreased during the 1990s, while those in Northern Kentucky residents rose. GAC use may also explain the differences in serum PFOA between Huntington, WV and Portsmouth, OH; Huntington, WV began using GAC in 1981 (Culp et al., 1981), while Portsmouth, OH does not.

Overall, GAC filtration or aquifer recharge decreased adjusted mean serum PFOA concentration 4 ng/mL (Model 1), while serum PFOA increases per year were 0.19–0.21 ng/mL lower than they would have been absent GAC (Model 2). These reductions were

40–60% of estimated serum PFOA concentrations. C-8 Project participants had similar reductions in their serum PFOA concentrations after GAC installation (Bartell et al., 2010). We also saw reductions in serum concentrations of other PFAS with GAC treatment; however, only PFOS saw a reduction in serum concentrations from GAC use (0.16 ng/mL) comparable to those observed with PFOA. This may be a result of lower sourcewater concentrations of other PFAS and not less efficacy for GAC. While GAC filtration did not reduce PFOA exposure to background level, it did mitigate PFOA exposure through drinking water.

Pharmacokinetic analyses under model criteria with the best fit for the entire population (Scenario 2 and PFOA elimination coefficients based on a 3.8yr half-life) indicate that GM serum PFOA concentrations in participants with multiple serum samples increased from 1991 to 2001, then declined to 2013. Under conditions where estimated drinking water PFOA concentrations were increasing, we saw very little variation in GM serum PFOA across all the half-lives we investigated. For both low initial PFOA exposure and the BCERP cohort, Scenario 1 (constant PFOA water concentrations) was a better fit than time-varying PFOA water concentrations (Scenario 2). Both the BCERP cohort and participants with lower starting PFOA had samples collected later than those in the FCC cohort or higher initial PFOA; therefore, Scenario 1 being a better fit may indicate that PFOA concentrations decreased rapidly between 2005 and 2009, or did not decrease as rapidly as we estimated after 2009. A 3.8-year half-life being the best fit in our pharmacokinetic analyses appears to contradict recent studies, which found the PFOA half-life to be 1.5–2.1 years shorter (Fu et al., 2016; Bartell et al., 2010). However, half-life studies where participants are still exposed to PFOA may result in a higher apparent half-life if all sources of PFOA are not properly accounted for (Russell et al., 2015). Because we did not have individual estimates of PFOA exposure through non-water sources, and used present drinking water consumption for past intake where necessary, the longer half-life may be accounting for errors in dose estimation.

Serum PFOA was significantly associated with water source, year sample collected, age at sample collection, tap water consumption, pregnancy, gravidity and breastfeeding in regression models. Although males have higher median serum PFOA concentrations than females in the general population (2.3 ng/mL vs. 1.8 ng/mL in 2011–2012) (CDC, 2015) and sex was highly significant in our univariate models, sex dropped out when models included pregnancy, reproductive history and breastfeeding. Maternal PFOA decreases associated with pregnancy, gravidity and breastfeeding may explain some of the observed sex differences (Liu et al., 2011). While not assessed in our study, menstruation may account for some of the remaining difference in serum PFOA concentrations between males and females (Lorber et al., 2015).

Significant associations between serum PFOA and tap water consumption, the Ohio River and Ohio River Aquifer strongly suggest drinking water is a predominant PFOA exposure source in these study populations. After covariate-adjustment, each additional liter of tap water consumed daily increased serum PFOA 0.17 ng/mL. Each additional year drinking Ohio River water treated with GAC increased serum PFOA 0.13 ng/mL, while each additional year drinking Ohio River water without GAC increased serum PFOA 0.34 ng/mL; one additional year drinking Ohio River Aquifer water increased serum PFOA 0.15 ng/mL. These associations point to drinking water as a major exposure source for our study participants.

Inter-PFAS correlations also suggest that drinking water is a major PFOA exposure source. Such correlations are strong in the general population (e.g., NHANES; PFOA-PFOS $\rho = 0.66$) (Calafat et al., 2007), and where the primary PFAS source is food (PFOA-PFOS $\rho = 0.54$) (Ji et al., 2012). Conversely, correlations are weaker

for populations exposed to PFOA through drinking water: PFOA-PFOS $\rho = 0.30$ in C-8 study participants (Frisbee et al., 2009), and 0.11–0.32 in Arnsberg, Germany (Holzer et al., 2008). Inter-PFAS correlations in our study population, especially PFOA-PFOS ($\rho = 0.41$), were lower than those previously reported for BCERP participants in Greater Cincinnati ($\rho = 0.62$) or the San Francisco Bay Area ($\rho = 0.60$), but comparable to BCERP participants living in Northern Kentucky ($\rho = 0.38$) (Pinney et al., 2014). Inter-PFAS correlations are also comparable to those measured in the C-8 project community (Frisbee et al., 2009). While PFAS have multiple environmental sources, these weaker correlations suggest different PFOA exposure source(s) predominate in our population than the US general population.

Isomer profiles can suggest exposure sources (De Silva and Mabury, 2006). PFAS were predominantly produced using electrochemical fluorination (ECF) in the US until 2002, when 3M ceased production. Telomerization was used by the largest remaining US PFAS manufacturer, DuPont (Prevedouros et al., 2006). ECF produces 70%–80% linear and 20–30% branched PFOS and PFOA; telomerization produces linear products from linear feedstocks (Buck et al., 2011). Human PFOA isomer pharmacokinetics are unknown, but rodents eliminate branched PFOA faster than *n*-PFOA (De Silva et al., 2009). Taken together, these findings suggest our participants were predominantly exposed to telomerization-produced PFOA (De Silva and Mabury, 2006). However we cannot rule out participant exposure to ECF-produced PFOA; mean 97% *n*-PFOA might have resulted, at least in part, from preferential branched PFOA isomer excretion, and *n*-PFOA retention, before sera collection.

The trends in study participants' measured and predicted serum PFOA mirror measured Ohio River water PFOA concentrations. PFAS concentrations were measured throughout the Ohio River Valley in September 2009 (Emery et al., 2010). Additional Ohio River PFOA measurements were conducted near Parkersburg, WV in 2003 (GIST, 2003) and average Ohio River PFOA concentrations were estimated for 1951–2003 (Paustenbach et al., 2007). Ohio River water PFBS, PFHxS, PFOS, PFNA and PFDeA concentrations in 2009 were less than 6 ng/L between Huntington, WV and Louisville, KY. Reported Cincinnati Ohio River water PFOA concentrations were 100 ng/L in 2005, 21 ng/L in 2006, and 13.1 ng/L in 2009 (Personal communication with Greater Cincinnati Water Works; Emery et al., 2010). 2009 Ohio River water PFOA concentrations at Huntington were 19.1 ng/L and 3.4–5.3 ng/L at points above Parkersburg, WV, 209 km upstream of Huntington; 2009 water PFOA concentrations were also low (1.48–6.22 ng/L) in Ohio River tributaries in our study area. Measured Ohio River PFOA concentrations at Parkersburg were 35 ng/L in 2009 (Emery et al., 2010) and 295 ng/L in 2003 (GIST, 2003); average estimated annual PFOA concentration was 690 ng/L in 1999 (Paustenbach et al., 2007). Measured PFOA concentrations in Ohio River Aquifer water 2002–2005 near Parkersburg were 58–4800 ng/L. The Ohio River Aquifer is partially recharged by the Ohio River, contaminating it with PFOA (Paustenbach et al., 2007).

Three known industrial sources discharge PFOA into the Ohio River: DuPont's Fort Washington Works and on-site landfill, Dry Run Landfill in Washington, WV, and Letart Landfill in Letart, WV. DuPont reported industrial discharges of approximately 150000 lbs. of PFOA (as ammonium perfluorooctanoate) to the Ohio River in the 1980s, 350000 lbs. in the 1990s and 75000 lbs. 2000–2003 (Paustenbach et al., 2007). The 2004 annual releases were 6000 lbs./yr., decreasing to 705 lbs./yr. in 2011 (United States Environmental Protection Agency, 2015b). These discharges contaminated municipal drinking water (Emmett et al., 2006; Paustenbach et al., 2007) and private wells (Hoffman et al., 2011) in the C-8 project area. Industrial discharges to Letart and Dry Run

are unknown, but surface water PFOA concentrations were 107–40290 ng/L for Dry Run and 154–1960 ng/L for Letart (GIST, 2003). Other industrial facilities discharging PFOA into the Ohio River are unknown. Wastewater treatment plants (WWTPs) (Huset et al., 2008; Muller et al., 2011a) or non-point sources (Muller et al., 2011b) are possible PFOA sources. Ohio River water PFOA concentrations in 2009 increased 1–3 ng/L at WWTP outfalls, compared to measurements immediately upstream (Emery et al., 2010). However, water PFOA concentrations are much lower (1.4–7.4 ng/L) in rivers solely contaminated by WWTPs (Huset et al., 2008; Muller et al., 2011a), while industrial PFAS discharges predominate over WWTPs in waters contaminated by both (Castiglioni et al., 2015; Valsecchi et al., 2015; Zhu et al., 2015). Because PFOA contamination in other waters is reported to persist hundreds of kilometers downstream of PFAS manufacturers (Valsecchi et al., 2015; Zhu et al., 2015), PFOA discharged into the Ohio River 209–666 km upstream of our study area could have contaminated our participants' drinking water.

4.1. Strengths/limitations

All three study populations represented voluntary convenience samples, and therefore were not necessarily representative of the general population. Although the mean and median values of PFCS reported here do not necessarily represent general population values, our data certainly indicate that there was widespread exposure to PFOA among participants. We could not assess all exposure sources, including food, house dust or whether adult participants were breastfed as children. Complete occupational histories were unavailable, though no participants worked at PFAS-production facilities. Because we based Model 2 on residential addresses, not including occupational histories could result in overestimating water source exposure, especially in large urban areas with multiple water providers or water sources. This is mitigated for children, where residences and schools are more closely located. However, our regression models explained 44–47% of study participants' serum PFOA variation without including these other exposure sources or locations. Because other PFAS concentrations in our study population were similar to NHANES, it is unlikely that high PFOA serum concentrations in our study are mainly due to food and dust exposures. Participants may have improperly estimated average water intake; this was mitigated by using visual aids for serving sizes (Chambers et al., 2000). We assumed constant reported water consumption in analyses; this could overestimate water consumption when participants were children (Kant and Graubard, 2010), and underestimate consumption when participants were younger adults (Kant et al., 2009). One-compartment pharmacokinetic models may not properly estimate serum PFOA concentrations, though trends matched regression analyses. Rigorous collection and processing protocols strengthen this study. Serum was specifically collected, processed, and stored for bio-specimen analysis using research protocols developed by CDC. Anthropometrics were collected using similar protocols for all studies, data were collected prior to PFAS measurements, and questionnaires were repeatedly administered to BCERP and FCC cohorts, allowing for missing data collection and response validation.

5. Conclusions

Mid-Ohio River Valley residents (Huntington, WV to Louisville, KY) were likely exposed to PFOA, but not other PFAS, above US population background levels; serum PFOA concentrations were highest in the 1990s, declining in the 2000s. Serum PFOA concentrations are significantly associated with use of Ohio River and Ohio

River Aquifer water. While using GAC for water treatment resulted in lower serum PFOA, it did not reduce concentrations to background. Municipal drinking water, probably contaminated by industrial PFOA discharges 209–666 km upstream of our participants, appears to be the primary PFOA exposure source.

Funding sources

This publication was made possible by the Breast Cancer and the Environment Research Program awards U01ES012770 and U01ES019453 from the National Institute of Environmental Health Sciences and the National Cancer Institute; P30-ES006096, R21 ES017176 and T32-ES10957 from NIEHS; EPA-RD-83478801 from the United States Environmental Protection Agency, and CSTA-UL1RR026314 from the National Center for Research Resources. Funding sources had no involvement in study design beyond the initial suggestion that we measure serum biomarkers of PFC exposure, and no involvement in collection, analysis and interpretation of data, writing of the article and the decision to submit the article for publication. Its contents are solely the responsibility of the authors and do not necessarily represent the official views of the NIEHS, NCI, the National Institutes of Health or the Centers for Disease Control and Prevention.

Competing financial interests

None.

Acknowledgements

Dr. Renee Jeffreys-Heil, Valeriya Petrenko, Emily Jones and Kathy Kreitel assisted with interviews. Victoria Straughn, Rhoshonda Jones and Sandy Rota performed blood collection, storage and processing. Kayoko Kato, Charles Dodson, Zsuzsanna Kuklenyik, Xavier Bryant, Amal Wanigatunga, Brian Basden, Carmen Dunbar, Ayesha Patel, Jun Ma, Tao Jia and Jack Reidy at CDC conducted PFAS measurements. West Virginia, Ohio, Kentucky and Indiana utilities, regulatory and mapping agencies helped identify water sources. The late Dr. Paul Succop assisted with statistical analyses. Dr. Carol Rice provided editorial review.

Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envpol.2017.04.092>.

References

- Bartell, S.M., Calafat, A.M., Lyu, C., Kato, K., Ryan, P.B., Steenland, K., 2010. Rate of decline in serum PFOA concentrations after granular activated carbon filtration at two public water systems in Ohio and West Virginia. *Environ. Health Perspect.* 118, 222–228.
- Belisle, J., 1981. Organic fluorine in human serum: Natural versus industrial sources. *Science* 212, 1509–1510.
- Biro, F.M., Galvez, M.P., Greenspan, L.C., Succop, P.A., Vangeepuram, N., Pinney, S.M., Teitelbaum, S., Windham, G.C., Kushi, L.H., Wolff, M.S., 2010. Pubertal assessment method and baseline characteristics in a mixed longitudinal study of girls. *Pediatrics* 126, e583–e590. <http://dx.doi.org/10.1542/peds.2009-3079>.
- Braun, J.M., Chen, A., Romano, M.E., Calafat, A.M., Webster, G.M., Yoltson, K., Lanphear, B.P., 2016. Prenatal perfluoroalkyl substance exposure and child adiposity at 8 years of age: The HOME study. *Obes. (Silver Spring)* 24, 231–237. <http://dx.doi.org/10.1002/oby.21258>.
- Brede, E., Wilhelm, M., Goen, T., Muller, J., Rauchfuss, K., Kraft, M., Holzer, J., 2010. Two-year follow-up biomonitoring pilot study of residents' and controls' PFC plasma levels after PFOA reduction in public water system in Arnsberg, Germany. *Int. J. Hyg. Environ. Health* 213, 217–223.
- Buck, R.C., Franklin, J., Berger, U., Conder, J.M., Cousins, I.T., de Voogt, P., Jensen, A.A., Kannan, K., Mabury, S.A., van Leeuwen, S.P., 2011. Perfluoroalkyl and polyfluoroalkyl substances in the environment: terminology, classification, and origins. *Integr. Environ. Assess. Manag.* 7, 513–541. <http://dx.doi.org/10.1002/ieam.258>.
- Calafat, A.M., Kuklenyik, Z., Reidy, J.A., Caudill, S.P., Tully, J.S., Needham, L.L., 2007. Serum concentrations of 11 polyfluoroalkyl compounds in the U.S. population: data from the national health and nutrition examination survey (NHANES). *Environ. Sci. Technol.* 41, 2237–2242.
- Castiglioni, S., Valsecchi, S., Polesello, S., Rusconi, M., Melis, M., Palmiotto, M., Manenti, A., Davoli, E., Zuccato, E., 2015. Sources and fate of perfluorinated compounds in the aqueous environment and in drinking water of a highly urbanized and industrialized area in Italy. *J. Hazard. Mater.* 282, 51–60. <http://dx.doi.org/10.1016/j.jhazmat.2014.06.007>.
- Centers for Disease Control and Prevention, 2015. Fourth Report on Human Exposure to Environmental Chemicals. Updated Tables, (February, 2015). Centers for Disease Control and Prevention, Atlanta, GA.
- Chambers 4th, E., Godwin, S.L., Vecchio, F.A., 2000. Cognitive strategies for reporting portion sizes using dietary recall procedures. *J. Am. Diet. Assoc.* 100, 891–897.
- Culp, R.L., Faisst, J.A., Smith, C.E., 1981. Granular Activated Carbon Installations. United States Environmental Protection Agency, Office of Research and Development, Municipal Environmental Research Laboratory, Cincinnati, OH.
- De Silva, A.O., Benskin, J.P., Martin, L.J., Arsenault, G., McCrindle, R., Riddell, N., Martin, J.W., Mabury, S.A., 2009. Disposition of perfluorinated acid isomers in Sprague-Dawley rats; part 2: Subchronic dose. *Environ. Toxicol. Chem.* 28, 555–567. <http://dx.doi.org/10.1897/08-254.1>.
- De Silva, A.O., Mabury, S.A., 2006. Isomer distribution of perfluorocarboxylates in human blood: Potential correlation to source. *Environ. Sci. Technol.* 40, 2903–2909.
- D'Hollander, W., de Voogt, P., De Coen, W., Bervoets, L., 2010. Perfluorinated substances in human food and other sources of human exposure. *Rev. Environ. Contam. Toxicol.* 208, 179–215. http://dx.doi.org/10.1007/978-1-4419-6880-7_4.
- Emery, E., Spaeth, J., Mills, M., Nakayama, S., Frommel, J., 2010. A Screening Study Investigating the Presence of Emerging Contaminants within the Ohio River Basin. The Ohio River Valley Water Sanitation Commission (ORSANCO) and United States Environmental Protection Agency, Office of Research and Development, National Risk Management Research Laboratory, Cincinnati, OH.
- Emmett, E.A., Shofer, F.S., Zhang, H., Freeman, D., Desai, C., Shaw, L.M., 2006. Community exposure to perfluorooctanoate: Relationships between serum concentrations and exposure sources. *J. Occup. Environ. Med.* 48, 759–770. <http://dx.doi.org/10.1097/01.jom.0000232486.07658.74>.
- Ericson, I., Nadal, M., van Bavel, B., Lindstrom, G., Domingo, J.L., 2008. Levels of perfluorochemicals in water samples from Catalonia, Spain: Is drinking water a significant contribution to human exposure? *Environ. Sci. Pollut. Res. Int.* 15, 614–619. <http://dx.doi.org/10.1007/s11356-008-0040-1>.
- Fernald Community Cohort (FCC) website. <http://med.uc.edu/eh/research/projects/fcc>. (Last accessed 4 April 2017).
- Frisbee, S.J., Brooks Jr., A.P., Maher, A., Flensburg, P., Arnold, S., Fletcher, T., Steenland, K., Shankar, A., Knox, S.S., Pollard, C., Halverson, J.A., Vieira, V.M., Jin, C., Leyden, K.M., Ducatman, A.M., 2009. The C8 health project: Design, methods, and participants. *Environ. Health Perspect.* 117, 1873–1882. <http://dx.doi.org/10.1289/ehp.0800379>.
- Fu, J., Gao, Y., Cui, L., Wang, T., Liang, Y., Qu, G., Yuan, B., Wang, Y., Zhang, A., Jiang, G., 2016. Occurrence, temporal trends, and half-lives of perfluoroalkyl acids (PFAAs) in occupational workers in China. *Sci. Rep.* 6, 38039.
- Groundwater Investigation Steering Team (GIST), 2003. Ammonium Perfluorooctanoate (C-8) Groundwater Investigation Steering Team Report. Groundwater Program. Division of Water and Waste Management, Charleston, WV.
- Guy, W.S., Taves, D.R., Brey Jr., W.S., 1976. Organic fluorocompounds in human plasma: Prevalence and characterization. *ACS Symp. Ser.* 28.
- Hoffman, K., Webster, T.F., Bartell, S.M., Weisskopf, M.G., Fletcher, T., Vieira, V.M., 2011. Private drinking water wells as a source of exposure to perfluorooctanoic acid (PFOA) in communities surrounding a fluoropolymer production facility. *Environ. Health Perspect.* 119, 92–97. <http://dx.doi.org/10.1289/ehp.1002503>.
- Holzer, J., Midasch, O., Rauchfuss, K., Kraft, M., Reupert, R., Angerer, J., Kleeschulte, P., Marschall, N., Wilhelm, M., 2008. Biomonitoring of perfluorinated compounds in children and adults exposed to perfluorooctanoate-contaminated drinking water. *Environ. Health Perspect.* 116, 651–657. <http://dx.doi.org/10.1289/ehp.11064>.
- Hornung, R.W., Reed, L.D., 1990. Estimation of average concentration in the presence of nondetectable values. *Appl. Occup. Environ. Hyg.* 5, 46–51.
- Huset, C.A., Chiaia, A.C., Barofsky, D.F., Jonkers, N., Kohler, H.P., Ort, C., Giger, D.W., Field, J.A., 2008. Occurrence and mass flows of fluorochemicals in the Glatt Valley watershed. *Switz. Environ. Sci. Technol.* 42, 6369–6377.
- Ji, K., Kim, S., Kho, Y., Paek, D., Sakong, J., Ha, J., Kim, S., Choi, K., 2012. Serum concentrations of major perfluorinated compounds among the general population in Korea: Dietary sources and potential impact on thyroid hormones. *Environ. Int.* 45, 78–85. <http://dx.doi.org/10.1016/j.envint.2012.03.007>.
- Kant, A.K., Graubard, B.I., 2010. Contributors of water intake in US children and adolescents: Associations with dietary and meal characteristics—National Health and Nutrition Examination Survey 2005–2006. *Am. J. Clin. Nutr.* 92, 887–896. <http://dx.doi.org/10.3945/ajcn.2010.29708>.
- Kant, A.K., Graubard, B.I., Atchison, E.A., 2009. Intakes of plain water, moisture in foods and beverages, and total water in the adult US population—nutritional, meal pattern, and body weight correlates: National Health and Nutrition Examination Surveys 1999–2006. *Am. J. Clin. Nutr.* 90, 655–663. <http://dx.doi.org/10.3945/ajcn.2009.27749>.

- Kato, K., Wong, L.Y., Chen, A., Dunbar, C., Webster, G.M., Lanphear, B.P., Calafat, A.M., 2014. Changes in serum concentrations of maternal poly- and perfluoroalkyl substances over the course of pregnancy and predictors of exposure in a multiethnic cohort of Cincinnati, Ohio pregnant women during 2003–2006. *Environ. Sci. Technol.* 48, 9600–9608. <http://dx.doi.org/10.1021/es501811k>.
- Kato, K., Basden, B.J., Needham, L.L., Calafat, A.M., 2011. Improved selectivity for the analysis of maternal serum and cord serum for polyfluoroalkyl chemicals. *J. Chromatogr. A* 1218, 2133–2137. <http://dx.doi.org/10.1016/j.chroma.2010.10.051>.
- Kuklennyik, Z., Needham, L.L., Calafat, A.M., 2005. Measurement of 18 perfluorinated organic acids and amides in human serum using on-line solid-phase extraction. *Anal. Chem.* 77, 6085–6091.
- Lau, C., Anitole, K., Hodes, C., Lai, D., Pfahles-Hutchens, A., Seed, J., 2007. Perfluoroalkyl acids: A review of monitoring and toxicological findings. *Toxicol. Sci.* 99, 366–394 doi: kfm128 [pii].
- Liu, J., Li, J., Liu, Y., Chan, H.M., Zhao, Y., Cai, Z., Wu, Y., 2011. Comparison on gestation and lactation exposure of perfluorinated compounds for newborns. *Environ. Int.* 37, 1206–1212. <http://dx.doi.org/10.1016/j.envint.2011.05.001>.
- Lorber, M., Eaglesham, G.E., Hobson, P., Toms, L.M., Mueller, J.F., Thompson, J.S., 2015. The effect of ongoing blood loss on human serum concentrations of perfluorinated acids. *Chemosphere* 118, 170–177.
- Muller, C.E., Gerecke, A.C., Alder, A.C., Scheringer, M., Hungerbuhler, K., 2011a. Identification of perfluoroalkyl acid sources in Swiss surface waters with the help of the artificial sweetener acesulfame. *Environ. Pollut.* 159, 1419–1426. <http://dx.doi.org/10.1016/j.envpol.2010.12.035>.
- Muller, C.E., Spiess, N., Gerecke, A.C., Scheringer, M., Hungerbuhler, K., 2011b. Quantifying diffuse and point inputs of perfluoroalkyl acids in a nonindustrial river catchment. *Environ. Sci. Technol.* 45, 9901–9909. <http://dx.doi.org/10.1021/es202140e>.
- Newton, S., McMahan, R., Stoeckel, J.A., Chislock, M., Lindstrom, A., Strynar, M., 2017. Novel polyfluorinated compounds identified using high resolution mass spectrometry downstream of manufacturing facilities near Decatur, Alabama. *Environ. Sci. Technol.* 51, 1544–1552.
- Olsen, G.W., Burris, J.M., Ehresman, D.J., Froehlich, J.W., Seacat, A.M., Butenhoff, J.L., Zobel, L.R., 2007. Half-life of serum elimination of perfluorooctanesulfonate, perfluorohexanesulfonate, and perfluorooctanoate in retired fluorochemical production workers. *Environ. Health Perspect.* 115, 1298–1305.
- Paustenbach, D.J., Panko, J.M., Scott, P.K., Unice, K.M., 2007. A methodology for estimating human exposure to perfluorooctanoic acid (PFOA): A retrospective exposure assessment of a community (1951–2003). *J. Toxicol. Environ. Health A* 70, 28–57 doi: V8124674W361L307 [pii].
- Pinney, S.M., Biro, F.M., Windham, G.C., Herrick, R.L., Yaghjian, L., Calafat, A.M., Succop, P., Sucharew, H., Ball, K.M., Kato, K., Kushi, L.H., Bornschein, R., 2014. Serum biomarkers of polyfluoroalkyl compound exposure in young girls in greater Cincinnati and the San Francisco Bay area, USA. *Environ. Pollut.* 184, 327–334. <http://dx.doi.org/10.1016/j.envpol.2013.09.008>.
- Prevedouros, K., Cousins, I.T., Buck, R.C., Korzeniowski, S.H., 2006. Sources, fate and transport of perfluorocarboxylates. *Environ. Sci. Technol.* 40, 32–44.
- Rahman, M.F., Peldszus, S., Anderson, W.B., 2014. Behaviour and fate of perfluoroalkyl and polyfluoroalkyl substances (PFASs) in drinking water treatment: A review. *Water Res.* 50, 318–340. <http://dx.doi.org/10.1016/j.watres.2013.10.045>.
- Russell, M.H., Waterland, R.L., Wong, F., 2015. Calculation of chemical elimination half-life from blood with an ongoing exposure source: The example of perfluorooctanoic acid (PFOA). *Chemosphere* 129, 210–216.
- Taves, D.R., Grey, W.S., Brey Jr., W.S., 1976. Organic fluoride in human plasma: Its distribution and partial identification. *Toxicol. Appl. Pharmacol.* 37, 120.
- Thompson, J., Lorber, M., Toms, L.M., Kato, K., Calafat, A.M., Mueller, J.F., 2010. Use of simple pharmacokinetic modeling to characterize exposure of Australians to perfluorooctanoic acid and perfluorooctane sulfonic acid. *Environ. Int.* 36, 390–397. <http://dx.doi.org/10.1016/j.envint.2010.02.008>.
- United States Environmental Protection Agency, 2015a. 2010/2015 PFOA Stewardship Program.
- United States Environmental Protection Agency, 2015b. ICIS Detailed Reports: WASHINGTON WORKS, NPDES WV0001279.
- Valsecchi, S., Rusconi, M., Mazzoni, M., Viviano, G., Pagnotta, R., Zaghi, C., Serrini, G., Polesello, S., 2015. Occurrence and sources of perfluoroalkyl acids in Italian river basins. *Chemosphere* 129, 126–134. <http://dx.doi.org/10.1016/j.chemosphere.2014.07.044>.
- Wang, Z., Cousins, I.T., Scheringer, M., Buck, R.C., Hungerbuhler, K., 2014. Global emission inventories for C4–C14 perfluoroalkyl carboxylic acid (PFCA) homologues from 1951 to 2030, Part I: Production and emissions from quantifiable sources. *Environ. Int.* 70, 62–75. <http://dx.doi.org/10.1016/j.envint.2014.04.013>.
- Westerhoff, G., Atha, K., Pohlman, R., 2009. The Cincinnati GAC Experience: Improving Water Quality and Public Trust in Ohio. *Government Engineering*, pp. 30–32. March–April.
- Wones, R., Pinney, S., Buckholz, J., Deck-Tebbe, C., Freyberg, R., Pesce, A., 2009. Medical monitoring: A beneficial remedy for residents living near an environmental hazard site. *J. Occup. Environ. Med.* 51, 1374–1374–1383.
- Zhu, Z., Wang, T., Meng, J., Wang, P., Li, Q., Lu, Y., 2015. Perfluoroalkyl substances in the Daling River with concentrated fluorine industries in China: Seasonal variation, mass flow, and risk assessment. *Environ. Sci. Pollut. Res. Int.* 22, 10009–10018. <http://dx.doi.org/10.1007/s11356-015-4189-0>.