Understanding Diverse Exposure Pathways for PFAS

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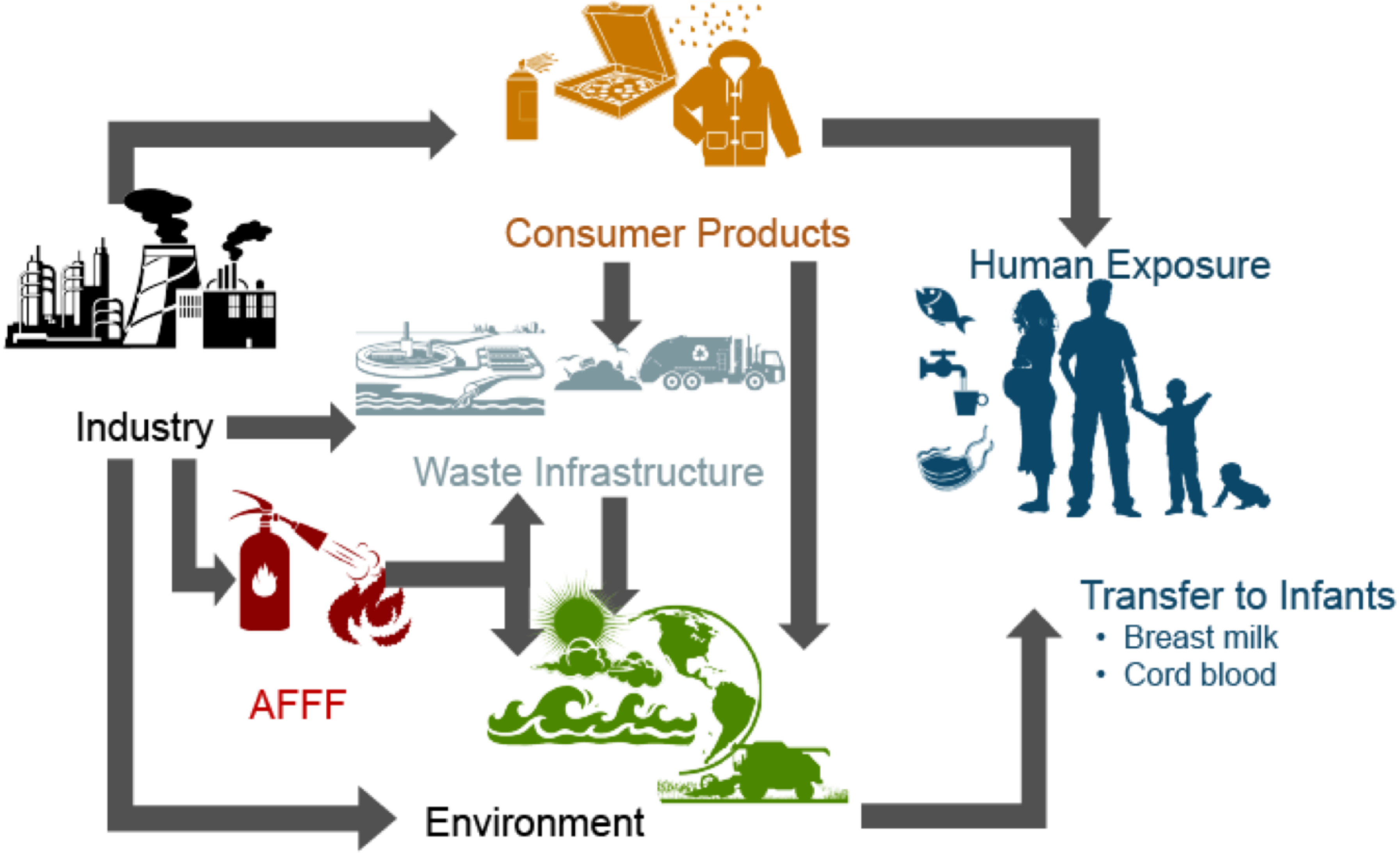
Biogeochemistry of Global Contaminants
HARVARD

STEEP
Sources, Transport, Exposure & Effects of PFASs
UNIVERSITY OF RHODE ISLAND SUPERFUND RESEARCH PROGRAM

Human exposures to PFAS are diverse: Some can be addressed/mitigated faster than others



Our focus:

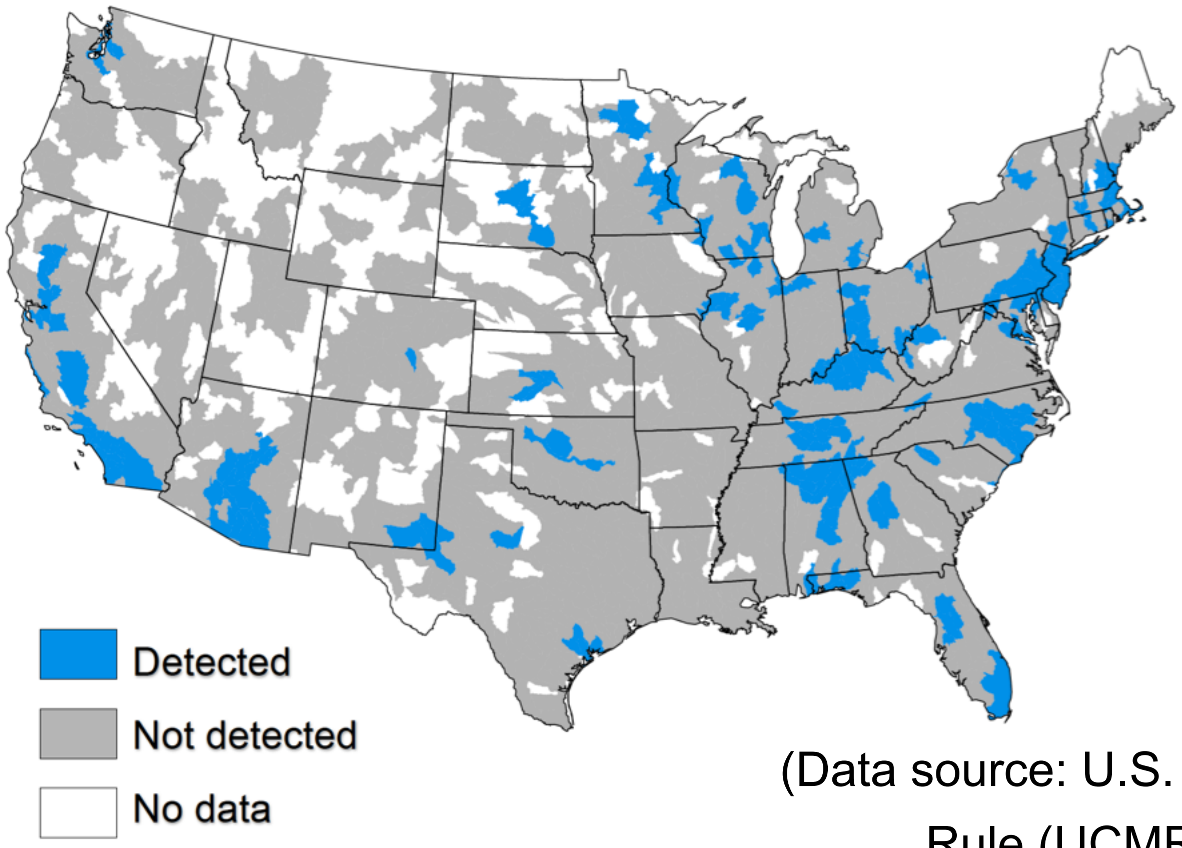
1. **Drinking water**
2. **Seafood**
3. **Consumer Products**

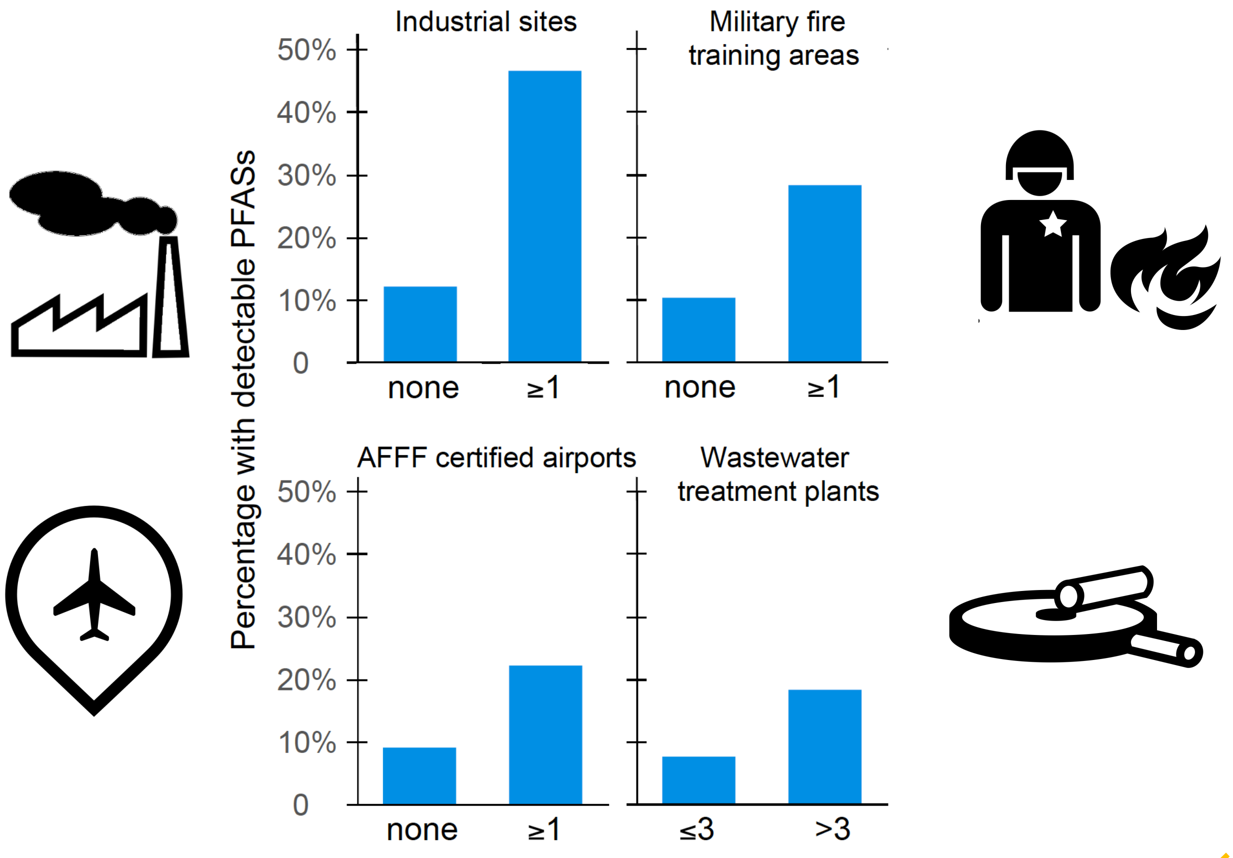
TIMESCALES

Sunderland et al., 2019, JESEE

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

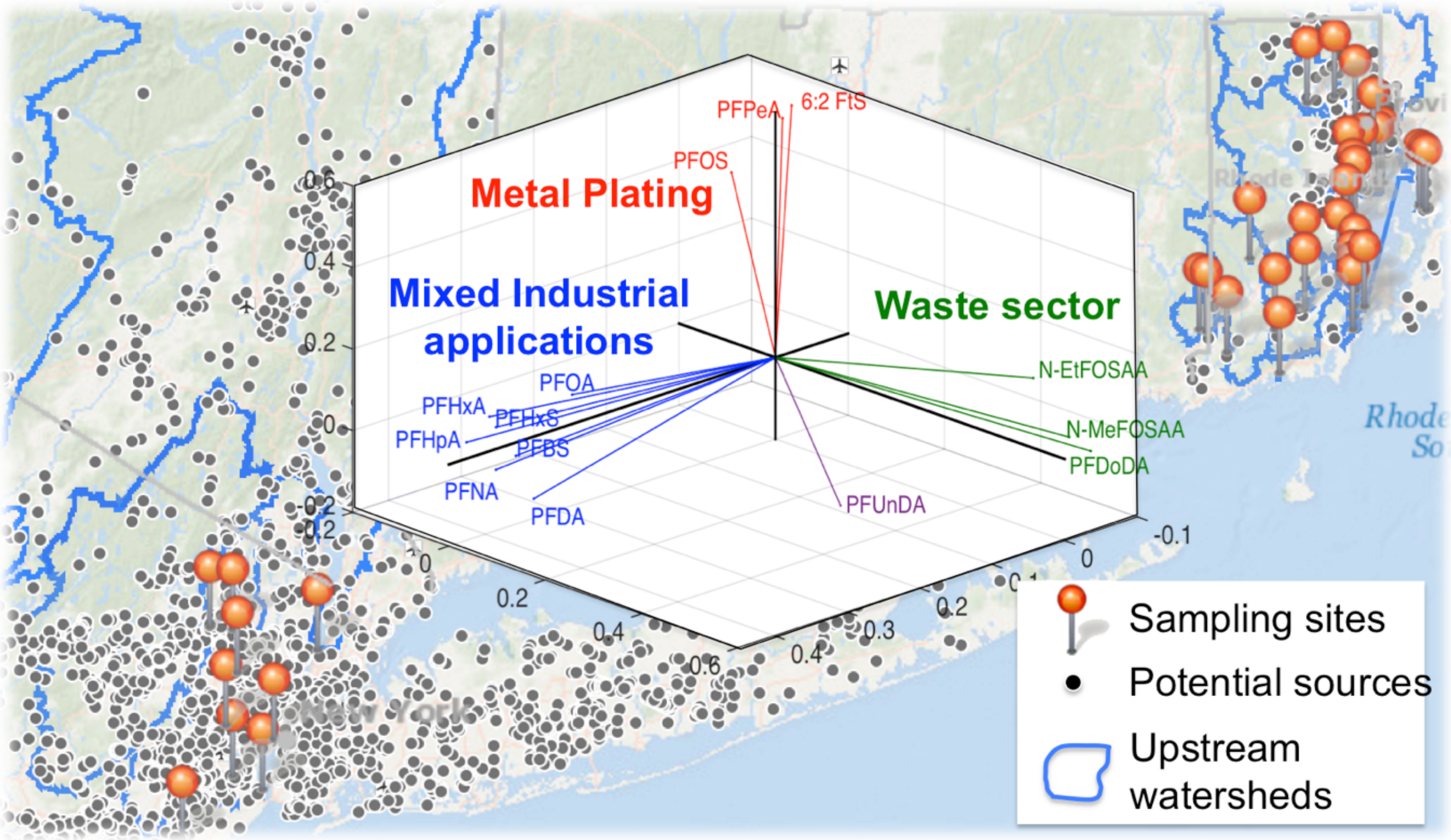
Hydrological units with detectable PFASs





(Data source: U.S. EPA 3rd Unregulated Contaminants Monitoring Rule (UCMR3), 2013-2015) (Hu et al., *ES&T Letters*, 2016)

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

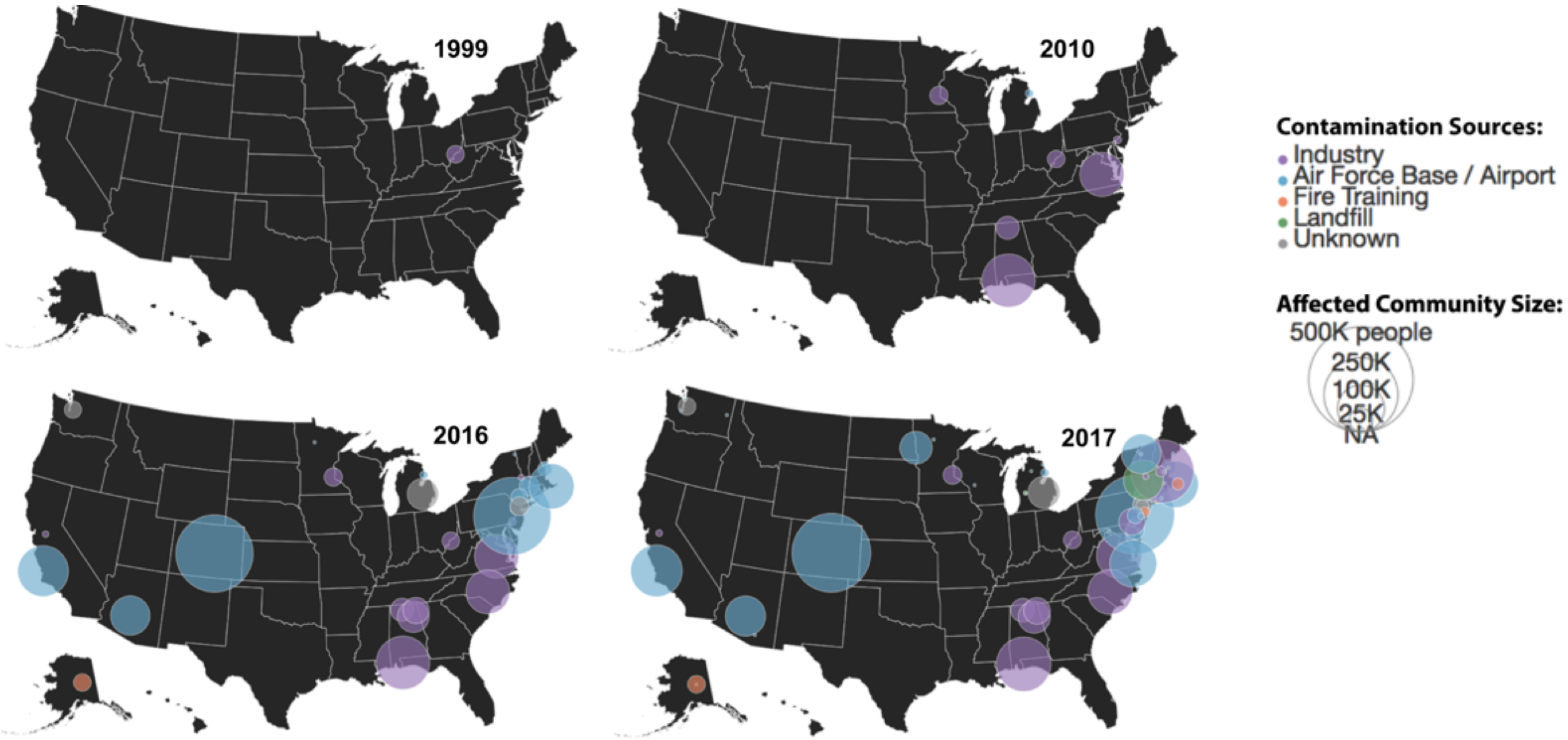
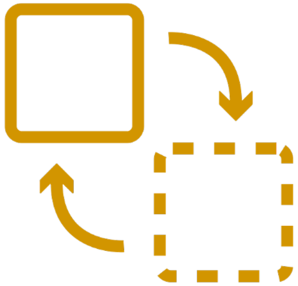


Figure adapted from data compiled by Northeastern University's Social Science Environmental Health Research Institute (SSEHRI)

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

Dietary

Life style

Residential location

1989/1990

Serum and tap water collection

n = 32,826

What are exposures now?

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

Reference dose → Exposure assessment → Concentration in drinking water

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

Entity	RSC	Source
Federal EPA, most states	20% for all PFAS	EPA grey literature
Minnesota, Maine, New Hampshire	20% - 60% for PFOA and PFOS	Human biomonitoring studies
Alaska, Texas	100%	Developed for remediation and clean-up of contaminated sites
Our study (NHS)	2.2% - 34% for five PFAS	Prospective cohort (the Nurses' Health Study)

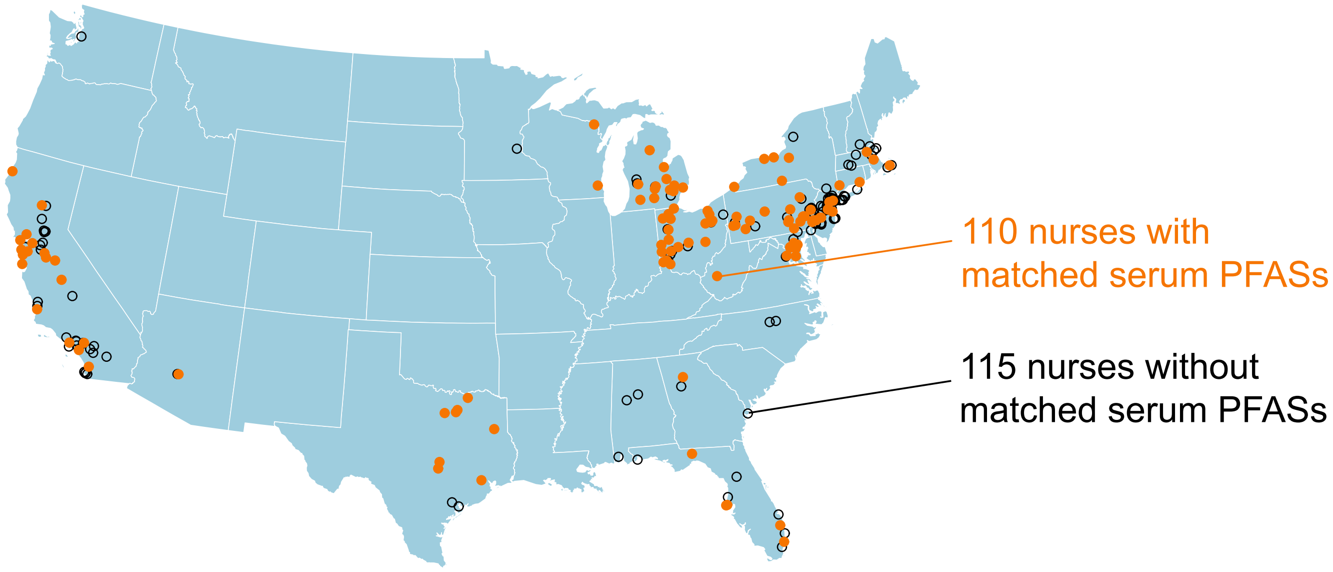
All other exposure assumption being equal, lower RSC values correspond to lower drinking water guideline levels

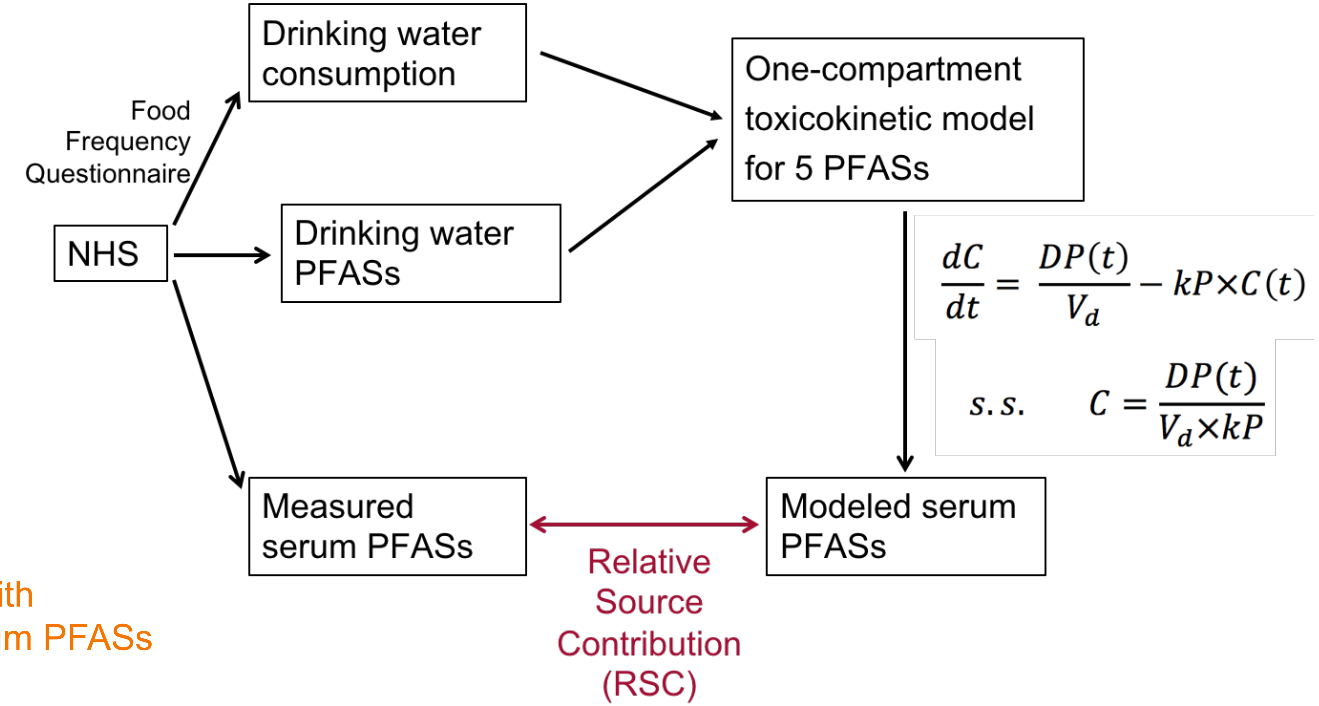
(Cordner et al., *JESEE*, 2019;
Hu et al., *EHP*, 2019;
Ali et al., ECEC19, 2019)

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

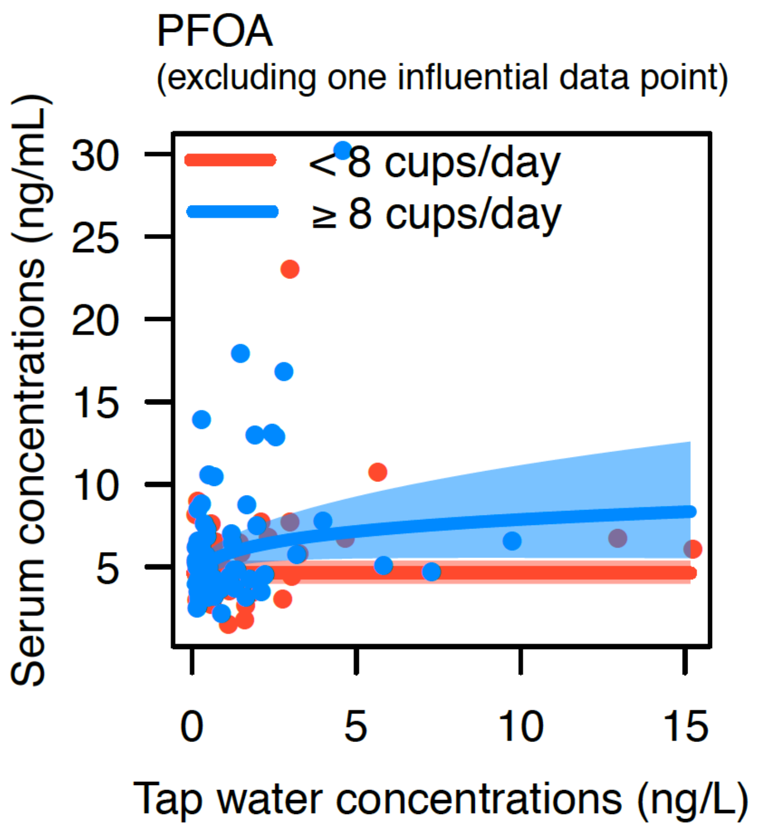
Representative of US background population

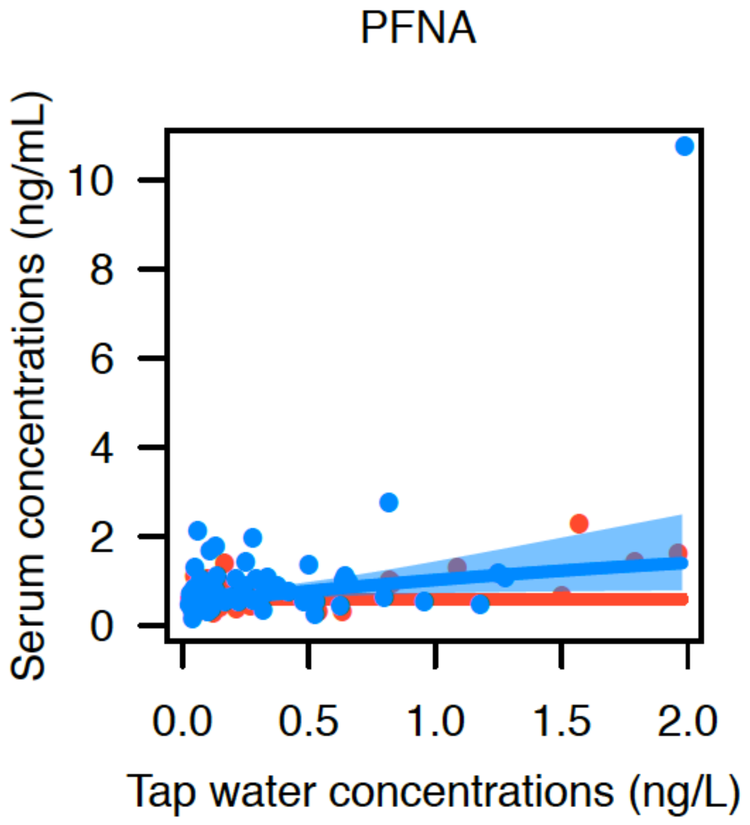




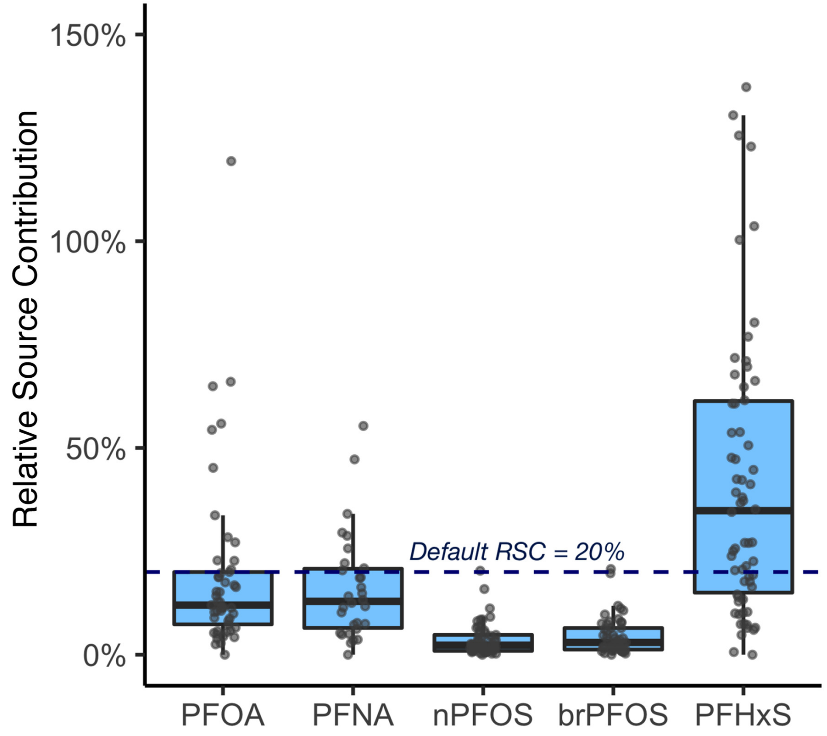
Hu et al., 2019, EHP

Tap water PFOA and PFNA are statistically significant predictors of serum in 1990



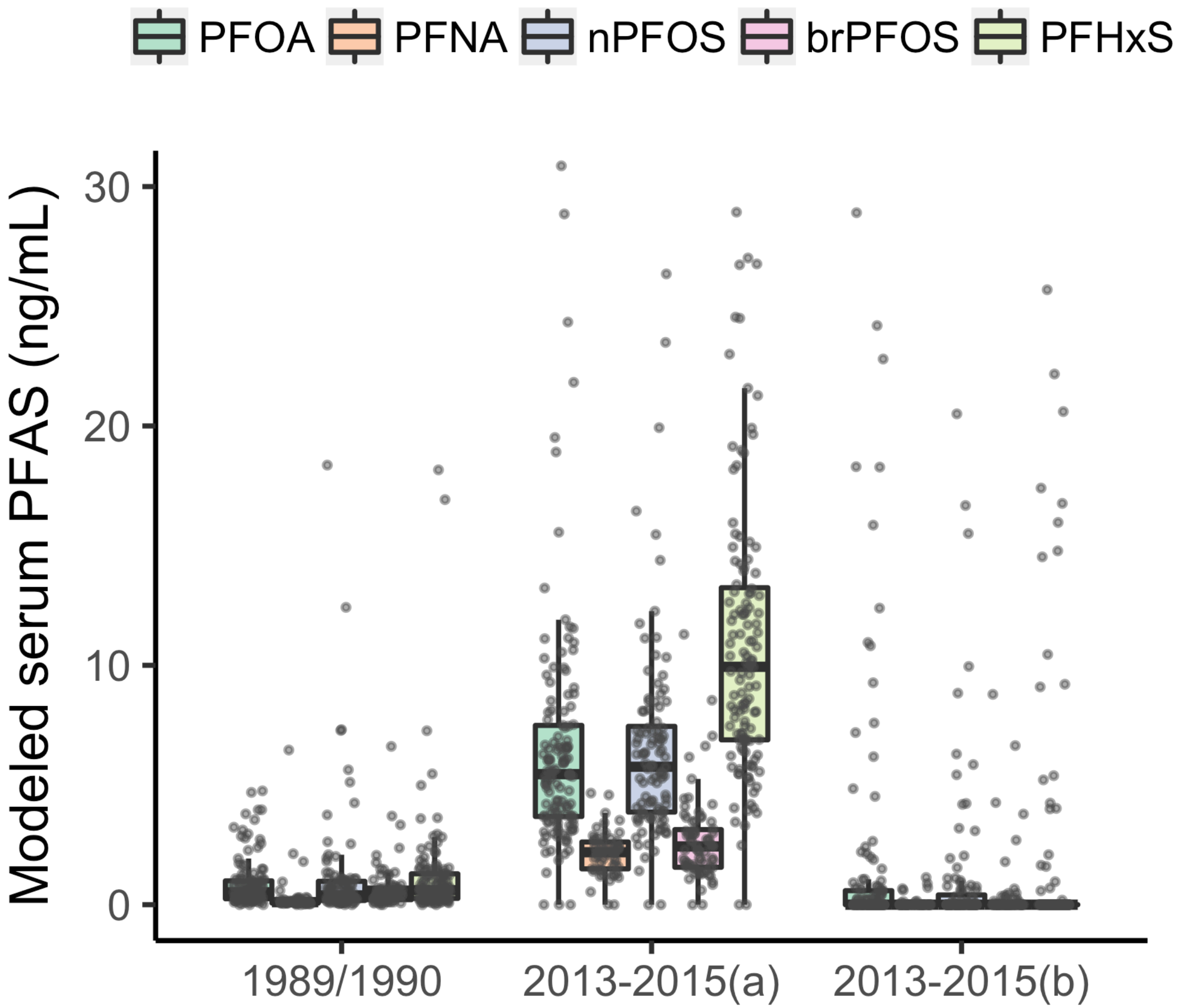


RSC = 2% - 34%



Hu et al., 2019, EHP

Modeled NHS serum PFAS concentrations from drinking water exposures based on drinking water in 1989/1990 and 2013-2015



Hu et al., 2019, EHP

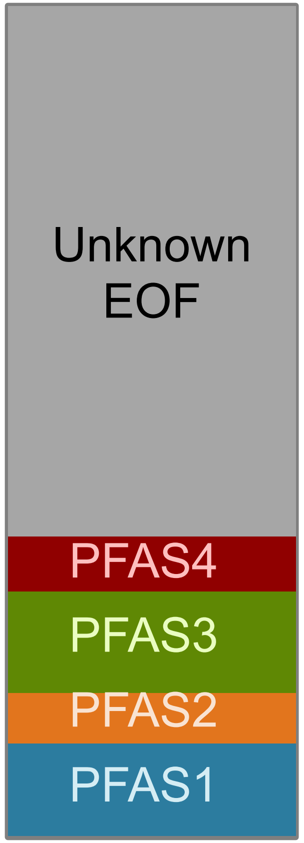
Pilot data suggest large increase in unidentified PFAS in drinking water: Consistent with production trends

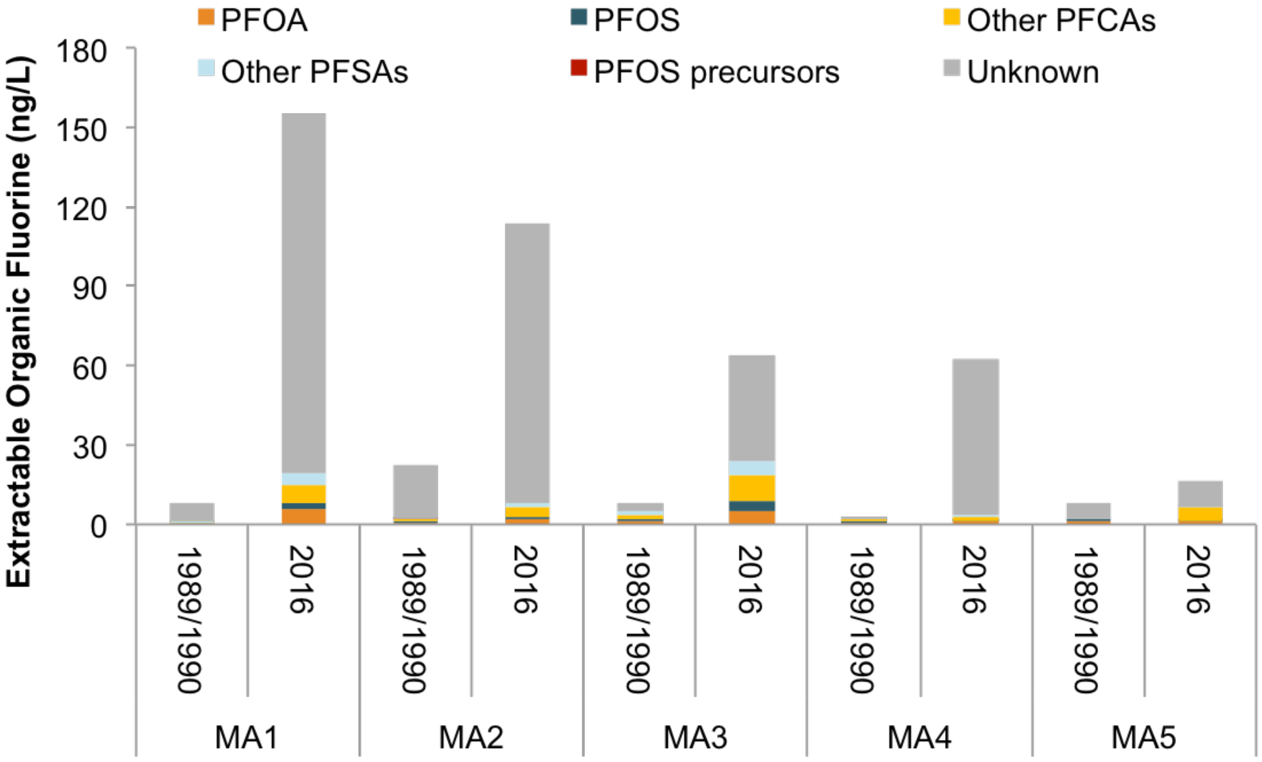
Extractable organic fluorine (EOF)

4700 PFASs

>200 detected

Toxicity of alternative PFASs not well understood

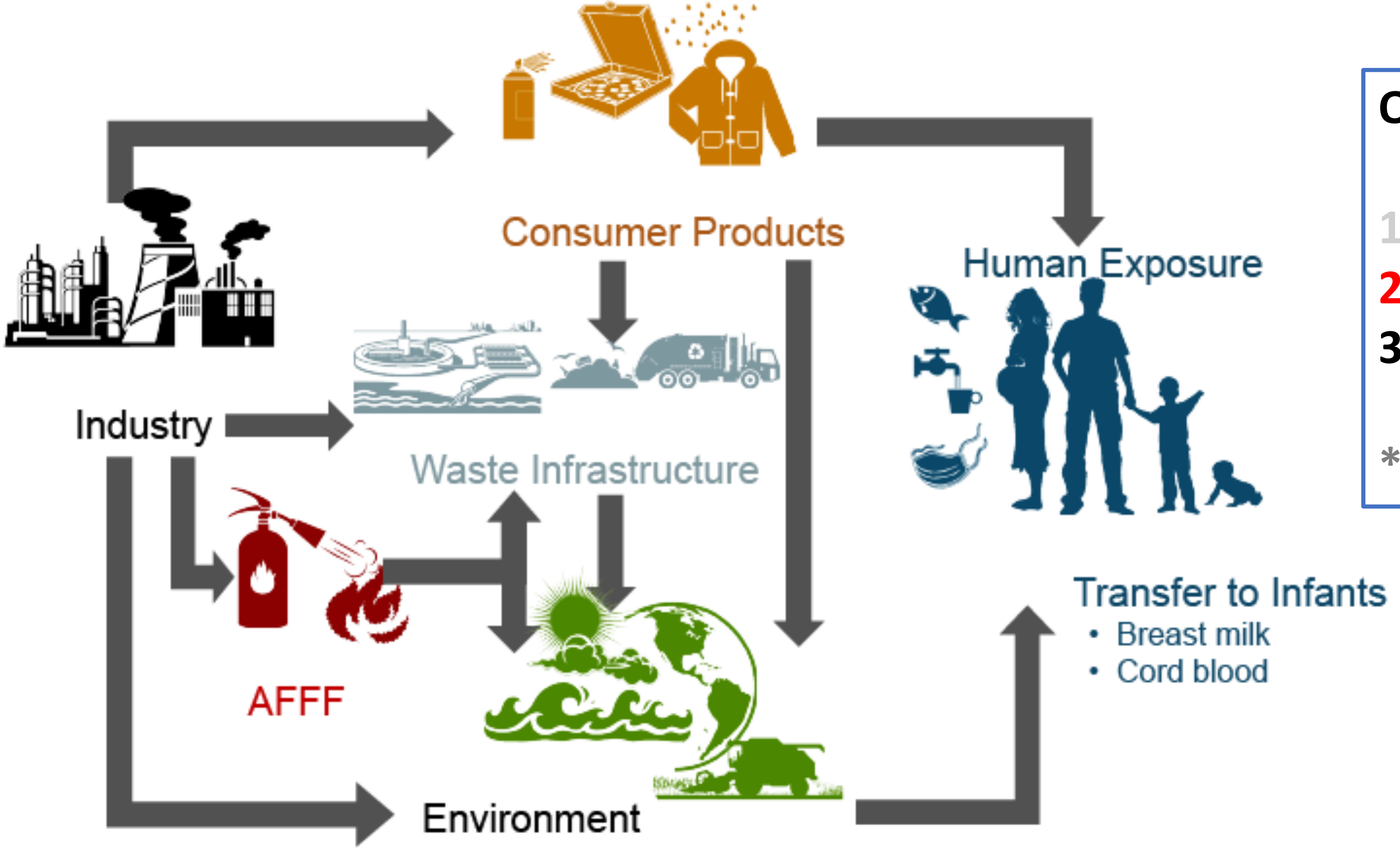




% unknown EOF: 8% - 89% in 1989/1990; 60% - 94% in 2016

Hu et al., 2019, EHP

Human Exposures to PFAS are Diverse: Some Can be Addressed/Mitigated Faster than Others



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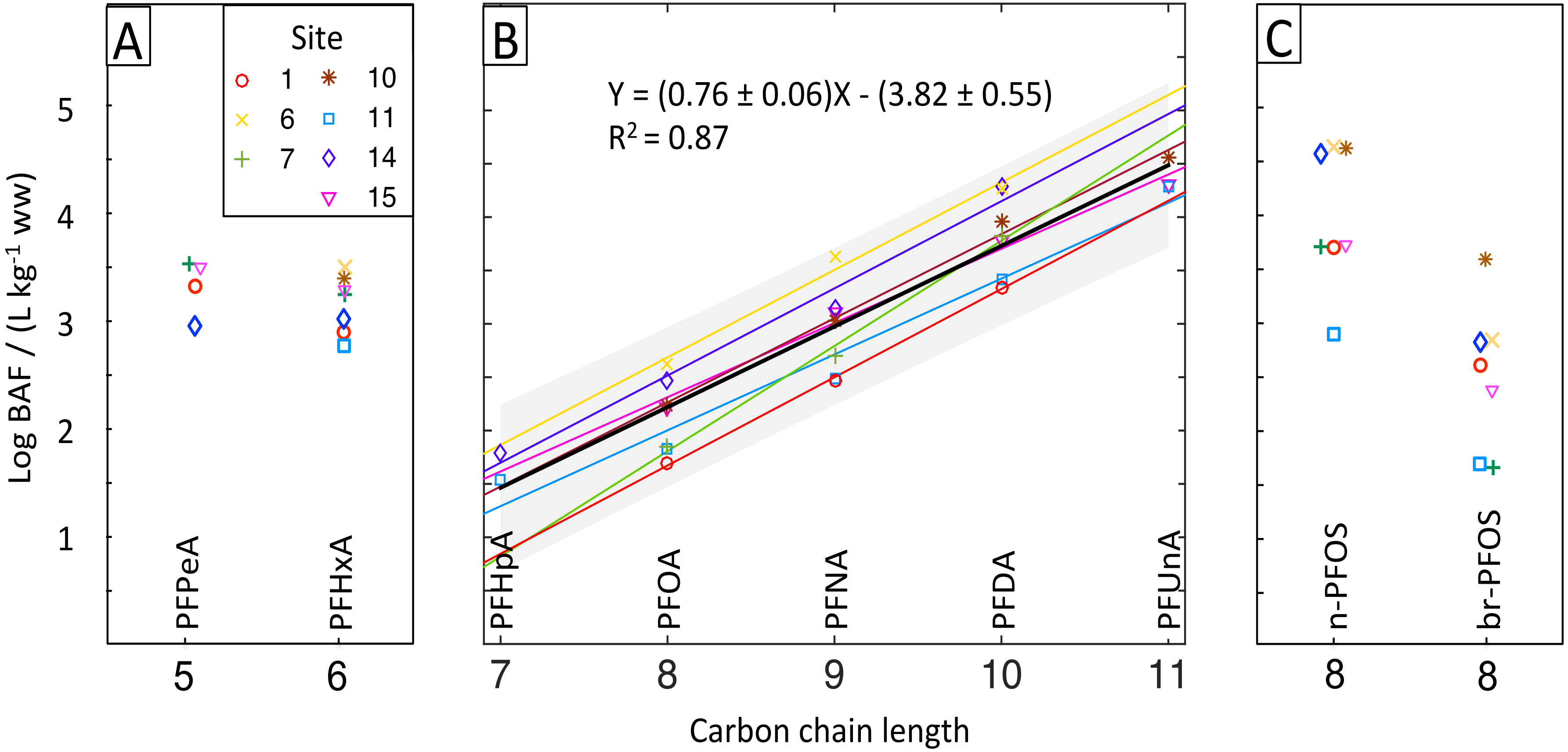
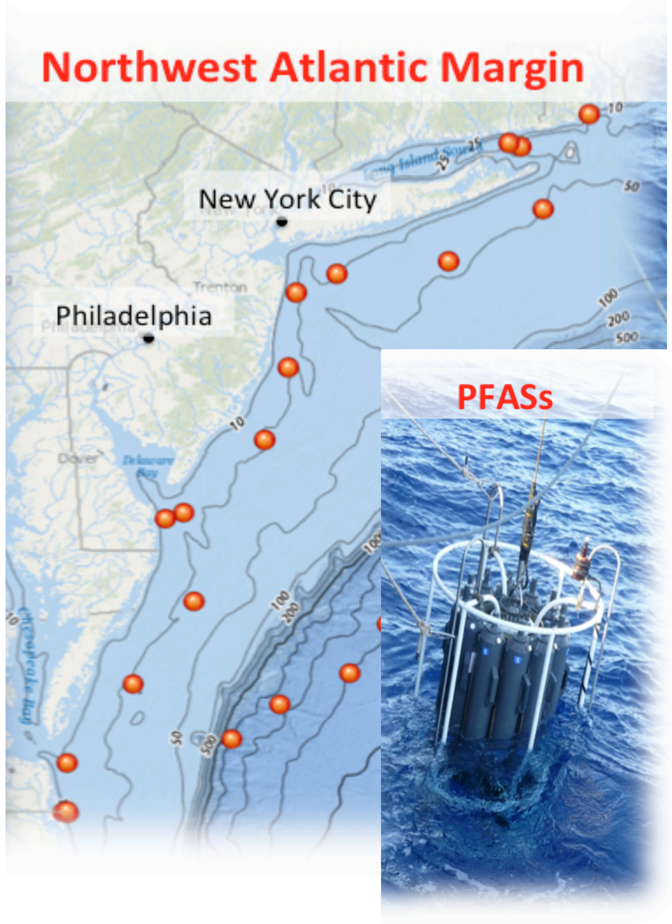
TIMESCALES

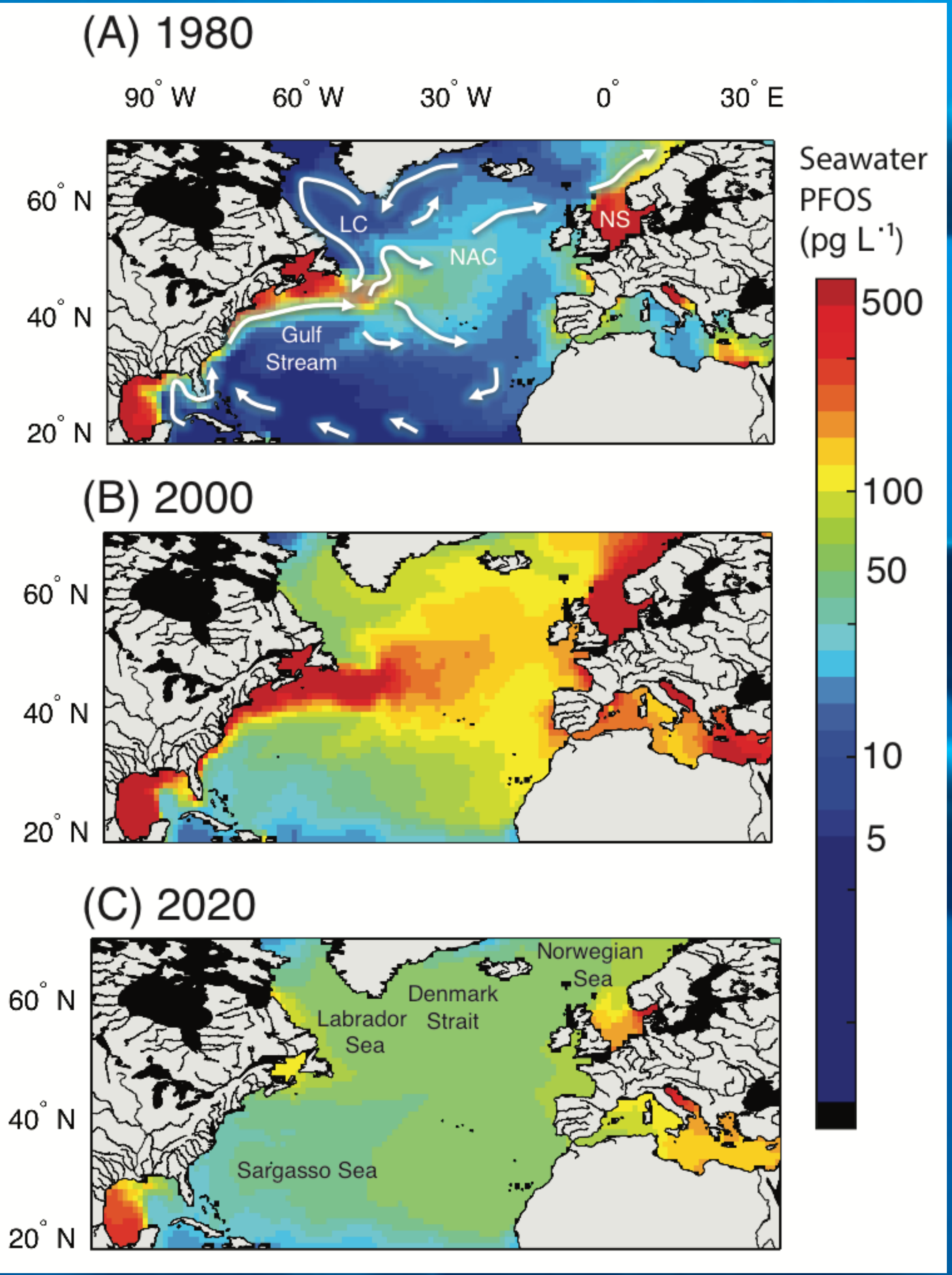
Sunderland et al., 2019, JESEE

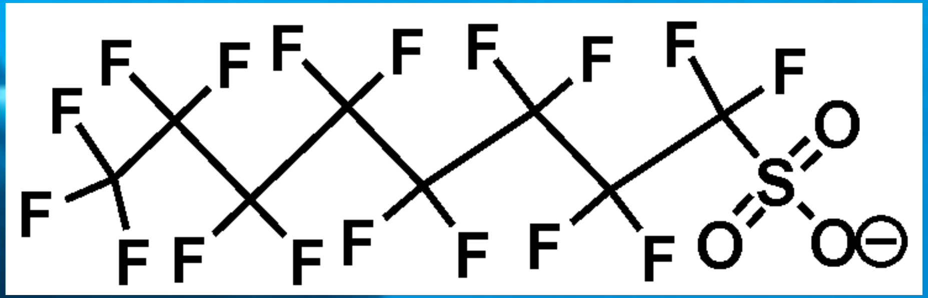
PFAS measurements in plankton suggest some precursors and linear isomers may bioaccumulate more than the terminal PFAAs





Zhang et al., 2019, Environmental Science & Technology

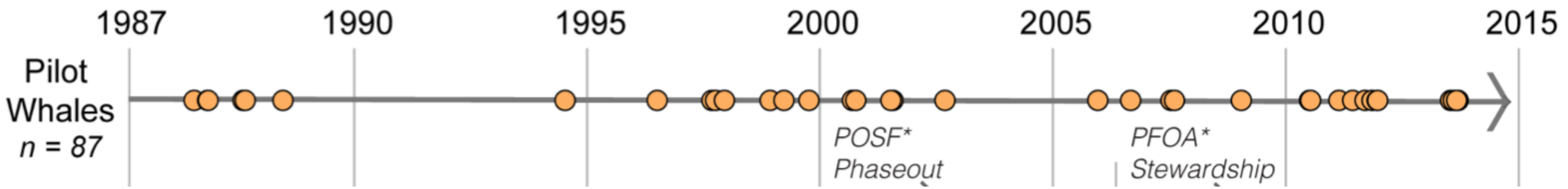


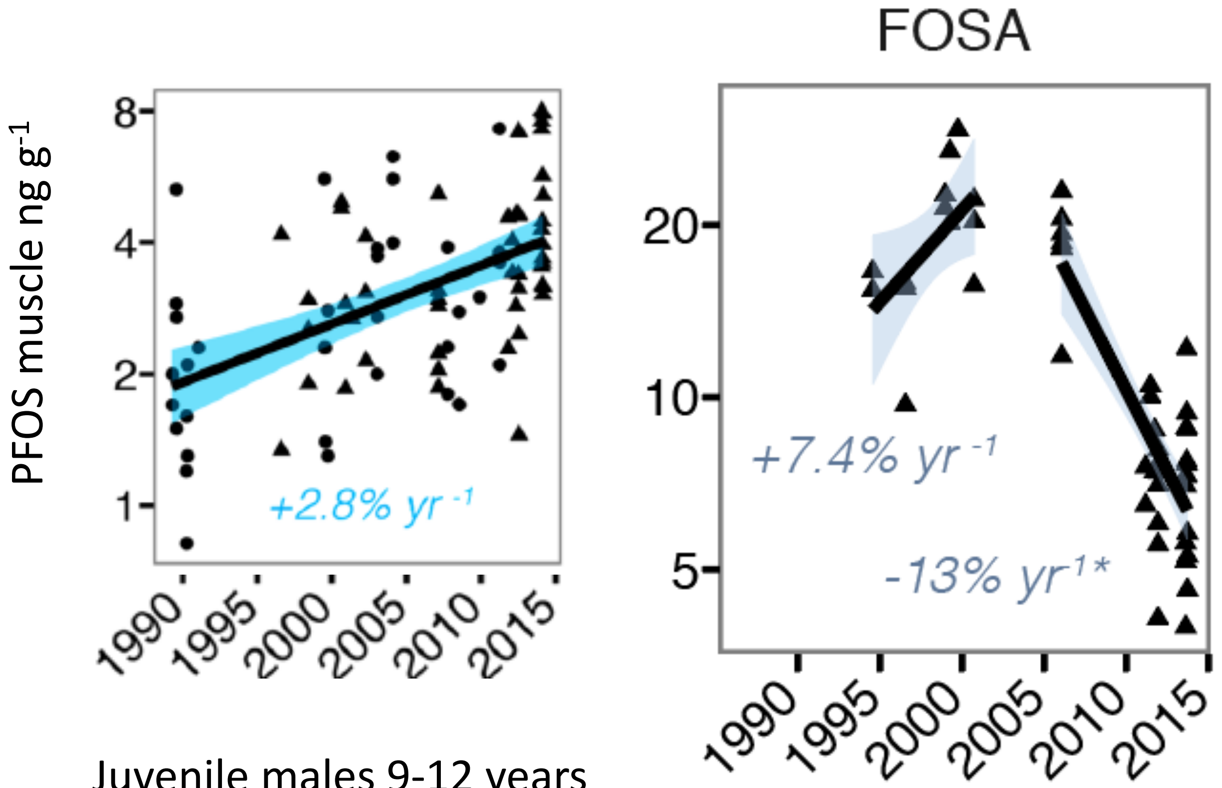


Large and rapid declines in modeled North Atlantic seawater PFOS (10 m)

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



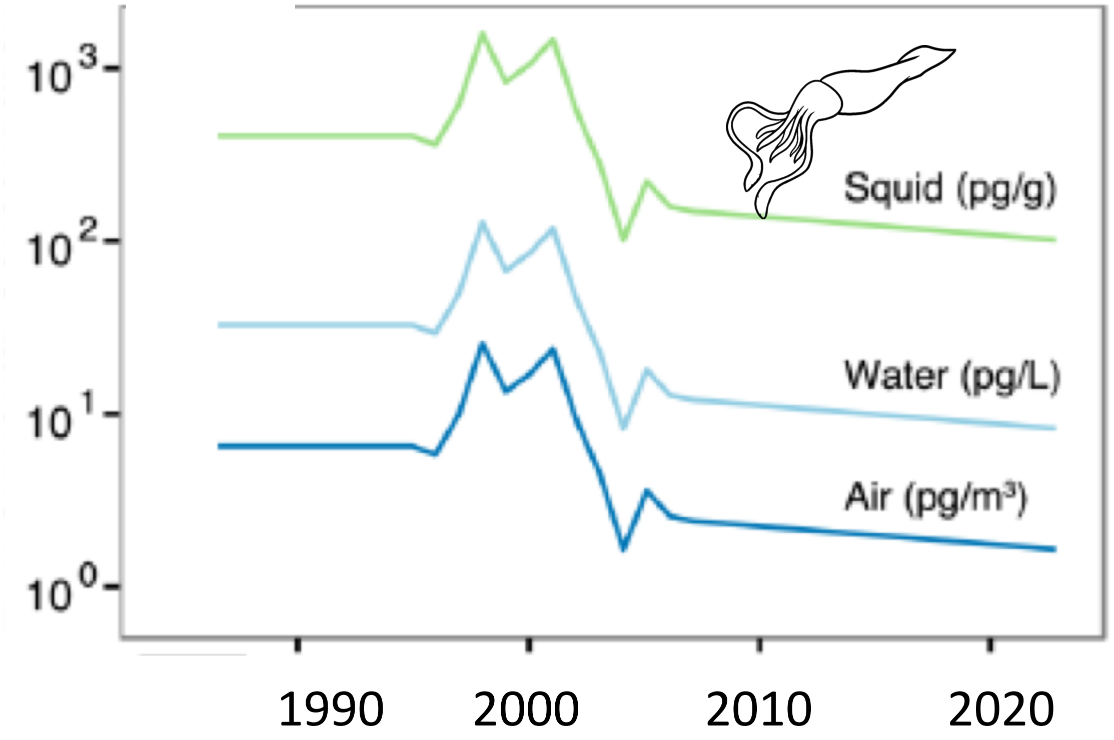


Juvenile males 9-12 years

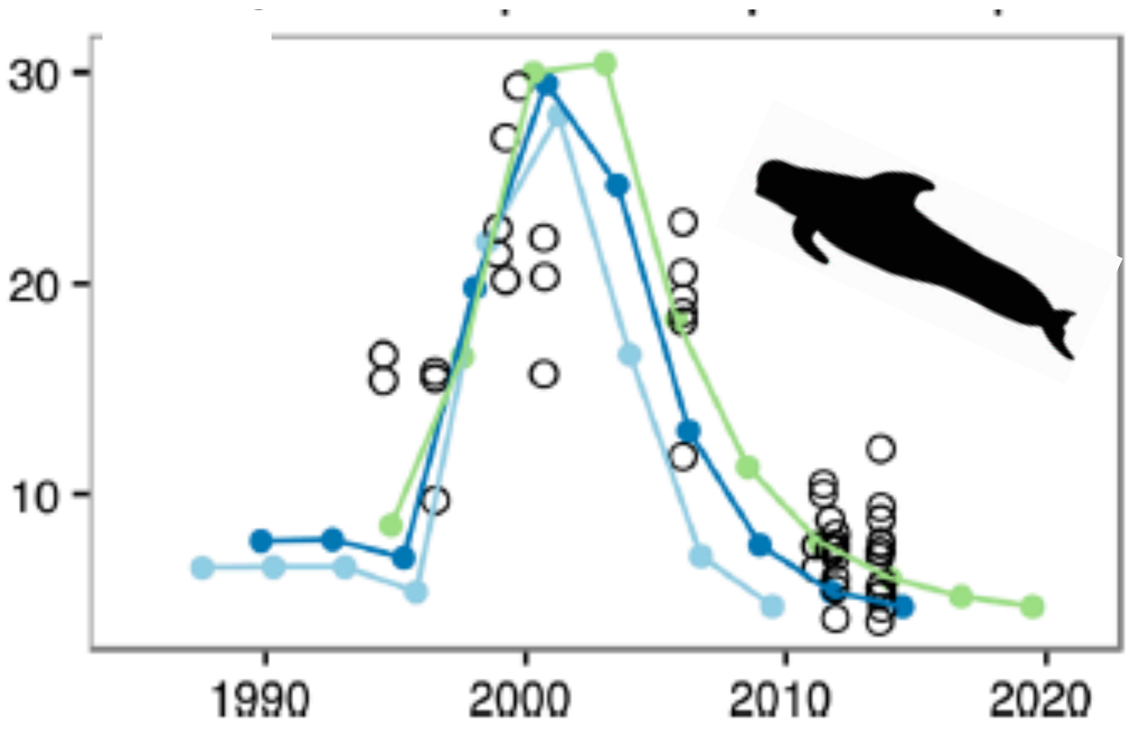
Dassuncao et al., 2017, ES&T

Declining atmospheric FOSA successfully predicts observed changes in pilot whale FOSA concentrations

Environmental Concentration

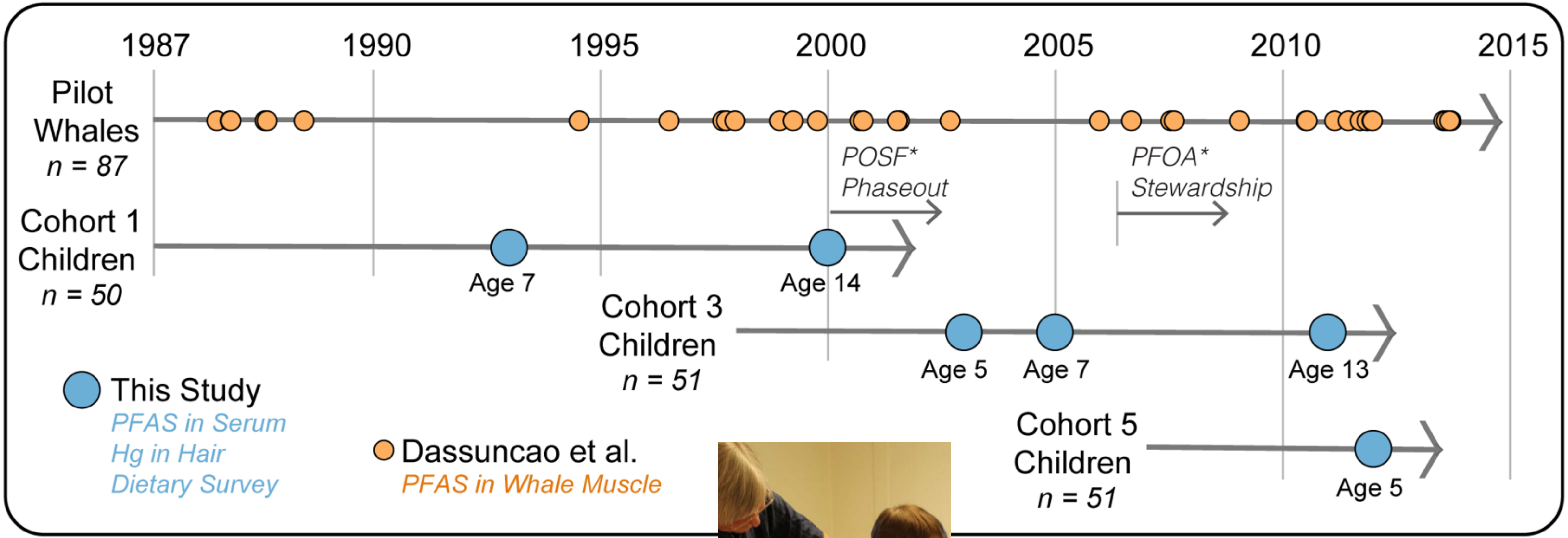


Pilot whale FOSA concentration (ng^{-1} g^{-1})



Dassuncao et al., 2017, ES&T

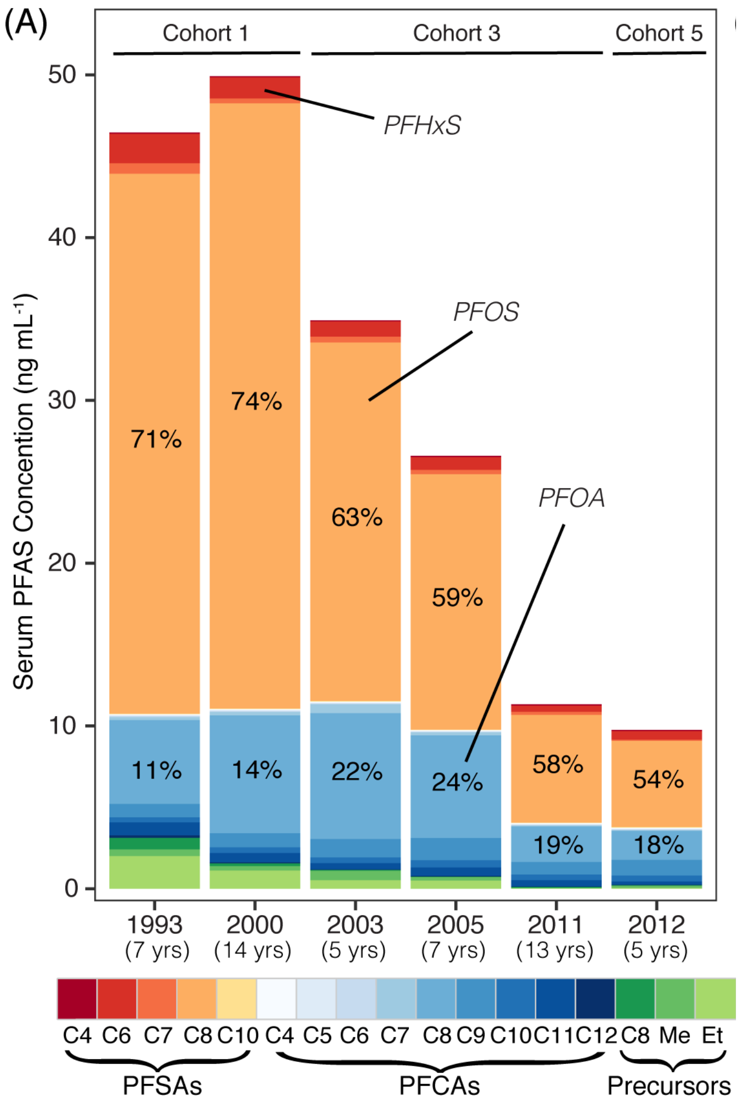
Are high seafood consuming populations mainly exposed to PFAS from seafood? Longitudinal measurements in Faroese kids

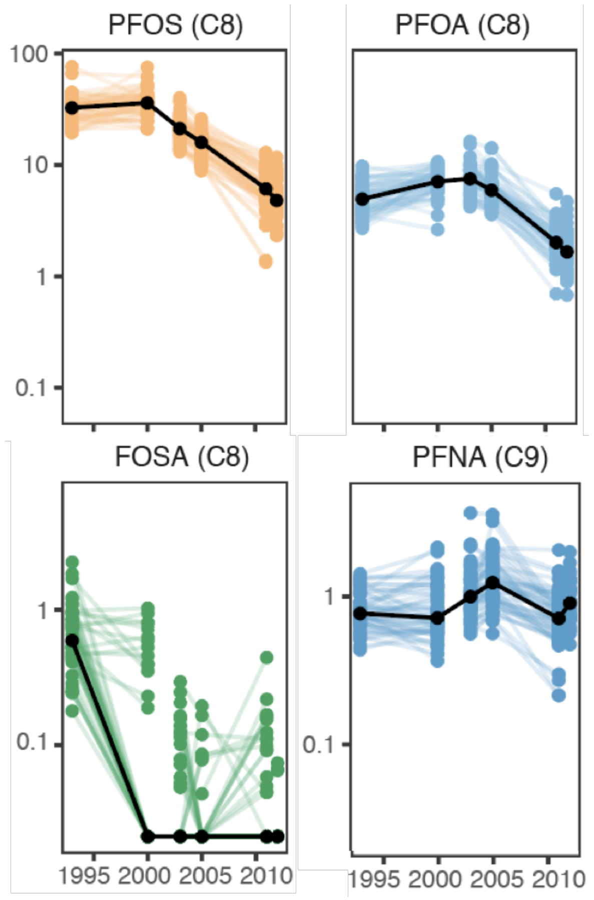


Dassuncao et al., 2018, ES&T

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





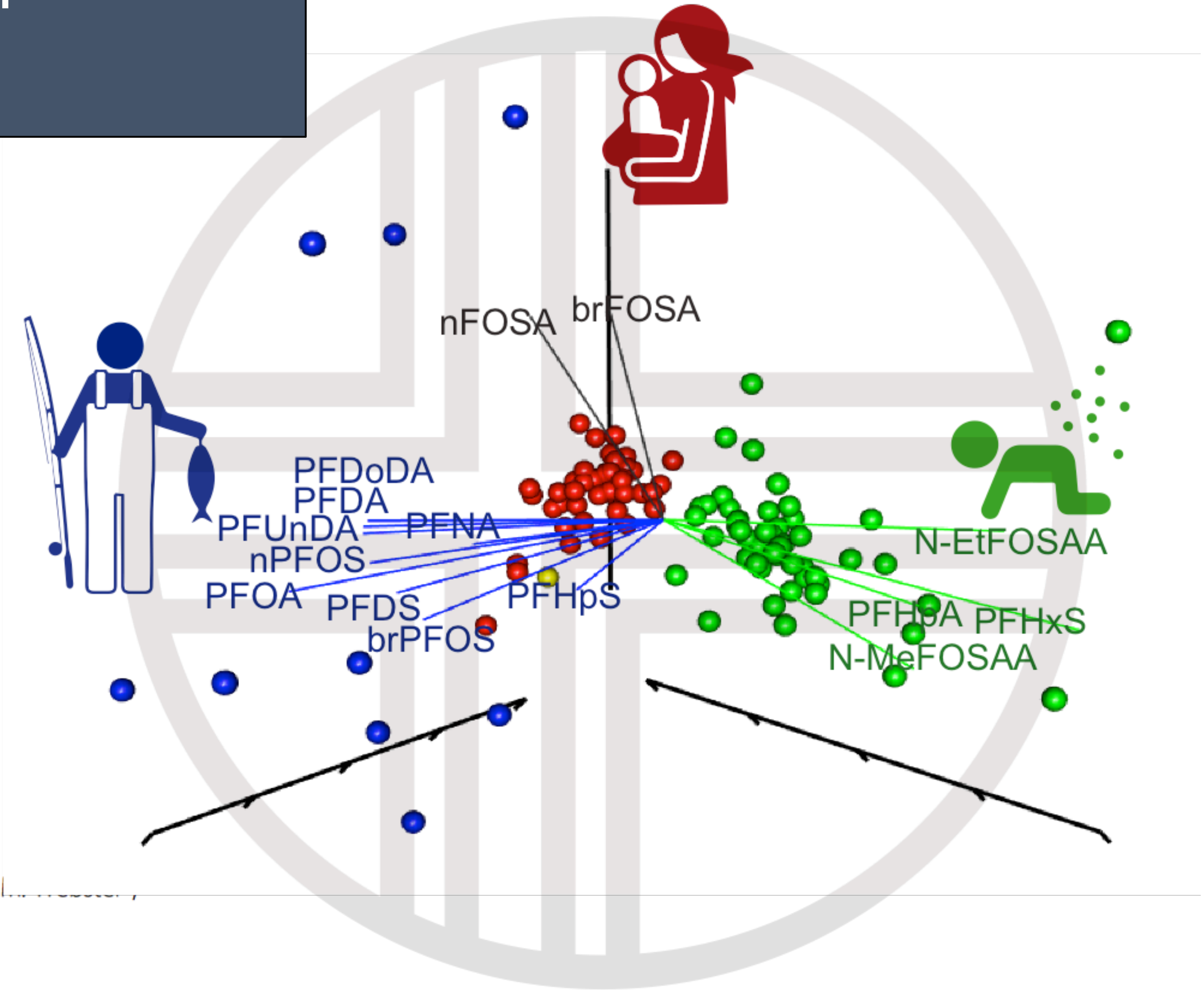
Dassuncao et al., 2018

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

Environmental Health

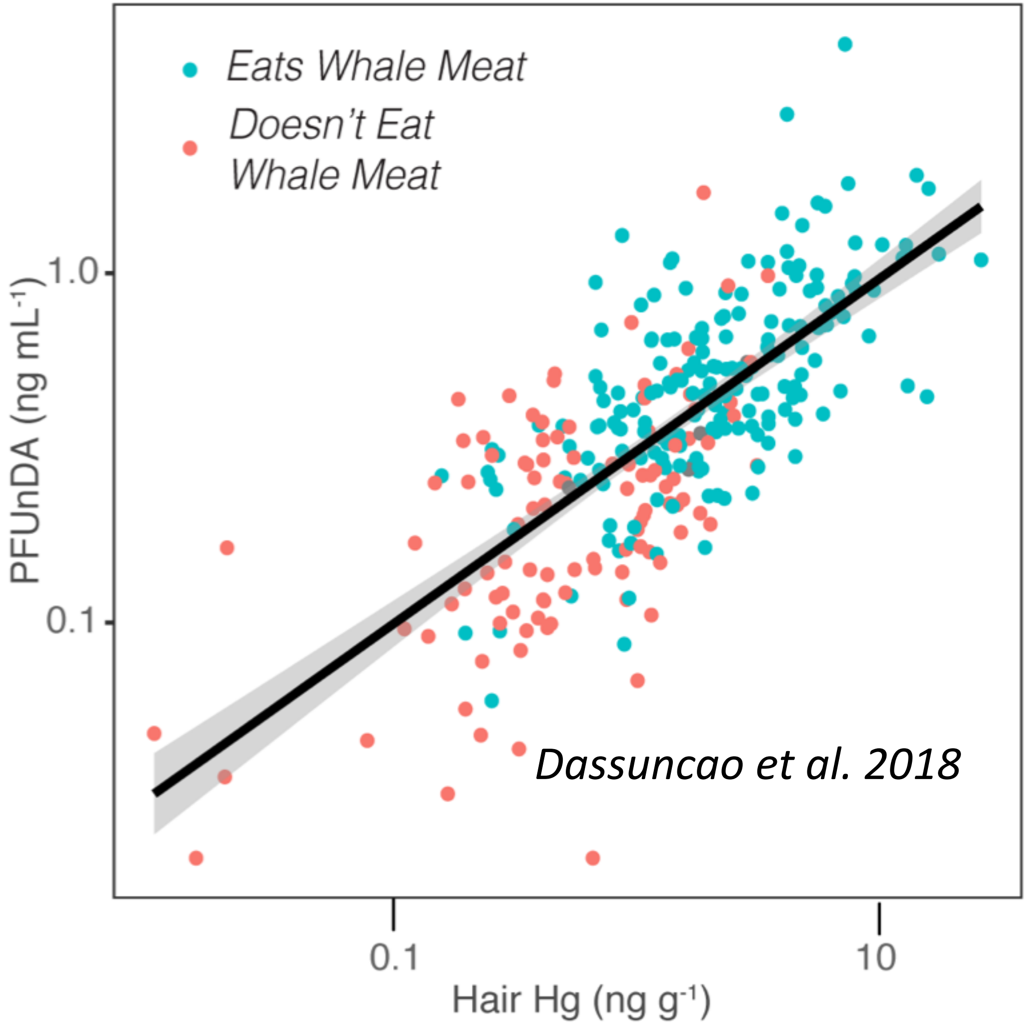
Can profiles of poly- and Perfluoroalkyl substances (PFASs) in human serum provide information on major exposure sources?

Xindi C. Hu[1,2*], Clifton Dassuncao[1,2], Xianming Zhang[2], Philippe Grandjean[1,3], Pál Weihe[4], Glenys
Flemming Nielsen[3] and Elsie M. Sunderland[1,2]

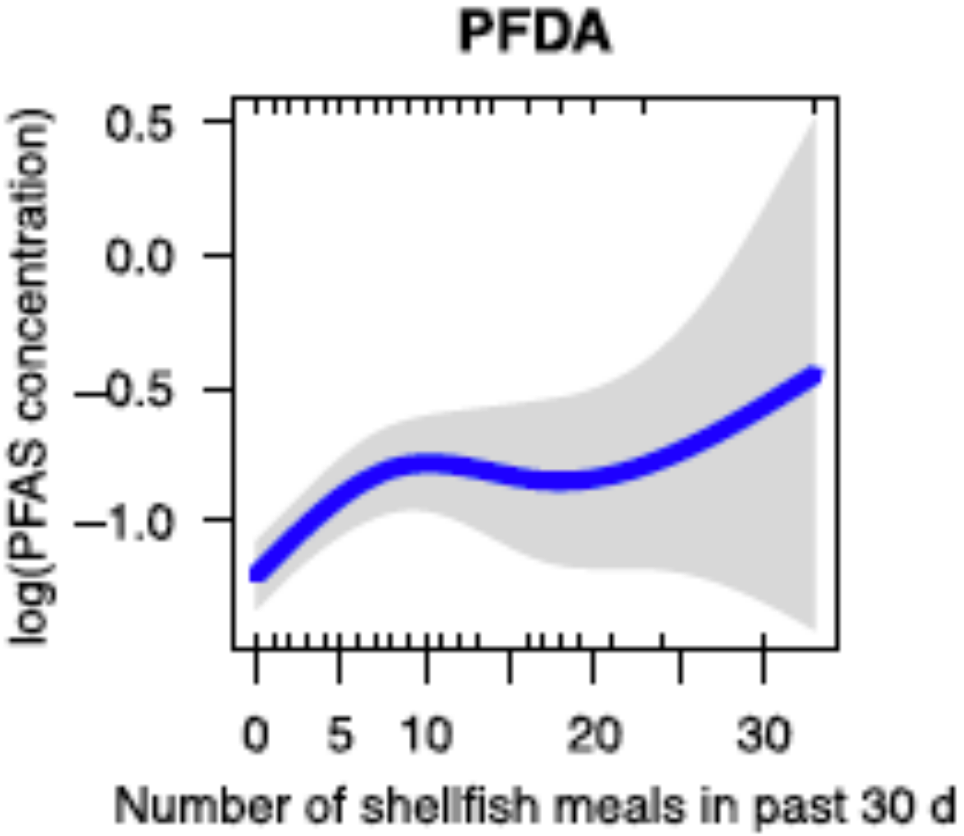


Long-Chained PFCAs strongly associated with seafood consumption

Faroese Children

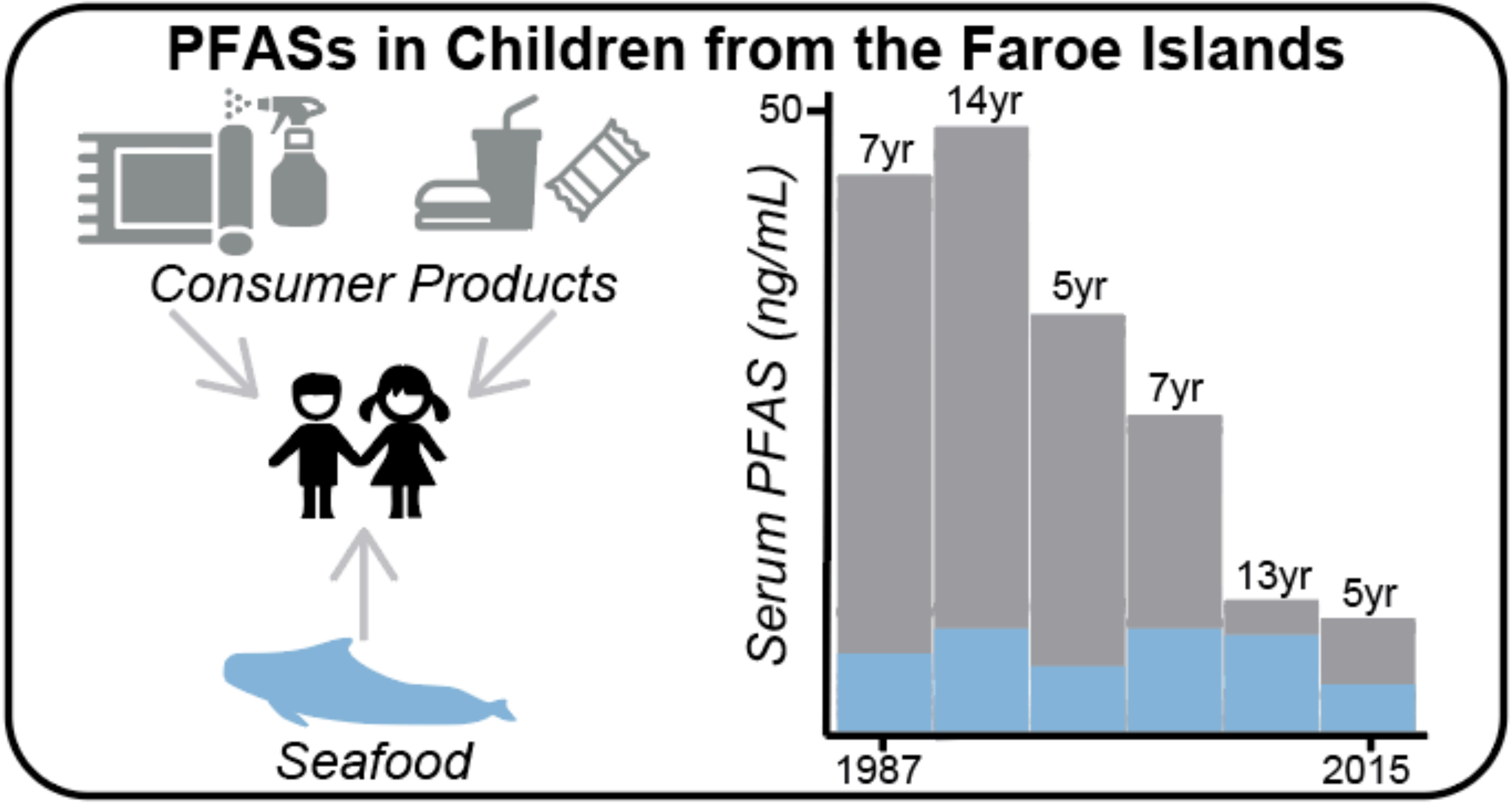


NHANES 2005-2006



Hu et al. 2018

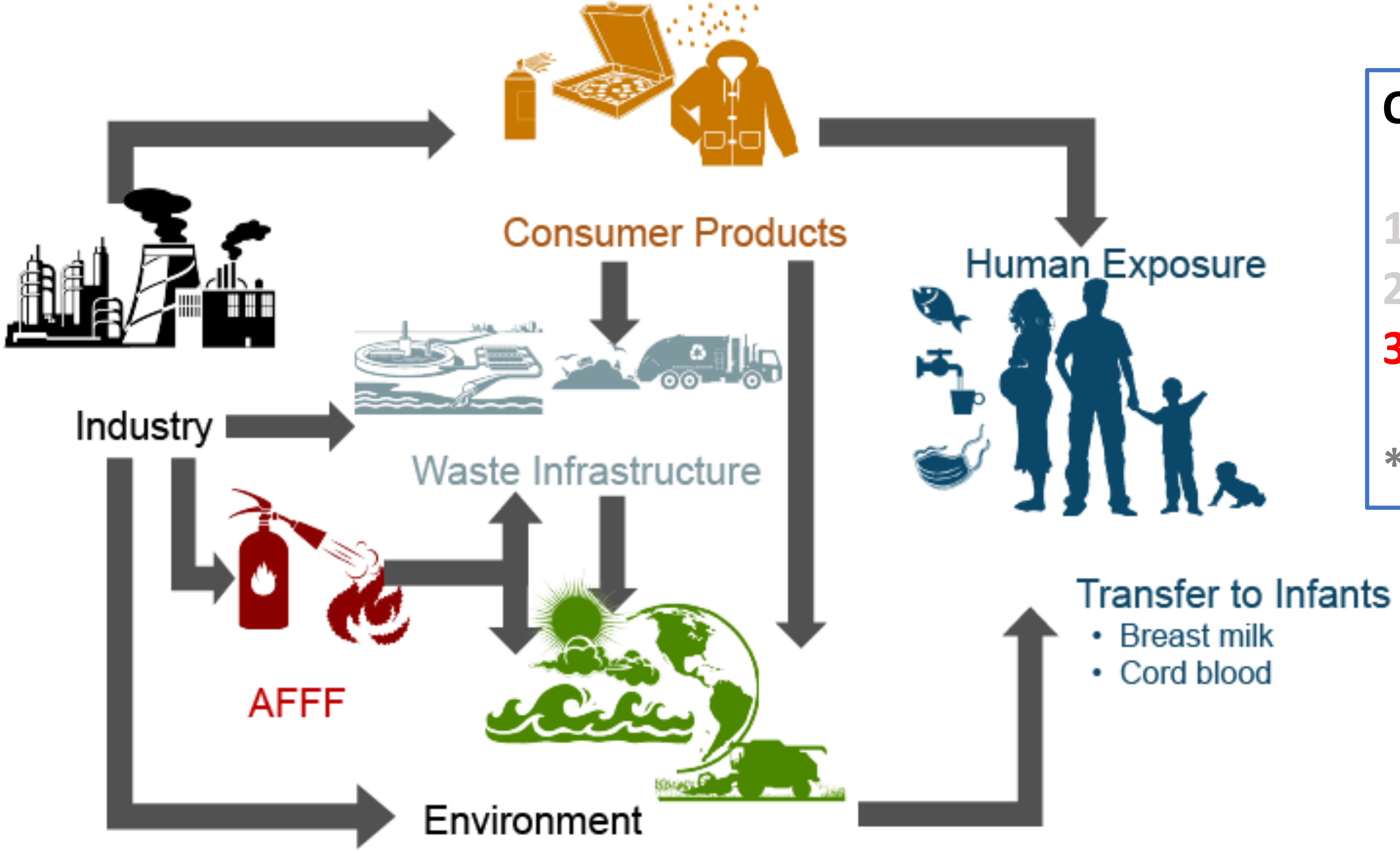
Decline in serum PFAS concentrations can not be explained by shifts exposure from seafood consumption



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.

Dassuncao et al. 2018

Human Exposures to PFAS are Diverse: Some Can be Addressed/Mitigated Faster than Others



Our focus:

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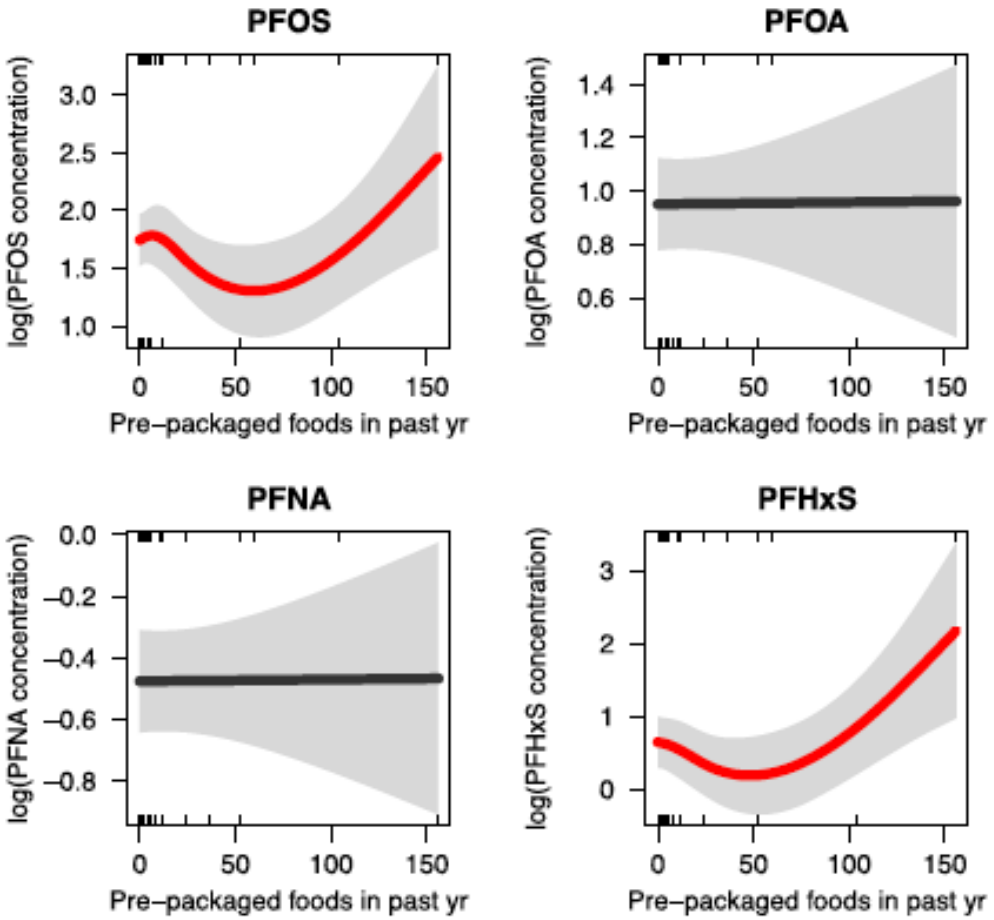
TIMESCALES

Sunderland et al., 2019, JESEE

Serum concentrations of some PFAS have been linked to use of food packaging in the general U.S. population



Hu et al., 2018, Environmental Health

Banning PFAS in food packaging (i.e., Denmark) could lead to a rapid reduction in general population exposures

BY SARAH GIBBENS 10 OCTOBER 2019

FAST FOOD INCREASES EXPOSURE TO A FOREVER CHEMICAL CALLED PFAS

Used in fast food packaging, the long-lasting chemicals can seep into food—and build up in our bodies.

FOLLOW SHARE TWEET

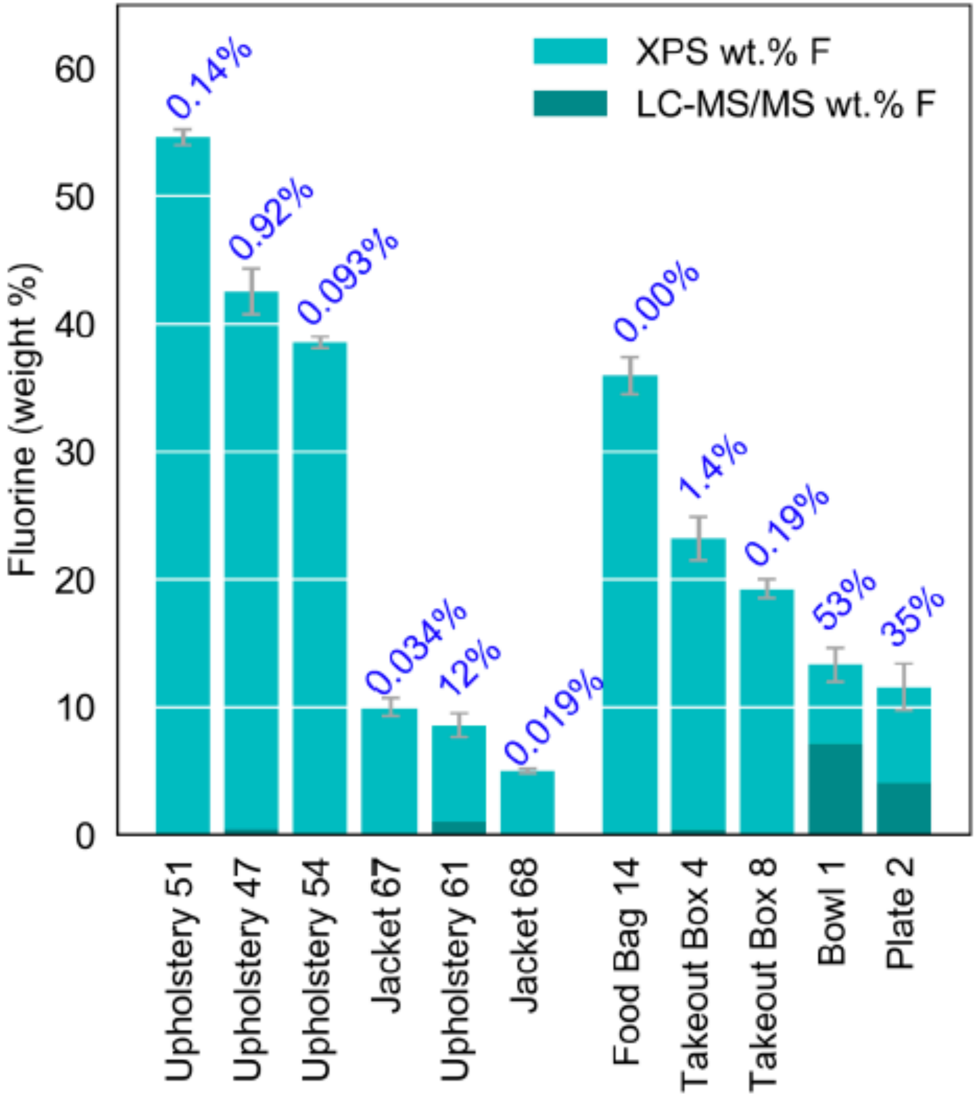
Targeted LC-MS/MS measurements make up SMALL fraction of total PFAS in consumer products

Environmental Science & Technology Letters
Letter
Cite This: Environ. Sci. Technol. Lett. 2019, 6, 38−43
pubs.acs.org/journal/estlcu

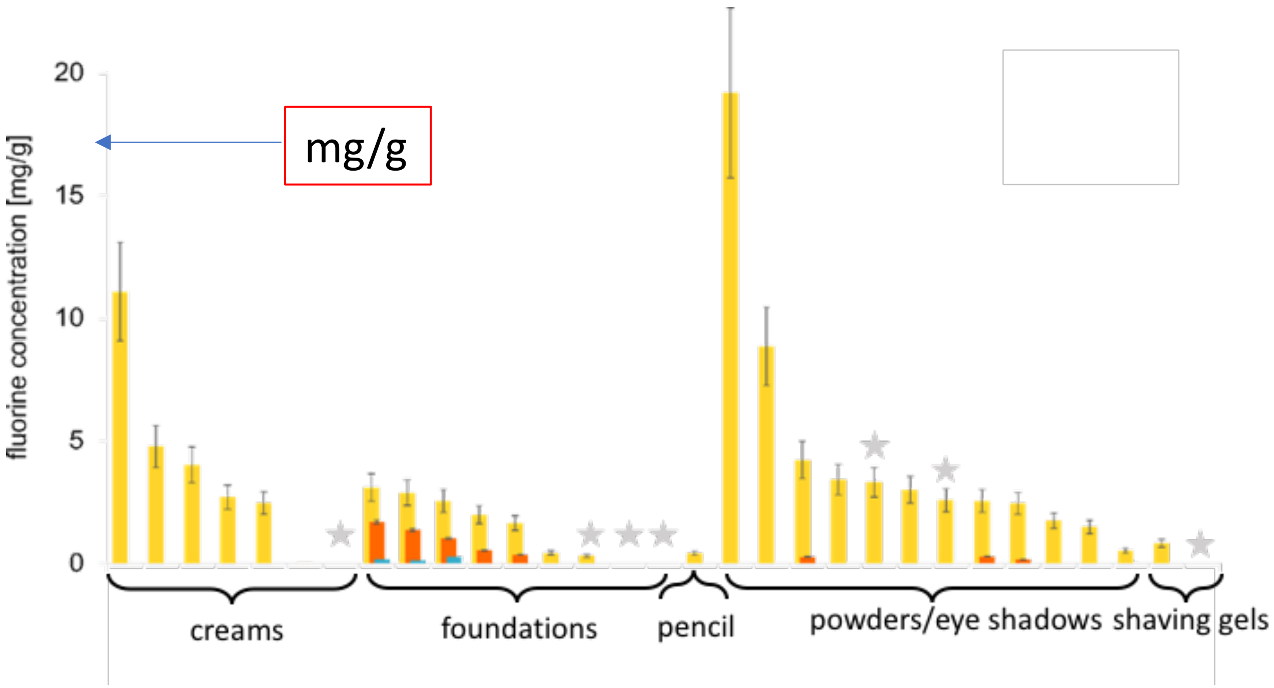
How Do We Measure Poly- and Perfluoroalkyl Substances (PFASs) at the Surface of Consumer Products?

Andrea K. Tokranov,[*,†] Nicole Nishizawa,[†] Carlo Alberto Amadei,[†] Jenny E. Zenobio,[‡] Heidi M. Pickard,[†] Joseph G. Allen,[§] Chad D. Vecitis,[†] and Elsie M. Sunderland[†,§]

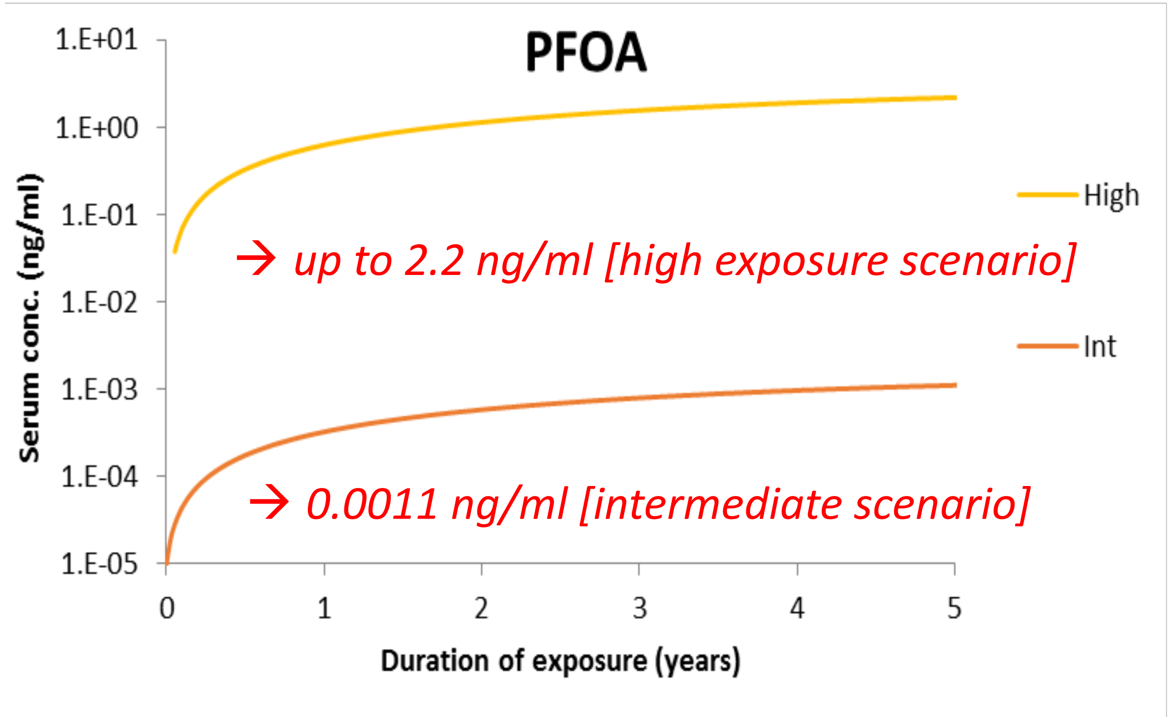
Tokranov et al., 2019, ES&T



Extremely high concentrations of fluorinated compounds measured in a variety of cosmetic products



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



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

Schultes et al., 2018

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.