Understanding Diverse Exposure Pathways for PFAS
Elsie M. Sunderland (ems@seas.harvard.edu)
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Human exposures to PFAS are diverse: Some can be addressed/mitigated faster than others.

Sunderland et al., 2019, JESEE

Our focus:
1. Drinking water
2. Seafood
3. Consumer Products

*TIMESCALES*
Detection of PFAS in U.S. drinking water statistically increased with higher point source abundance

PFAS composition provides insights into major sources

Zhang et al., *ES&T Letters*, 2016
Drinking water is the primary pathway of PFAS exposure next to many contaminated sites

Figure adapted from data compiled by Northeastern University’s Social Science Environmental Health Research Institute (SSEHRI)
Drinking water may **not** be the major exposure source for the general population outside of contaminated sites.

**What are the sources of PFAS occurrence in drinking water?**

**How important is drinking water to the overall human exposure to PFAS?**

**What about exposure to alternative PFASs?**
Nurses Health Study, HSPH, a large prospective study of US women est. 1976

1976
Enrollment
\( n = 121,319 \)

1989/1990
Serum and tap water collection
\( n = 32,826 \)

Dietary
Life style
Residential location

What are exposures now?
Relative source contribution (RSC): What is this? Why does it matter?

Exposure assessment
- Water ingestion rates
- Proportion of the daily dose supplied by drinking water relative to other exposure sources (RSC)

<table>
<thead>
<tr>
<th>Entity</th>
<th>RSC</th>
<th>Source</th>
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<tbody>
<tr>
<td>Federal EPA, most states</td>
<td>20% for all PFAS</td>
<td>EPA grey literature</td>
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<tr>
<td>Minnesota, Maine, New Hampshire</td>
<td>20% - 60% for PFOA and PFOS</td>
<td>Human biomonitoring studies</td>
</tr>
<tr>
<td>Alaska, Texas</td>
<td>100%</td>
<td>Developed for remediation and clean-up of contaminated sites</td>
</tr>
<tr>
<td>Our study (NHS)</td>
<td>2.2% - 34% for five PFAS</td>
<td>Prospective cohort (the Nurses’ Health Study)</td>
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All other exposure assumption being equal, lower RSC values correspond to lower drinking water guideline levels

(Cordner et al., JESEE, 2019; Hu et al., EHP, 2019; Ali et al., ECEC19, 2019)
We used paired tap water and serum to quantify the relative source contribution (RSC) of drinking water PFAS to overall exposure.

Participants from geographically diverse areas
Representative of US background population

Hu et al., 2019, EHP
Tap water PFOA and PFNA are statistically significant predictors of serum in 1990

Hu et al., 2019, EHP

Huo et al., 2019, EHP
Pilot data suggest large increase in unidentified PFAS in drinking water: Consistent with production trends

4700 PFASs

>200 detected

Toxicity of alternative PFASs not well understood

Hu et al., 2019, EHP
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*SITIESCALES*
PFAS measurements in plankton suggest some precursors and linear isomers may bioaccumulate more than the terminal PFAAs.

\[ Y = (0.76 \pm 0.06)X - (3.82 \pm 0.55) \]

\[ R^2 = 0.87 \]

Zhang et al., 2019, Environmental Science & Technology
Large and rapid declines in modeled North Atlantic seawater PFOS (10 m)

X. Zhang et al., 2017, Global Biogeochemical Cycles
Measured targeted PFAS concentrations in North Atlantic pilot whales shows a rapid decline in FOSA, a PFOS precursor since 2000.

Dassuncao et al., 2017, ES&T
Declining atmospheric FOSA successfully predicts observed changes in pilot whale FOSA concentrations

Environmental Concentration

Pilot whale FOSA concentration (ng\textsuperscript{-1} g\textsuperscript{-1})

Dassuncao et al., 2017, ES&T
Are high seafood consuming populations mainly exposed to PFAS from seafood? Longitudinal measurements in Faroese kids

Dassuncao et al., 2018, ES&T
Rapid declines in targeted PFAS in children’s serum driven mainly by PFOS, PFOA, and FOSA

Some long chain PFAS (i.e., PFNA) stable or increasing

Dassuncao et al., 2018
Long-chain PFAS in serum (i.e., C>9) good tracer for seafood consumption

Environmental Health

Can profiles of poly- and Perfluoroalkyl substances (PFASs) in human serum provide information on major exposure sources?

Xindi C. Hu¹, Clifton Dassuncao¹,², Xianming Zhang³, Philippe Grandjean¹,³, Pål Weihe⁴, Glenys Lamport⁵, Flemming Nielsen⁵ and Elsie M. Sunderland¹,²
Long-Chained PFCAs strongly associated with seafood consumption

Faroese Children

Dassuncao et al. 2018

NHANES 2005-2006

Hu et al. 2018
Decline in serum PFAS concentrations can not be explained by shifts exposure from seafood consumption.

Even in the Faroe Islands (remote high seafood consuming population), diverse consumer products appear to have accounted for the majority of exposures for children in the 1990-2000s.
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*SOURCES*

Sunderland et al., 2019, JESEE
Serum concentrations of some PFAS have been linked to use of food packaging in the general U.S. population. Banning PFAS in food packaging (i.e., Denmark) could lead to a rapid reduction in general population exposures.

Hu et al., 2018, Environmental Health
Targeted LC-MS/MS measurements make up SMALL fraction of total PFAS in consumer products

Tokranov et al., 2019, ES&T
Extremely high concentrations of fluorinated compounds measured in a variety of cosmetic products

Total fluorine
Extractable organic fluorine (EOF)
∑PFAS
No listed fluorinated ingredient

Schultes et al., 2018

NHANES 2013-2014
Geometric mean serum: 1.94 (1.76–2.14) ng/mL

→ up to 2.2 ng/ml [high exposure scenario]
→ 0.0011 ng/ml [intermediate scenario]
Summary

- RSC of 20% seems appropriate for legacy PFAS in drinking water but does not account for newer compounds that appear to be increasing;

- Concerted global regulatory actions on PFOS and PFOA, in partnership with industry, have led to rapid declines in human exposures to these compounds;

- Seafood is an important exposure pathway for legacy PFASs and may be growing in importance for some populations;

- Consumer products (direct use of PFAS) main driver of serum concentrations even among high-seafood consuming populations;

- Total organofluorine (PIGE, TOP, CIC, XPS) methods are needed to screen for new and unidentified PFAS.