Perfluorinated compounds (PFCs) in fish: What we learned from EPA probability-based surveys of U.S. urban rivers and the Great Lakes

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Are the fish safe to eat?
The National Study of Chemical Residues in Lake Fish Tissue

National Rivers & Streams Assessment
Assessing the Nation’s Waters

National Coastal Condition Assessment
Target Chemicals?

**NLFTS – 268 chemicals**
- metals (Hg and As)
- dioxins/furans
- PCB congeners
- pesticides
- semi-volatile organics (e.g., PAHs)

**CECs**
- Pharmaceuticals/personal care products
- Polybrominated diphenyl ethers (PBDEs)
- Perfluorinated compounds (PFCs)
What are perfluorinated compounds?

- A family of fluorine-containing synthetic compounds
- Chemical structure gives them unique properties
- Thermal stability and repel both water and oil
- Qualities capable of making consumer/industrial materials stain and stick resistant

Many forms of PFCs, but two familiar compounds are:

• PFOA or perfluorooctanoic acid, used to make Teflon® products, and
• PFOS or perfluorooctane sulfonate, a breakdown product of chemicals formerly used to make Scotchgard® products
Large volumes of PFCs have been produced since the 1950s. Their high production volume led to widespread distribution in the environment, particularly in water where they’re readily transported.

- Grease-resistant food packaging and paper products
- Stain protection treatment on carpet, furniture, and clothing (until 2002)
- Coatings on non-stick cookware
- Even waterproof clothing
Why are PFCs CECs?

In the environment…

- Released during manufacture, storage/transport, product use, and disposal
- Global environmental distribution noted in early 2000s
- Widely distributed in water, air, food, wildlife, humans
- High levels found in waters near PFC facilities, WWTPs, urban centers, and in urban groundwater
- Detected in bird and mammal species in remote Arctic
- Bioaccumulate through food webs (highest potential for longer-chain PFCs)
In birds, fish, and mammals, PFCs bind to proteins in blood/livers (not lipophilic like other organic pollutants).

Animal studies on toxic effects of PFCs (primarily PFOS and PFOA) show various effects on development, reproduction, and immune function.

PFOA increased the risk of tumors of liver, testicles, mammary glands, and pancreas in lab animals.

It isn’t clear that these chemical causes of cancers in animals would also translate to humans.
Why are PFCs CECs?

In humans...

• Occupational and non-occupational exposures (e.g., drinking water, carpet/upholstery dust, food packaging, non-stick cookware)
• Long half-life (4-8 years)
• 2005–2013 epidemiological study found probable links between elevated PFOA levels in human blood and ulcerative colitis, thyroid diseases, testicular cancer, kidney cancer, and preeclampsia
• Possible human health concerns remain due to continued use of PFOS, PFOA, and newer PFCs being introduced as substitutes
EPA worked to voluntarily phase out PFOS production (and related chemicals) in 2002.

EPA established a PFOA Stewardship Program with eight major companies toward a 95% reduction in emissions and product contents of PFOA (and long-chain PFCs) by 2010 and elimination by 2015.

Despite restrictions, PFCs are still produced in other parts of the world and enter the U.S. with imports.

PFC precursors produced in the U.S. can break down to persistent degradation products that will continue to be CECs.
Consumption of fish from contaminated waters may be a major source of exposure to PFOS or long-chain PFCs.

Sinclair et al. (2006) found average PFOS concentrations in fish to be 8,850-fold greater than levels in surface waters of New York State.

Why study PFCs in fish?

- Uncertainty associated with the role of food as an exposure pathway
- Recent modeling studies estimate that PFC contamination in food may account for more than 90% of human exposure to PFOS and PFOA.
How can we characterize PFC contamination in U.S. freshwater fish?

EPA conducted screening level studies of PFCs in freshwater fish on a national scale during the 2008-09 NRSA and in the Great Lakes during the 2010 NCCA.

162 urban river sites (5th order or greater in size)

157 randomly selected nearshore sites throughout the five Great Lakes

...allowing the first nationally and regionally representative assessment of PFCs using a statistically based sampling design.
Statistical design

Sampling sites selected using a probability-based approach (Stevens and Olsen, 1999) described as a Generalized Random Tessellation Stratified (GRTS) survey design by Stevens and Olsen (2004).

To implement the design, the population being sampled had to be explicitly described, every element in the population had the opportunity to be sampled with known probability, and the selection process included an explicit random element.
Statistical analysis

Incorporates elements of the probabilistic survey design and includes:

- survey design (sample) weights adjustment based on site status
- target population estimation (i.e., number of sites that met the study definition of urban river or nearshore Great Lakes location)
- estimation of the number/proportion of sites in the sampled population
- estimation of percentiles and cumulative distribution of tissue concentrations for the sampled population

Statistical results provide a regionally representative sample that can be extrapolated to a Great Lakes nearshore surface area of an estimated 11,091 km² (4,282 mi²)

Statistical results provide a nationally representative sample that can be extrapolated to a sampled population of an estimated river length of 17,509 km (10,880 miles)
Sample collection, preparation, and analysis

- **Target fish** -- ubiquitous, abundant, easily identified, commonly consumed by humans, and large enough to provide adequate tissue for analysis
- **Sampling** -- electrofishing May-September 2008/09 for the NRSA; gillnetting and hook and line June-November 2010 for the GL
- **Preparation** -- fish scaled and filleted in the lab and fillets compositied using the batch method
- **Analysis** -- fillet homogenates analyzed by high-performance liquid chromatography tandem mass spectrometry (HPLC-MS/MS) for 13 PFCs
Results

- **162** composite samples (682 fish)
- **80%** of the urban river samples contained some detectable PFCs
- **PFOS** was the most frequently detected chemical (in **73%** of samples)
- The statistically derived PFOS **median** in fillets was **10.7 ppb** for the urban river sampled population of 17,509 km (10,880 mi)
- The **maximum** PFOS concentration measured in fillet tissue was **127 ppb**

- **157** composite samples (423 fish)
- **100%** of the Great Lakes samples contained some detectable PFCs
- **PFOS** was the most frequently detected chemical (in **100%** of samples)
- The statistically derived PFOS **median** in fillets was **15.2 ppb** for the Great Lakes nearshore sampled population of 11,091 km$^2$ (4,282 mi$^2$)
- The **maximum** PFOS concentration measured in fillet tissue was **80 ppb**
Results

- PFOS appears to have the highest bioaccumulation potential in food webs (Houde et al. 2011)
- Highest PFOS values in urban rivers and Great Lakes in top carnivores
Human health screening values

- EPA proposed draft HH reference doses for PFOA and PFOS that are currently under review (Federal Register, 2014)
- National risk-based consumption limits or HH SVs have not been developed for PFCs in the U.S.
- Minnesota Department of Health included meal advice categories based on PFOS in fish for their statewide Fish Consumption Advisory recommending:
  - consumption of only one meal of fish per week if PFOS concentrations are >40–200 ng/g (wet weight),
  - one meal per month if levels are >200–800 ng/g,
  - and no consumption at levels greater than 800 ng/g
Cumulative Distribution Function (PFOS)

- No samples were at “do not eat” levels
- **11%** of the sampled population of U.S. urban rivers (1,856 river km) and **9%** of the Great Lakes sampled population (998 km²) had PFOS concentrations that exceeded the 40 ng/g SV recommending that fish consumption be restricted to no more than one meal per week.
Is continued PFC monitoring warranted?

- Temporal trend models do not show a decline in Lake Ontario lake trout PFCs despite the 2002 voluntary PFOS (and related chemical) production phase out (Gewurtz et al. 2012)
- Their power analysis results suggested that 15 years of PFOS data (and a within-year sample size of at least 10) would be required to obtain sufficient power (80%) to detect a 5% decreasing trend in PFOS contamination, which demonstrates the need for continued long-term monitoring

- Continued development and refinement of tissue advisory levels is warranted as knowledge increases on the toxicity, prevalence, sources, and environmental behavior of PFCs and the overall benefits of fish consumption
For more information? 

For a copy of the journal article contact: Alena Baldwin-Brown

For raw data summarized in this presentation go to: 
http://www.epa.gov/fish-tech/fish-tissue-data-collected-epa

For more information contact Leanne Stahl at: stahl.leanne@epa.gov
Thank you!