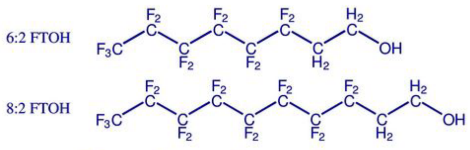
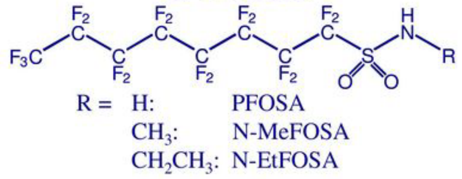
Global presence and fate of PFAS precursors

FTOHs



FOSAs



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NIH NIEHS

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More information about STEEP is available at: https://web.uri.edu/steep/ and https://tools.niehs.nih.gov/srp/programs/Program_detail.cfm?Project_ID=P42ES027706

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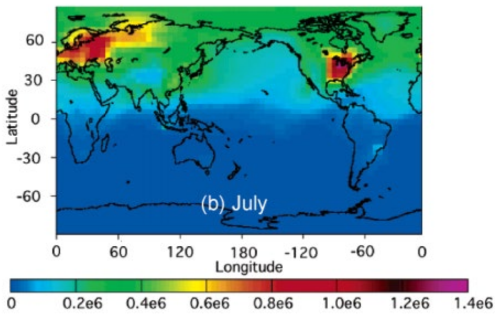
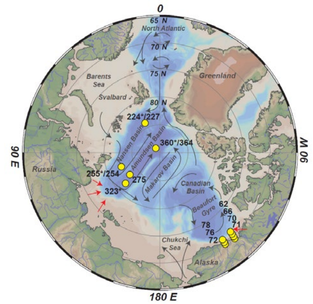
Presence of atmospheric FTOHs

- GLOBAL presence of FTOHs.

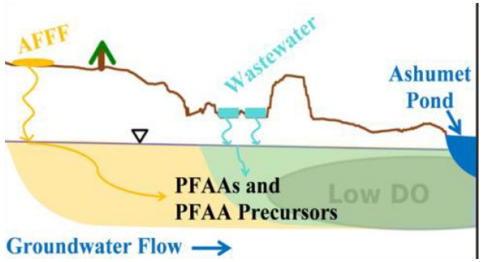


- Precursors affecting the Arctic and foodwebs



- Precursors in plants

- Microbial conversions of precursors



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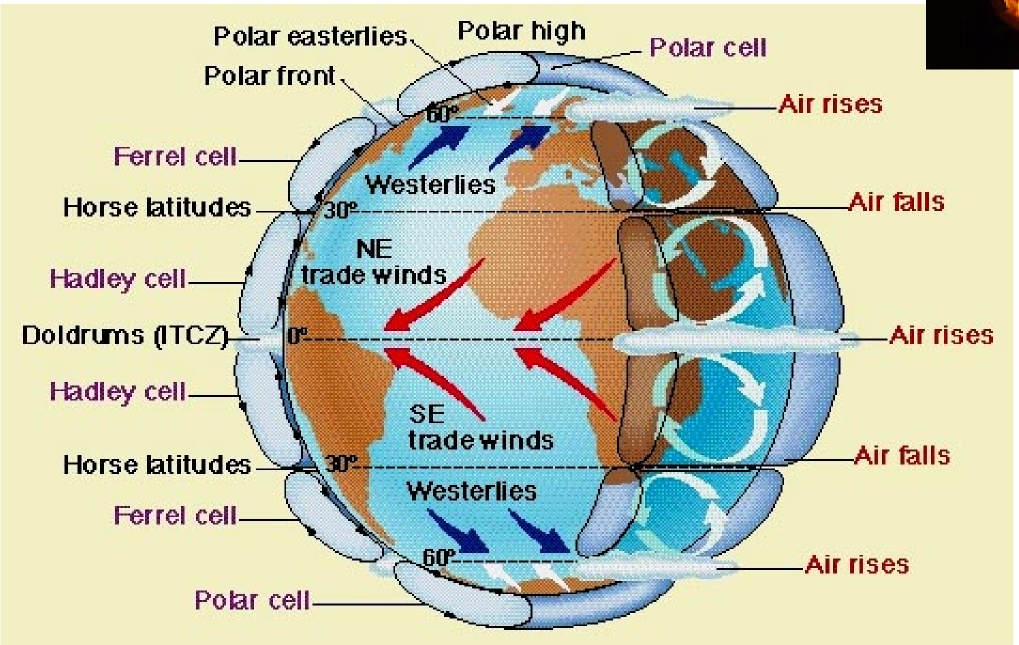
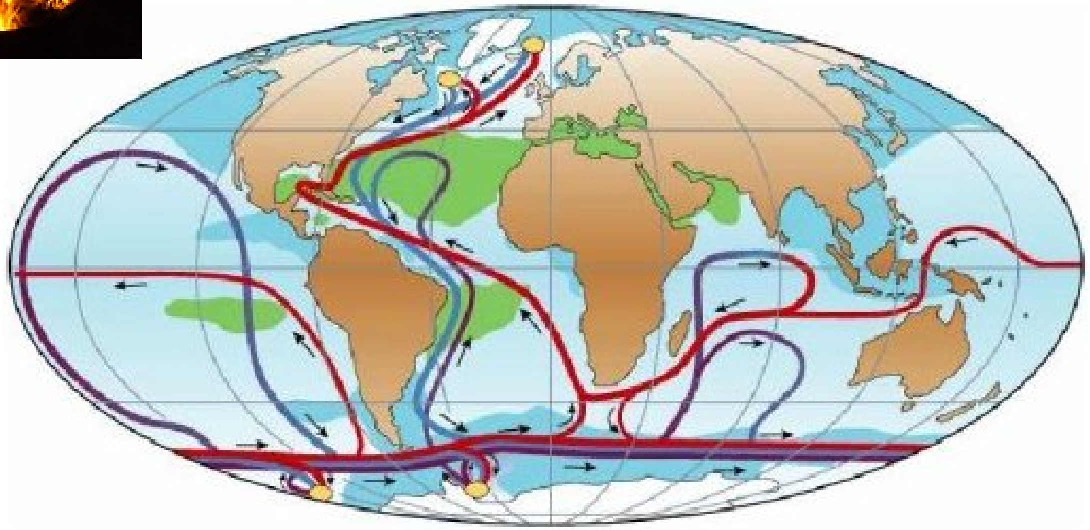
PFAS battle of the elements:

Winds vs Currents





http://www.oceansonline.com/winds.htm

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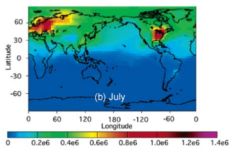
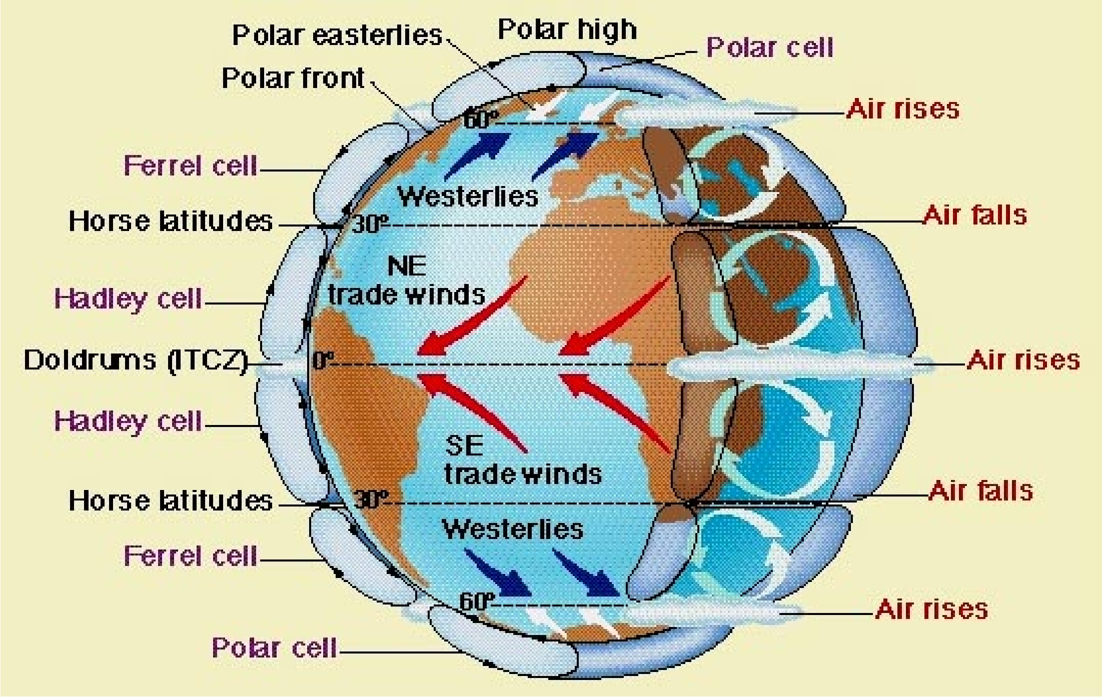
Volatile PFAS in the atmosphere: FTOHs





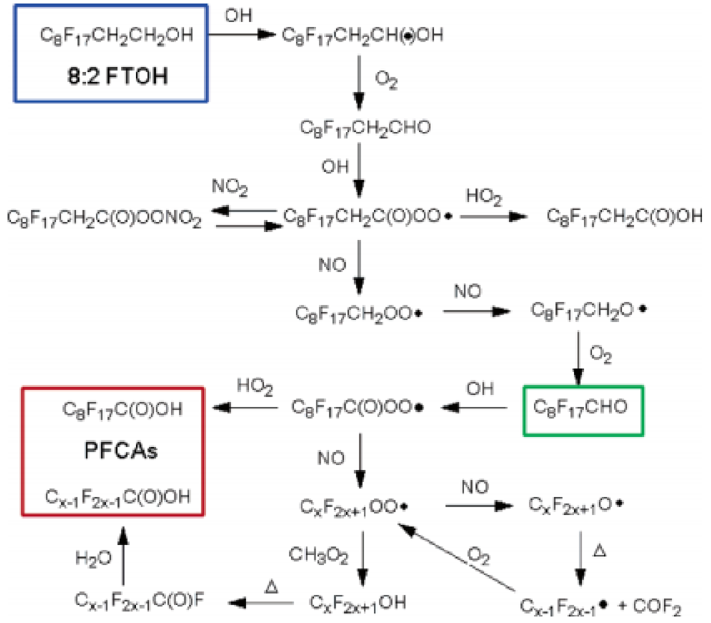


FIGURE 1. Simplified mechanism for the atmospheric degradation of 8:2 FTOH (blue box) illustrating its conversion into $C_8F_{17}CHO$ (green box) and the competition between NO and either HO_2 or CH_3O_2 radicals that limits the formation of perfluorocarboxylic acids (red box).

(Wallington et al., 2006)

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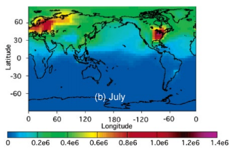

Time for radical ideas

TABLE 1. Atmospheric Oxidation Mechanism of 8:2 FTOH

reaction	rate
Atmospheric Chemistry of $C_8F_{17}CH_2CH_2OH$	
$C_8F_{17}CH_2CH_2OH + OH \rightarrow C_8F_{17}CH_2CHO$	$3.2 \times 10^{-11} \exp(-1000/T)^a$
Atmospheric Chemistry of $C_8F_{17}CH_2CHO$	
$C_8F_{17}CH_2CHO + OH \rightarrow C_8F_{17}CH_2C(O)OO$	$1.0 \times 10^{-10} \exp(-1000/T)^a$
$C_8F_{17}CH_2C(O)OO + NO \rightarrow C_8F_{17}CHO$	$8.1 \times 10^{-12} \exp(270/T)^a$
$C_8F_{17}CH_2C(O)OO + NO_2 \rightarrow C_8F_{17}CH_2C(O)OONO_2$	$1.1 \times 10^{-11} (T/298)^{-1.0\ a}$
$C_8F_{17}CH_2C(O)OONO_2 \rightarrow C_8F_{17}CH_2C(O)OO + NO_2$	$2.8 \times 10^{16} \exp(-13580/T)^b$
$C_8F_{17}CH_2C(O)OO + HO_2 \rightarrow$ products	$0.6 \times 4.3 \times 10^{-13} \exp(1040/T)^a$
$C_8F_{17}CH_2C(O)OO + HO_2 \rightarrow C_8F_{17}CHO + CO_2$	$0.4 \times 4.3 \times 10^{-13} \exp(1040/T)^a$
Atmospheric Chemistry of $C_8F_{17}CHO$	
$C_8F_{17}CHO + OH \rightarrow C_8F_{17}C(O)OO$	$1.7 \times 10^{-11} \exp(-1000/T)^a$
$C_8F_{17}CHO + hv \rightarrow C_8F_{17}O_2$	$\phi = 0.02$
$C_8F_{17}C(O)OO + NO \rightarrow C_8F_{17}O_2$	$8.1 \times 10^{-12} \exp(270/T)^a$
$C_8F_{17}C(O)OO + NO_2 \rightarrow C_8F_{17}C(O)OONO_2$	$1.1 \times 10^{-11} (T/298)^{-1.0\ a}$
$C_8F_{17}C(O)OONO_2 \rightarrow C_8F_{17}C(O)OO + NO_2$	$2.8 \times 10^{16} \exp(-13580/T)^b$
$C_8F_{17}C(O)OO + HO_2 \rightarrow C_8F_{17}COOH(PFNA^c) + O_3$	$0.10 \times 4.3 \times 10^{-13} \exp(1040/T)^a$
$C_8F_{17}C(O)OO + HO_2 \rightarrow C_8F_{17}O_2$	$0.90 \times 4.3 \times 10^{-13} \exp(1040/T)^a$
Atmospheric Chemistry of $C_8F_{17}O_2$	
$C_8F_{17}O_2 + NO \rightarrow C_8F_{17}O + NO_2$	$2.8 \times 10^{-12} \exp(300/T)^a$
$C_8F_{17}O_2 + HO_2 \rightarrow C_8F_{17}O + OH + O_2$	$4.1 \times 10^{-13} \exp(750/T)^a$
$C_8F_{17}O_2 + CH_3O_2 \rightarrow C_8F_{17}O + CH_3O$	$2.7 \times 10^{-12} \exp(-470/T)^a$
$C_8F_{17}O_2 + CH_3O_2 \rightarrow C_8F_{17}OH + HCHO$	$1.0 \times 10^{-13} \exp(660/T)^a$
$C_8F_{17}OH \rightarrow C_7F_{15}COOH(PFOA^d)$	$2.3 \times 10^{-6\ b}$

[a] Units of cm^3 $molecule^{-1}$ s^{-1}. [b] Units of s^{-1}. [c] PFNA = perfluorononanoic acid = $C_8F_{17}COOH$. [d] PFOA = perfluorooctanoic acid = $C_7F_{15}COOH$.

(Wallington et al., 2006)

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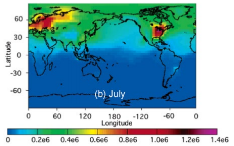
The spread of FTOHs across the atmosphere



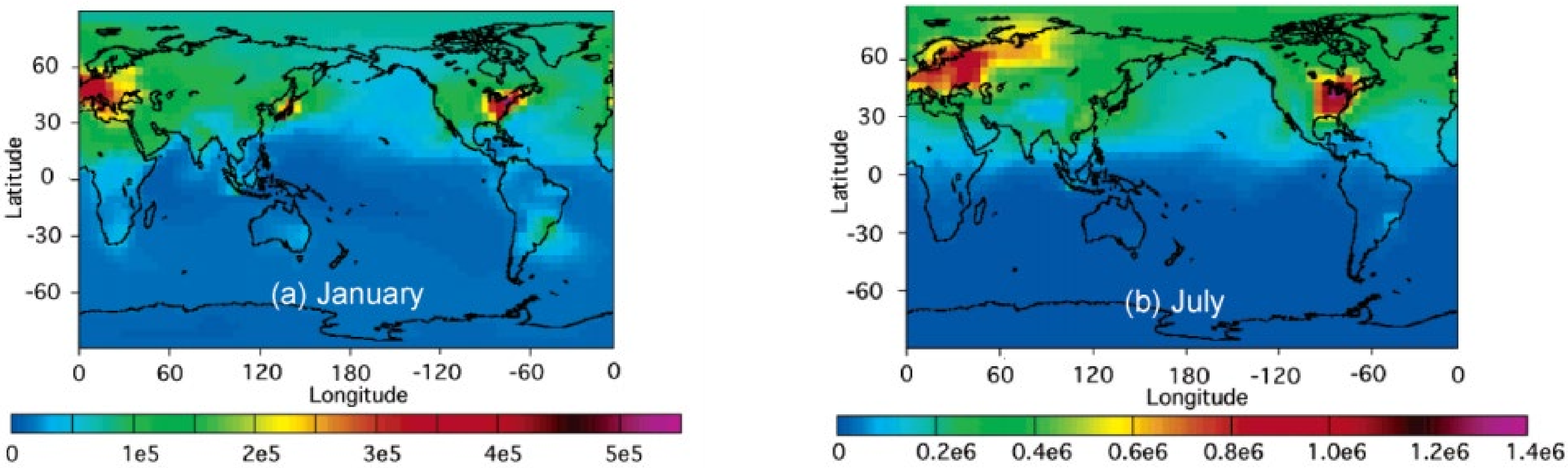


FIGURE 2. Summed concentration of 8:2 FTOH and all of its degradation products at 50 m in altitude for (a) January and (b) July. The color scale extends from (a) 0 to 5.5 × 10^5 molecule cm^{-2} and (b) 0 to 1.4 × 10^5 molecule cm^{-3}.

- Yield of 3-6% PFOA from 8:2 FT-OH; also smaller PFCAs
- Source: Arctic 5:1
- t_{life} ~ 20-40 days

(Wallington et al., 2006)

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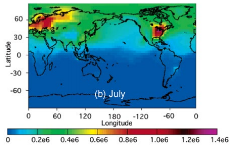
Toronto: Arctic 5:1, first set



Toronto: Costa Rica: Botswana 10:5:1

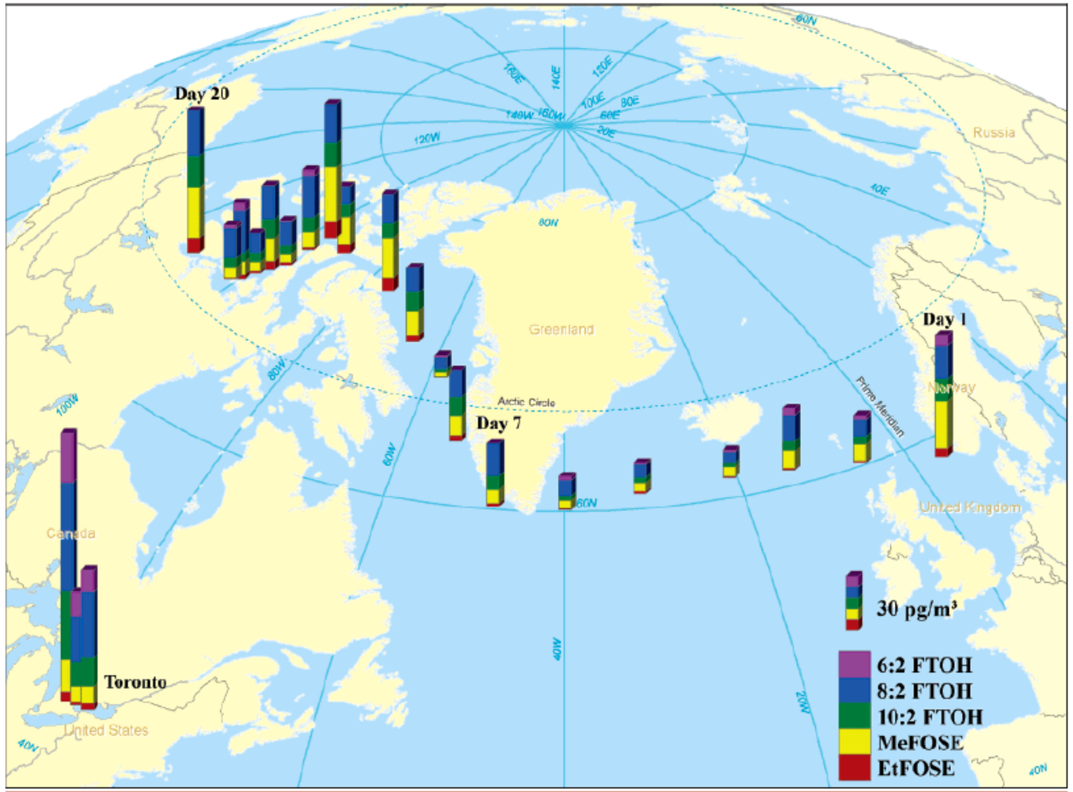
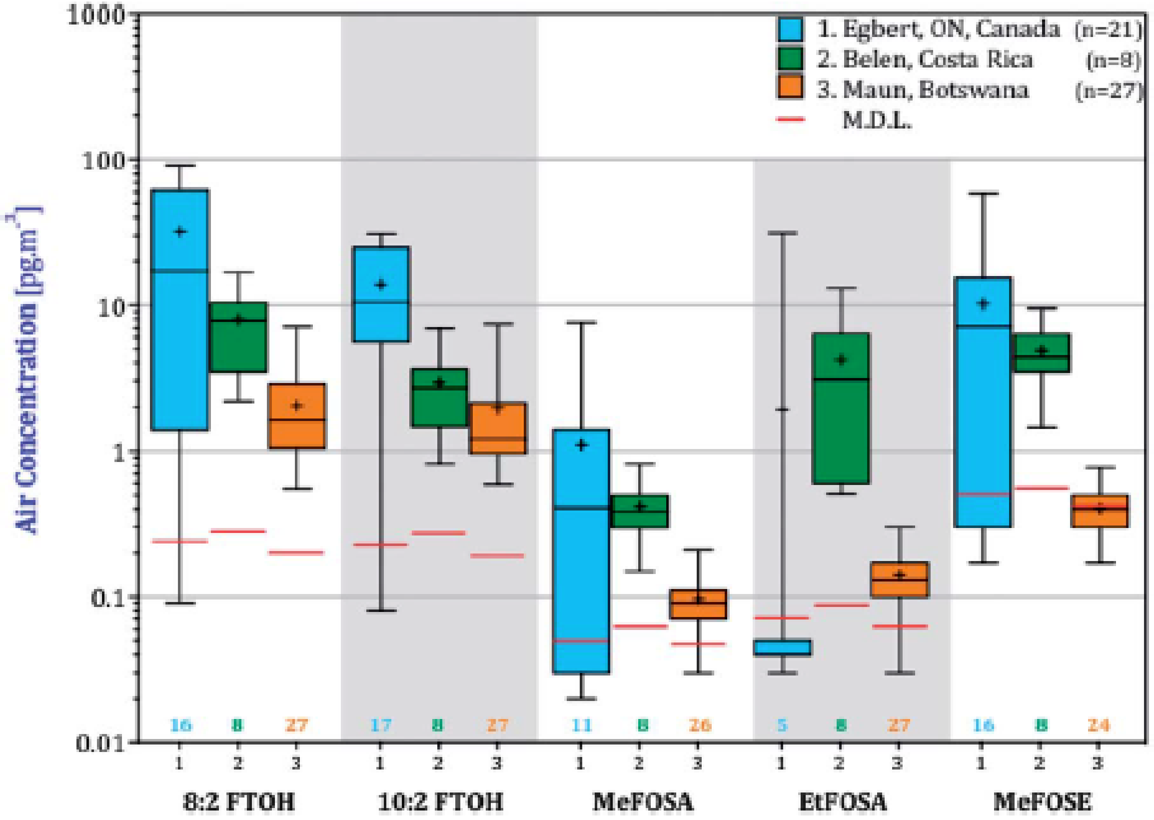


FIGURE 2. Total air concentrations (sum of gas phase and particle phase) for FTOHs and PFASs across the North Atlantic Ocean a Canadian Archipelago (see Figure 1 cruise track) and in Toronto, Canada.

(Shoeib et al., 2006)



(Gawor et al., 2014)

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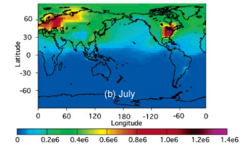
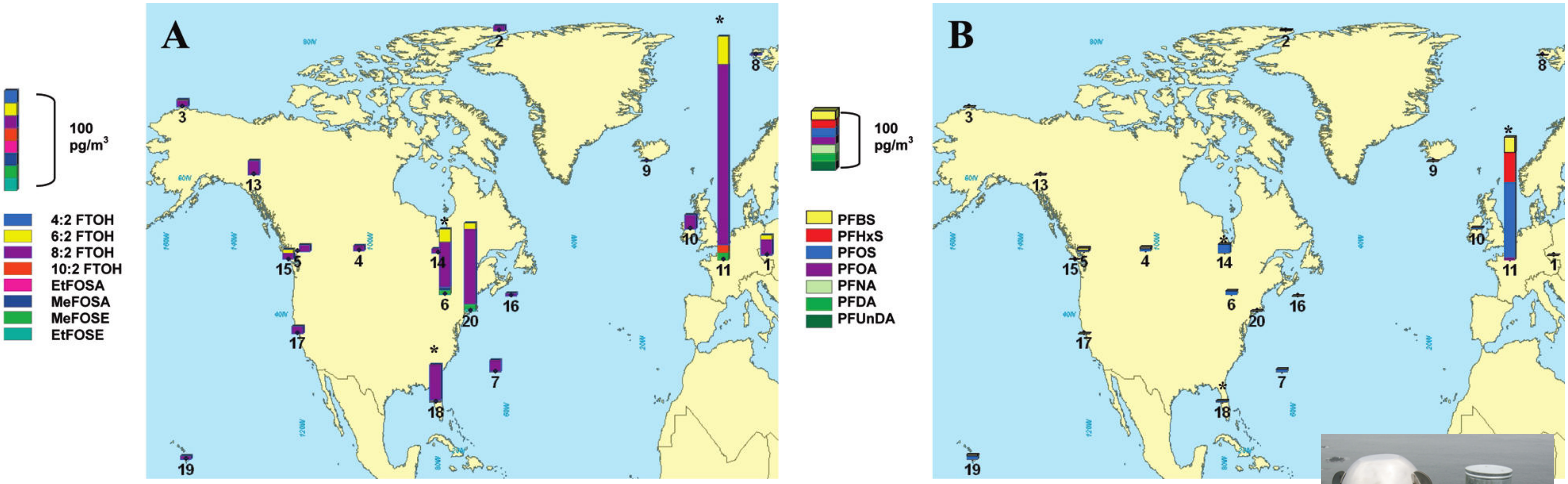
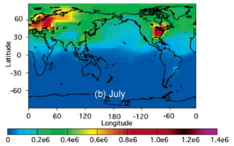
Some sites are cleaner





(Genualdi et al., 2010)

All clean down south?



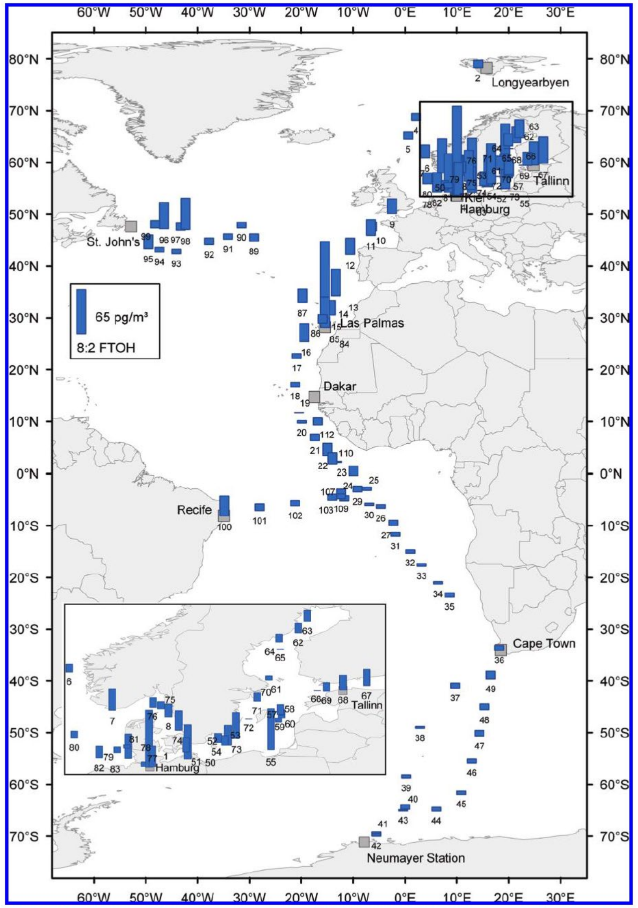


FIGURE 1. Spatial distribution of 8:2 FTOH gas-phase concentrations determined during several cruises in the Baltic Sea and the Atlantic and Southern Oceans. Note that the close-up of the Baltic Sea region is not to scale.

(Dreyer et al., 2009)

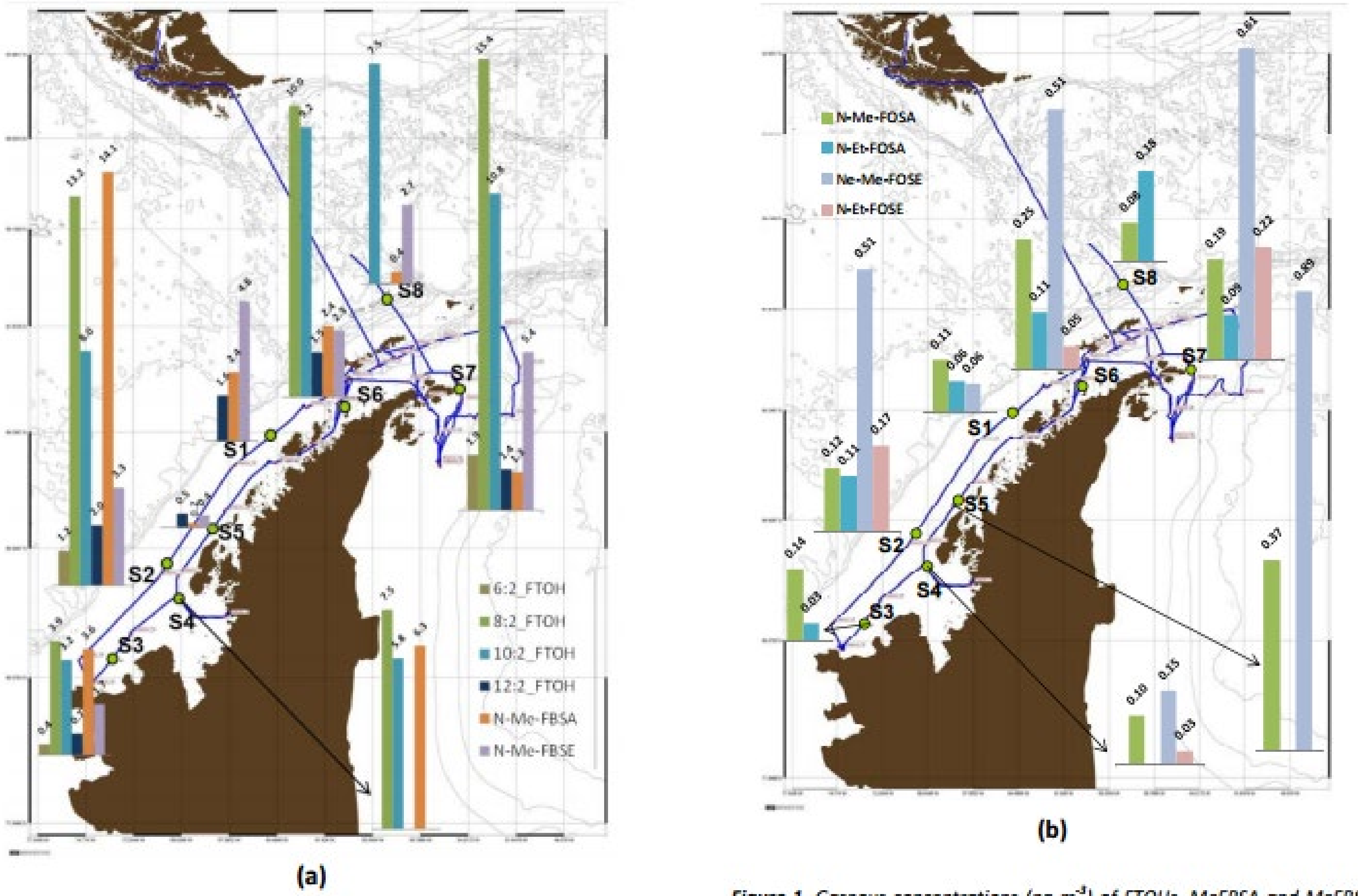


Figure 1. Gaseous concentrations (pg m⁻³) of FTOHs, MeFBSA and MeFBSE (a); FOSAs and FOSEs (b), measured during February 2009.

(Del Vento et al., 2012)

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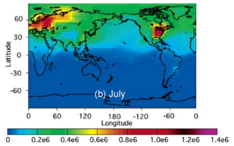
Cold condensation or fata morgana?



- Resurgence of FTOHs in Antarctic?
- Trends across Atlantic Ocean as expected
- Revolatilization of FTOHs from snow?
- Yet
  - NH 4x more land
  - NH 9 x more people

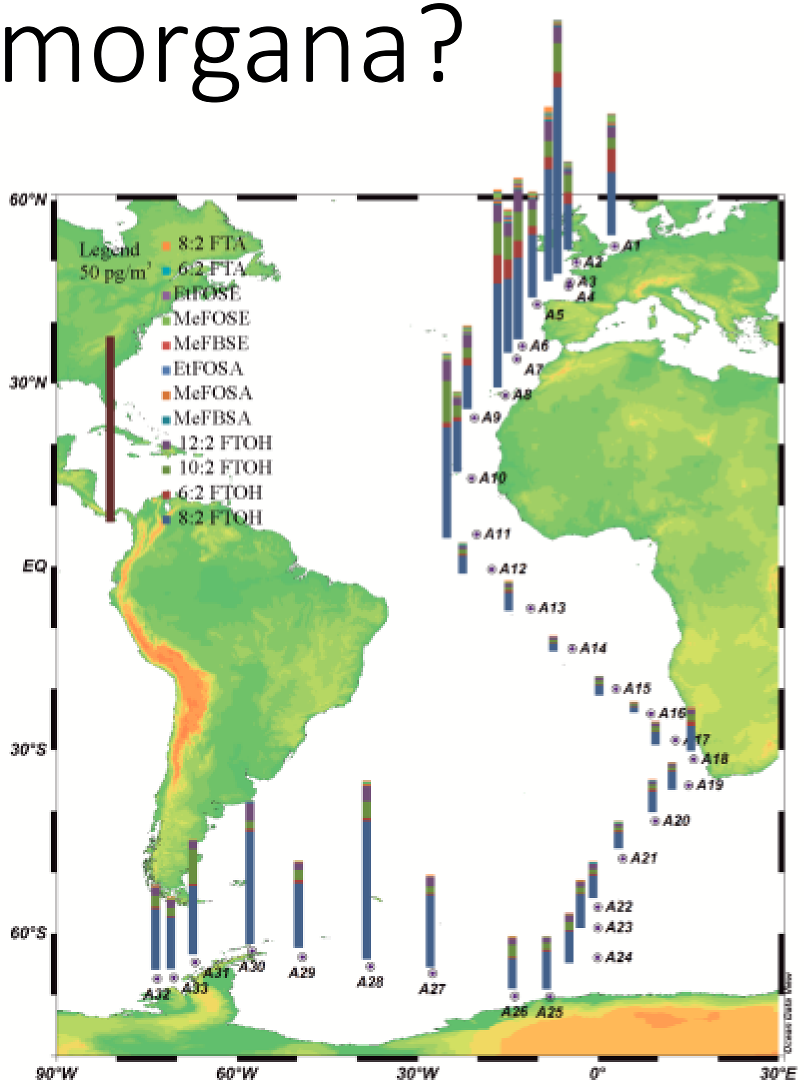


Figure 1. High volume air sampling sites during the cruise of the R/V *Polarstern* in ANT-XXII from October 2010 to January 2011 from Germany to the Antarctic, and concentrations (pg/m³), composition of individual PFASs in the marine atmosphere.

(Wang et al., 2015)

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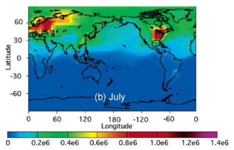
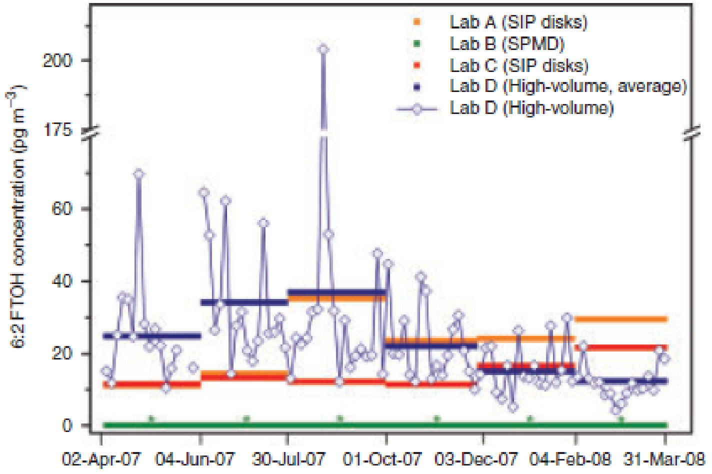
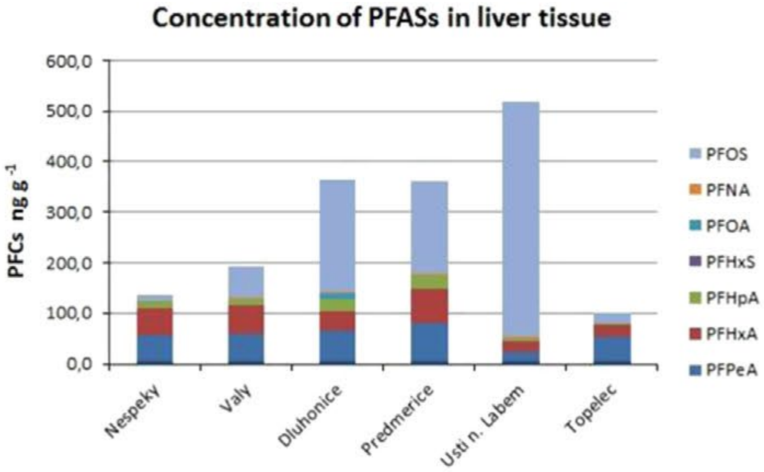


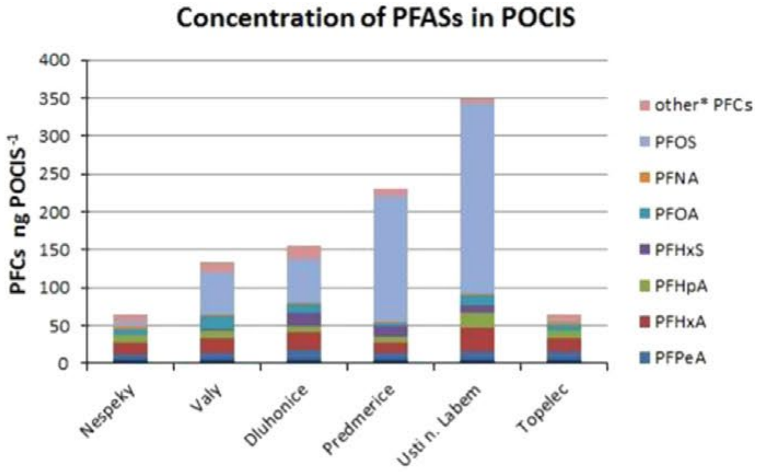
Why passives might be useful

- Time-integrated concentrations
- Pre-concentrations already in field
- Less handling in laboratory
- Proxy for bioaccumulation



(Dreyer et al., 2010)





(Cerveny et al., 2016)

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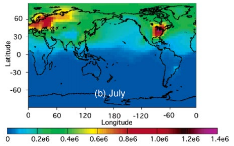
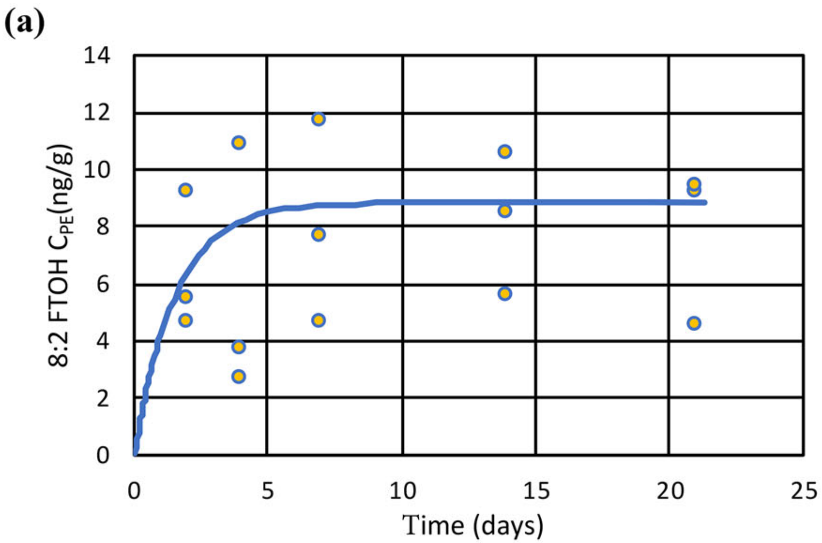
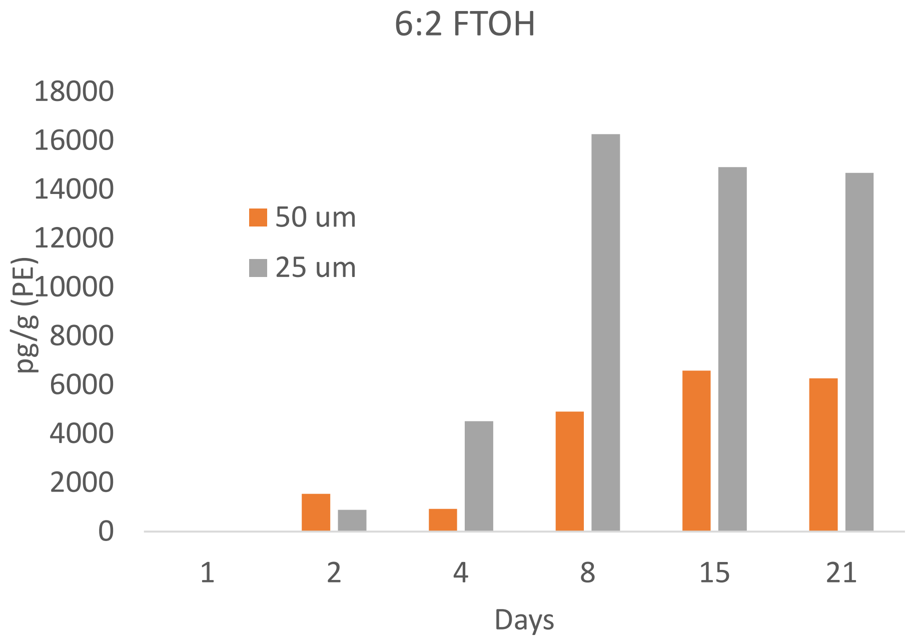
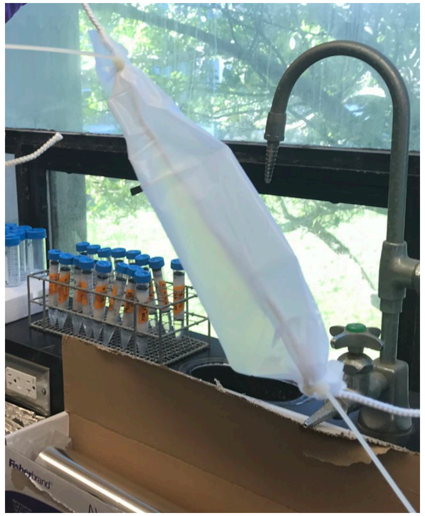


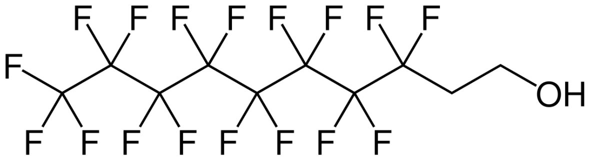
Testing of Polyethylene Sheets as Passive Samplers for Volatile PFAS in Indoor Air

Maya Morales-McDevitt et al.









(Dixon-Anderson and Lohmann, 2018)

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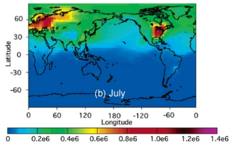

Plenty of other precursors out there (AFFF)

Table 2. Product FTOH Concentrations and Nontargeted Chemical Compositions

	targeted (ng μL^{-1})				nontargeted		
product	4:2 FTOH	6:2 FTOH	8:2 FTOH	10:2 FTOH	composition	peak center m/Q	mass error (ppm)
Masurf FS-115	26.1	578.5	106.8	43.0	$C_6F_{10}H_2O$	279.996	5.0
					$C_7F_{13}H_3O$	349.994	10.4
					$C_{16}F_{26}H_8O$	710.009	9.9
					$C_{18}F_{30}H_8O$	810.002	9.4
					$C_{20}F_{34}H_8O$	909.988	16.7
Zonyl FSA	9.4	948.1	130.7	17.9	$C_{14}F_{23}H_5O$	626.002	7.5
					$C_{16}F_{27}H_5O$	725.999	11.1
					$C_{18}F_{31}H_5O$	825.997	14.8
Capstone FS-35	6.7	644.6	–	–	$C_4F_5H_5O$	164.025	6.4
					$C_5F_7H_5O$	214.023	0.6
					$C_7F_{11}H_5O$	314.015	4.7
					$C_{14}F_{24}H_6O$	645.996	11.7
Arctic 3 AFFF	–	1.6	0.3	–	$C_7F_{11}H_5O$	314.018	4.9
					$C_{16}F_{27}H_5O$	726.002	15.3
					$C_{18}F_{31}H_5O$	825.986	1.8

(Riedel et al, 2019)

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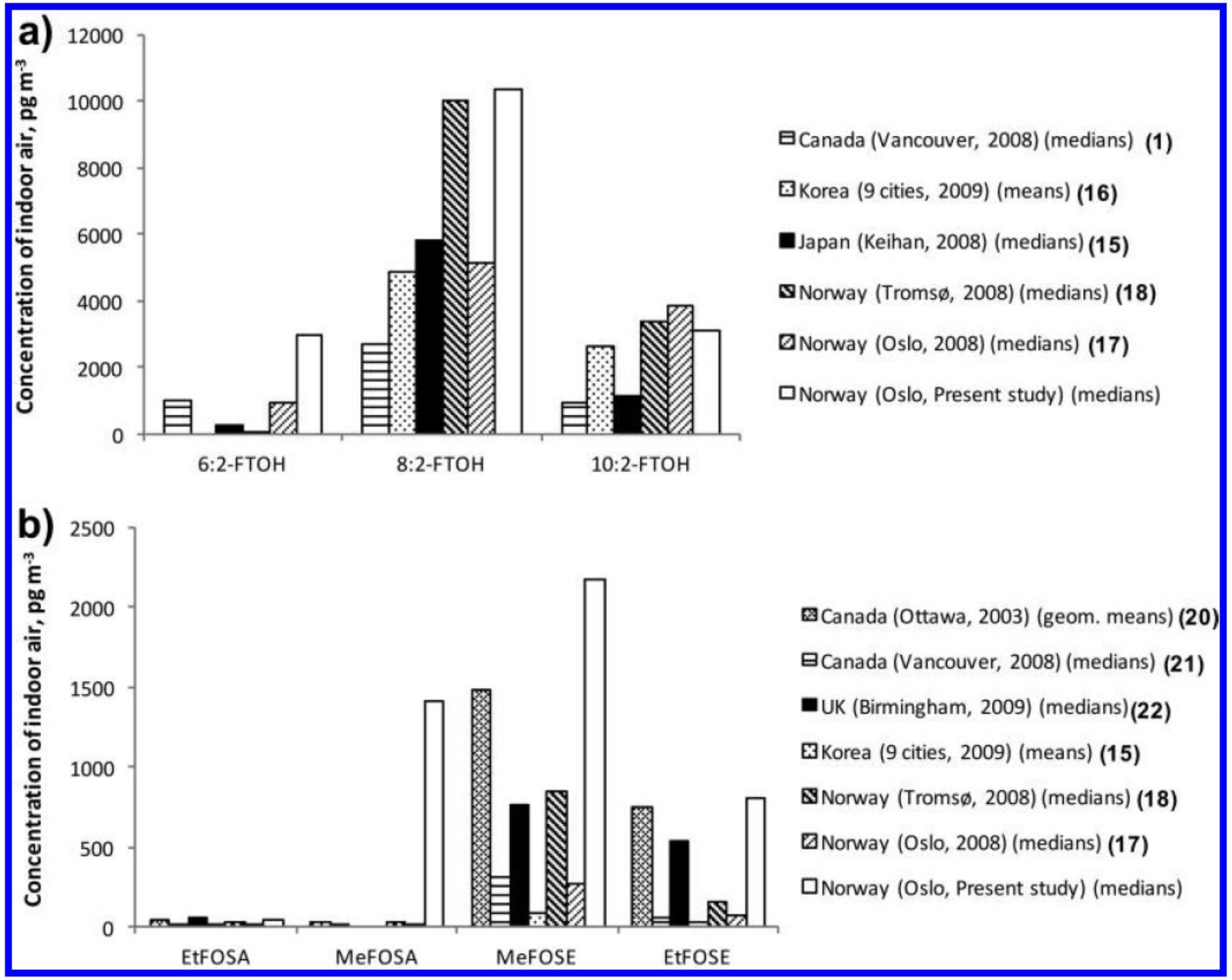
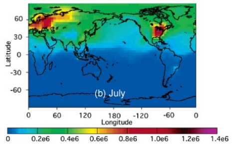
Plenty of other precursors in there



(Padilla-Sanchez et al, 2017)



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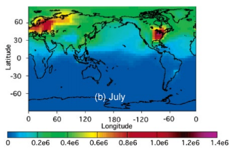
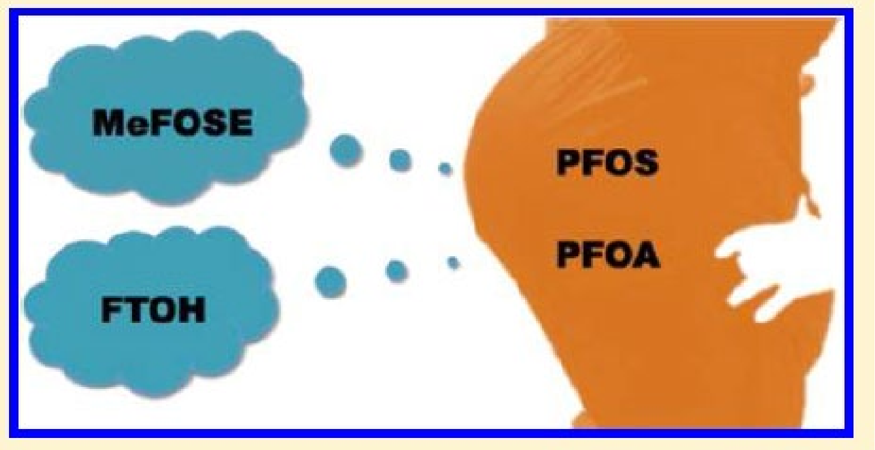
Something in the air..

… and in your blood



Environmental Science & Technology
Article
pubs.acs.org/est

Airborne Precursors Predict Maternal Serum Perfluoroalkyl Acid Concentrations



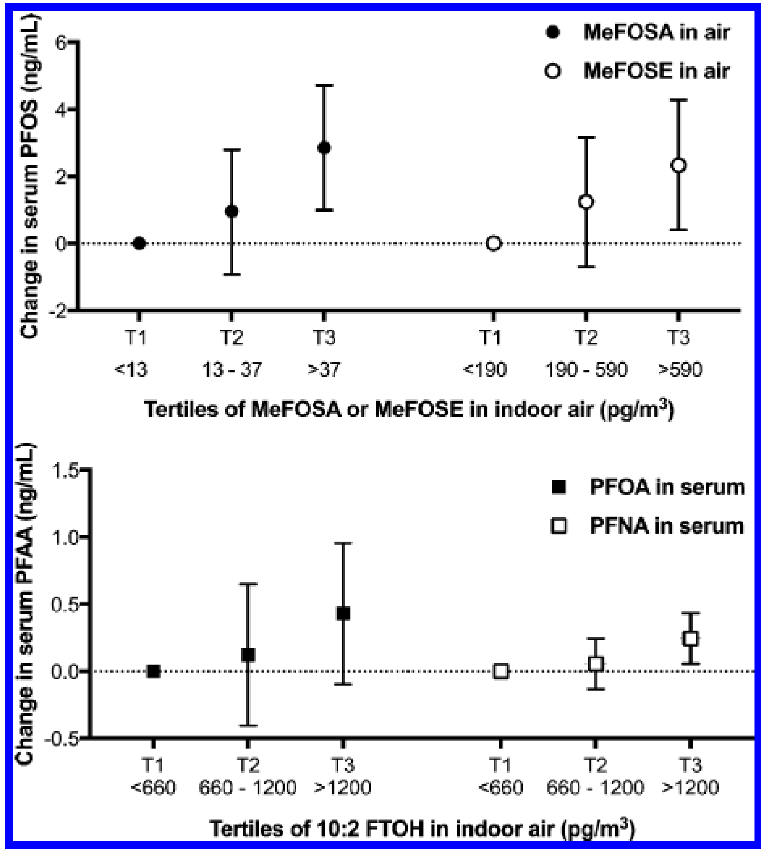


Figure 1. Exposure categories [low (n = 17), medium (n = 16), and high (n = 16)] of PreFAA in air predicting serum PFAA levels in 50 women.

(Makey et al. 2017)

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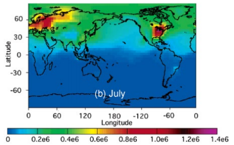

Global presence –FTOHs

- Different campaigns – routine detection in NH;
- Gradients from source to sink regions
- Much stronger gradients indoor vs outdoor
- Passive samplers as 1 tool for routine detection
- Presence of long-chain acids in humans (pre-cursors)?

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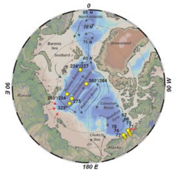

Precursors into the Arctic

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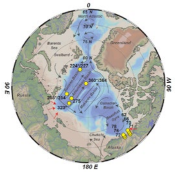
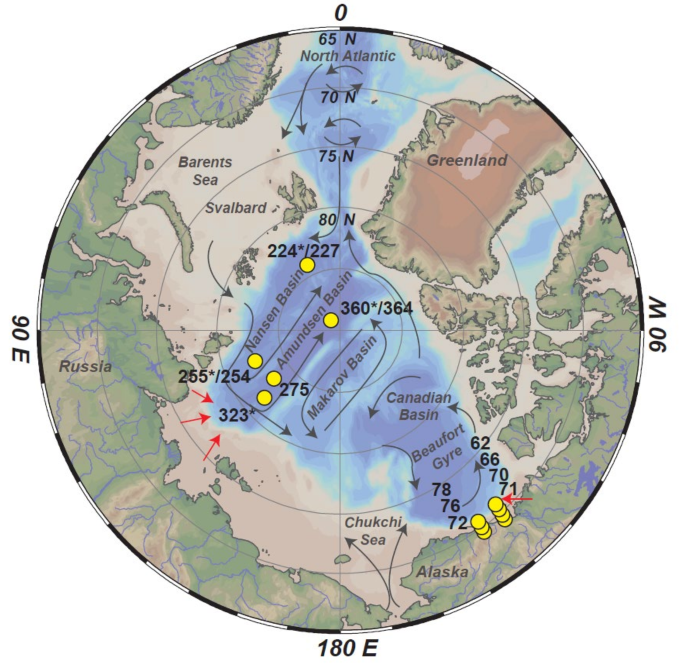
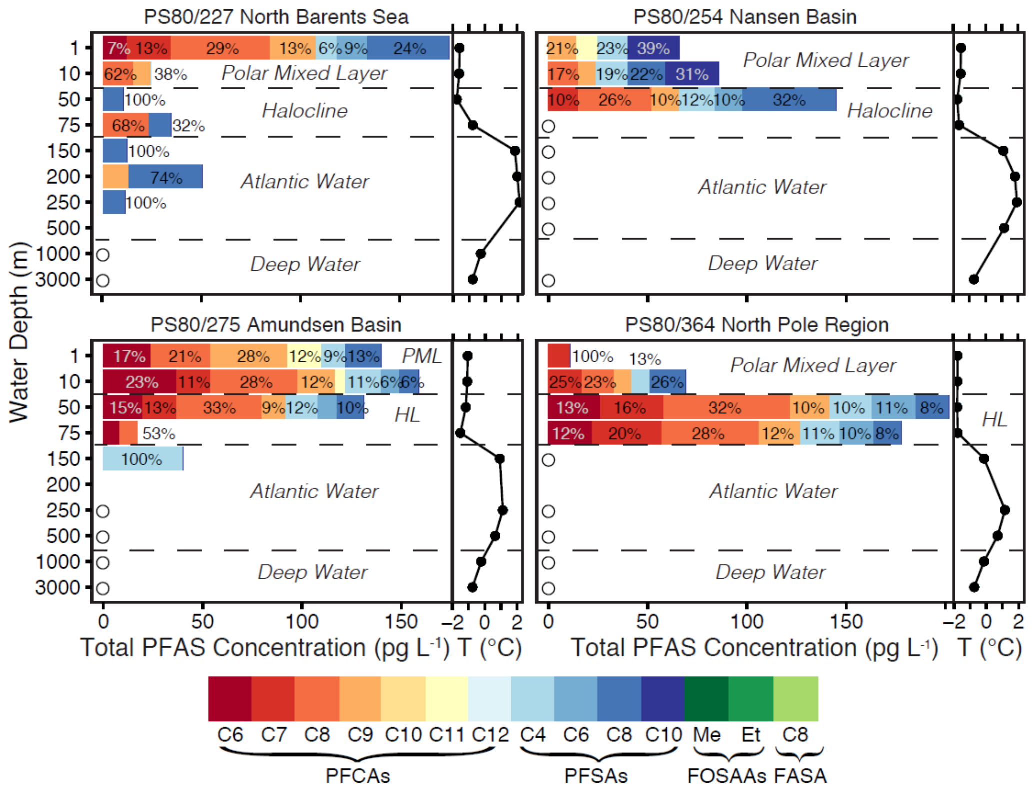
First signs of Atlantic PFASs?







(Yeung et al., EST 2017)

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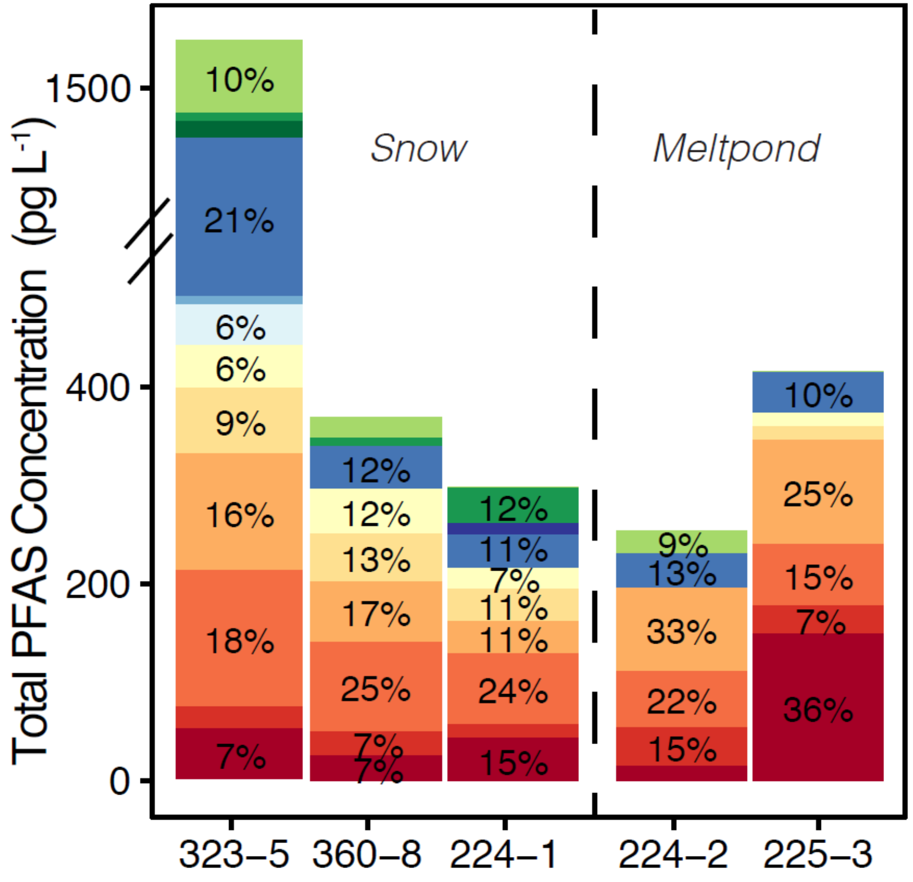
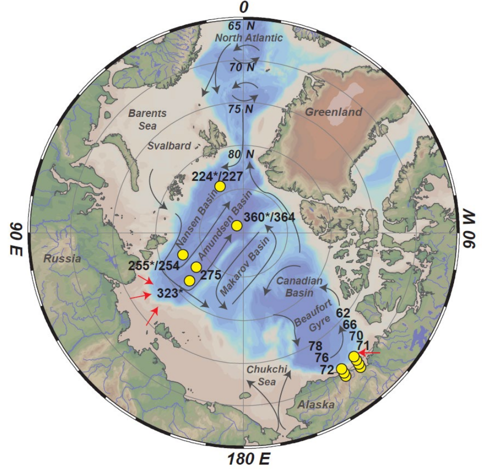
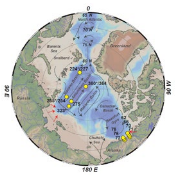
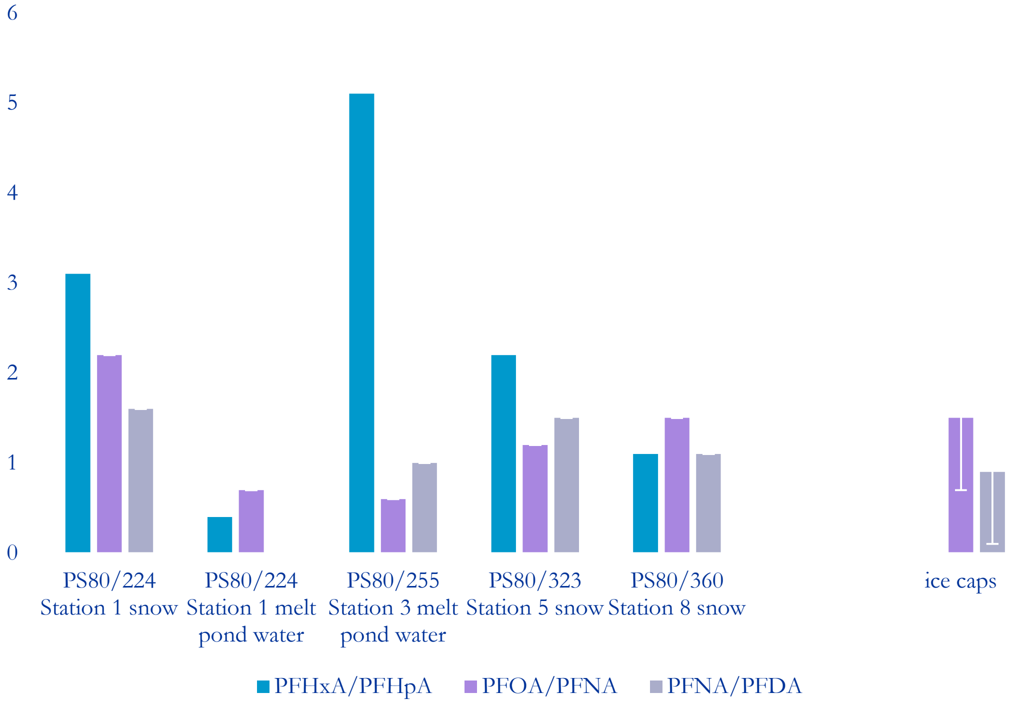
Origin of PFASs on ice?

- Similar ratios on ice and in snow: deposition!



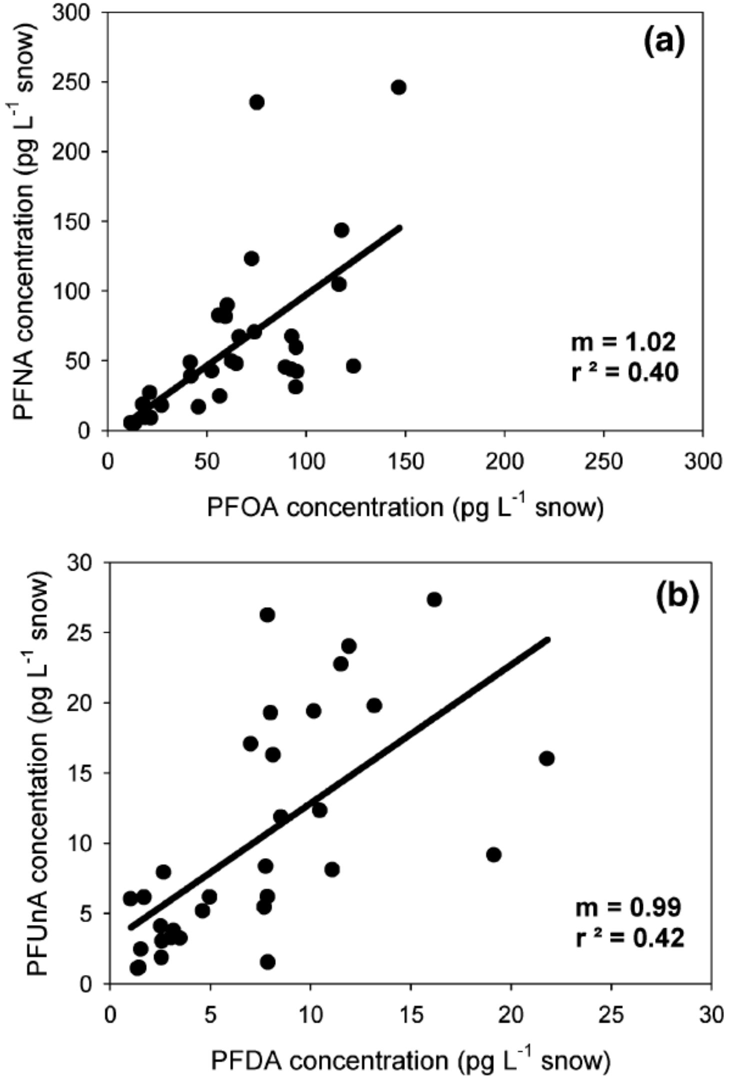






(Young et al., EST 2007)

PFASs in Devon Ice Cap



- Only atmospheric input
- Little from sea spray

- Oxidation of 8:2 FTOH

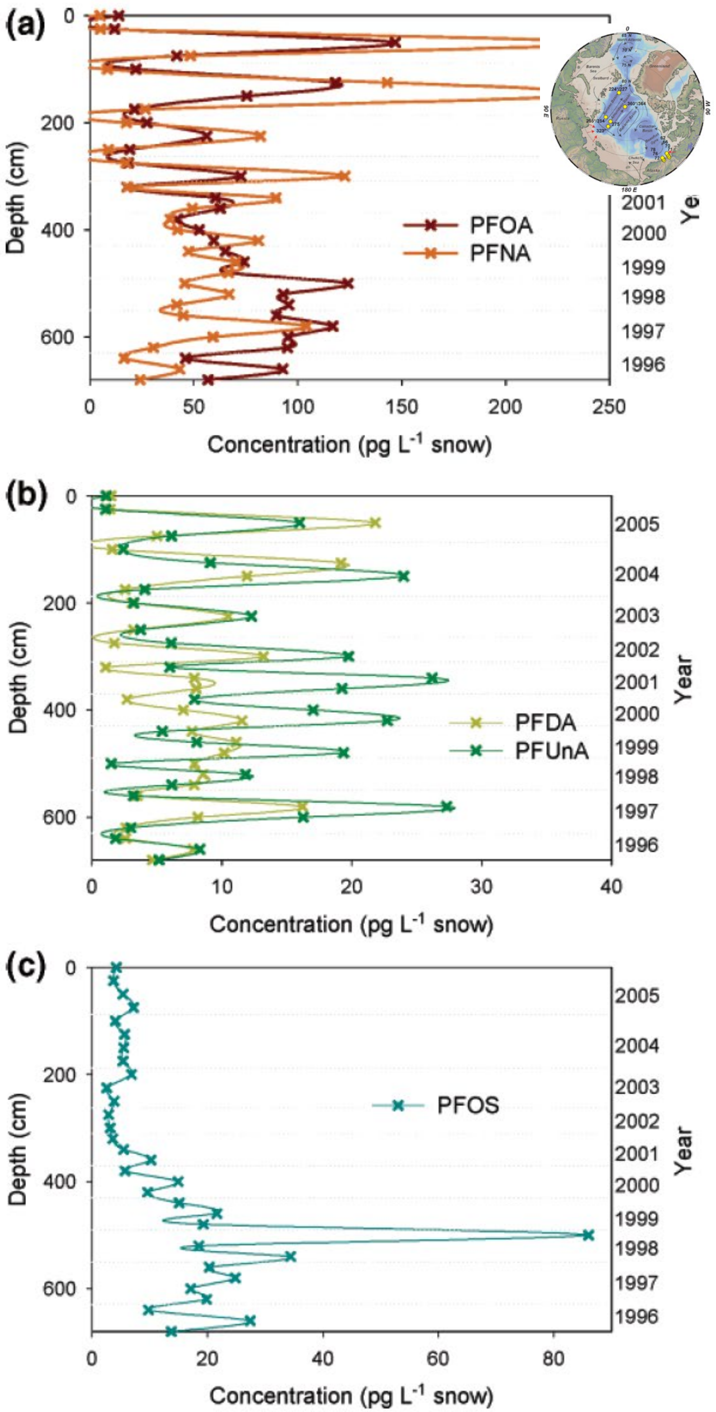
-> PFOA, PFNA

- Oxidation of 10:2 FTOH

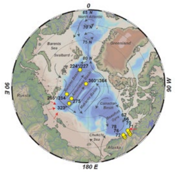
-> PFDA, PFUnDA

(Little commercial production)

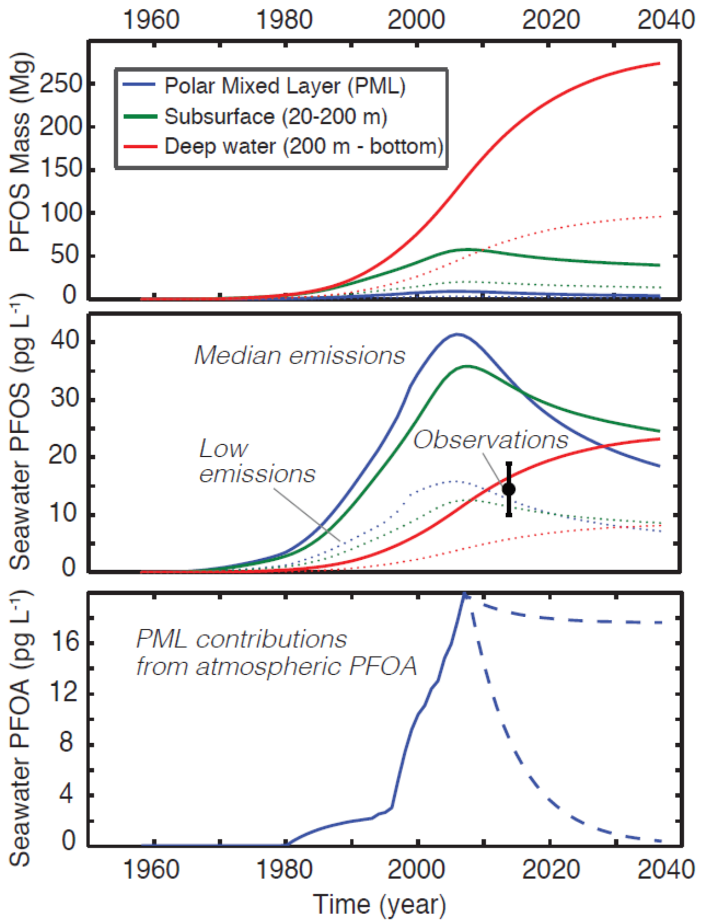
(Young et al., 2007)



Back in the Arctic



- Only traces of PFASs in deeper Arctic water (Atlantic water mass)
- Little evidence for vertical transport
- Model suggest PFASs in deeper Arctic will increase
- Both atmosphere and ocean transport
  - For PFOS up to 30% from air
  - For PFOA 30-60% from air



(Zhang et al., GBC 2017)

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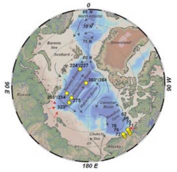
Bioaccumulation of PFASs in the Arctic



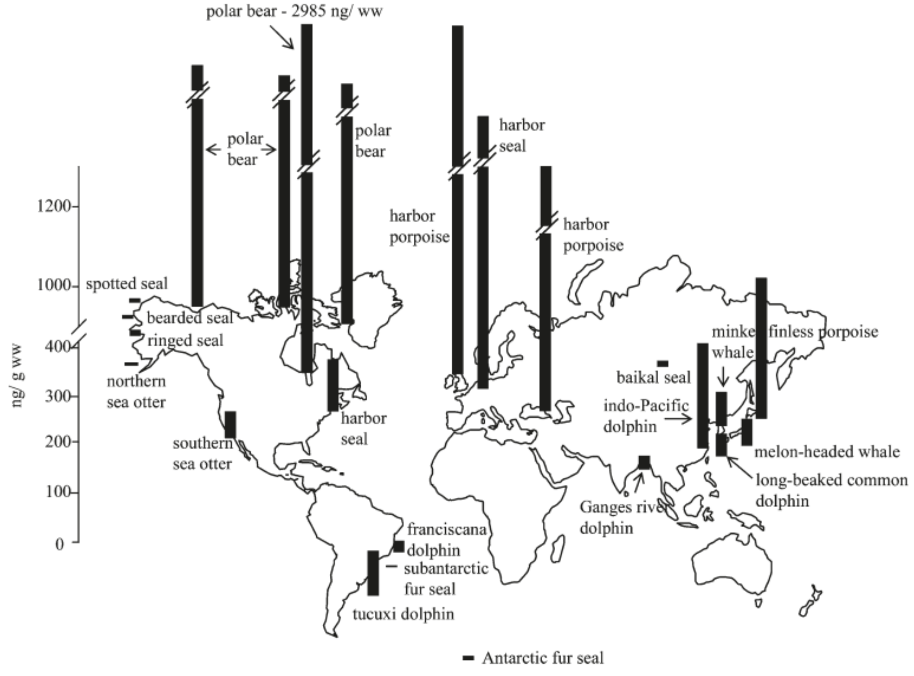
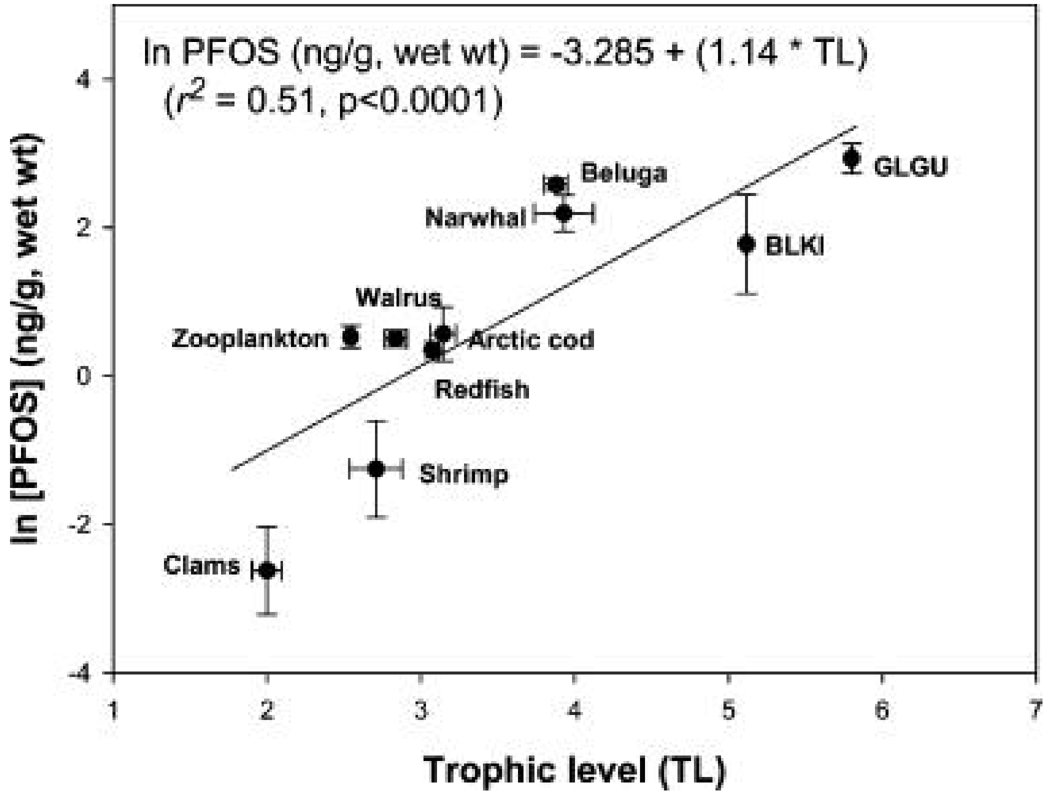


Figure 1. Recently reported (2006−2010) PFOS concentrations in liver of marine mammals worldwide.[6−8,10,12,13,15,19,21,30,36,65−67,102−104]

Monitoring of Perfluorinated Compounds in Aquatic Biota: An Updated Review

PFCs in Aquatic Biota

Magali Houde,[*,†] Amila O. De Silva,[‡] Derek C. G. Muir,[‡] and Robert J. Letcher[§]



(Tomy et al., 2004)

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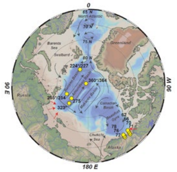
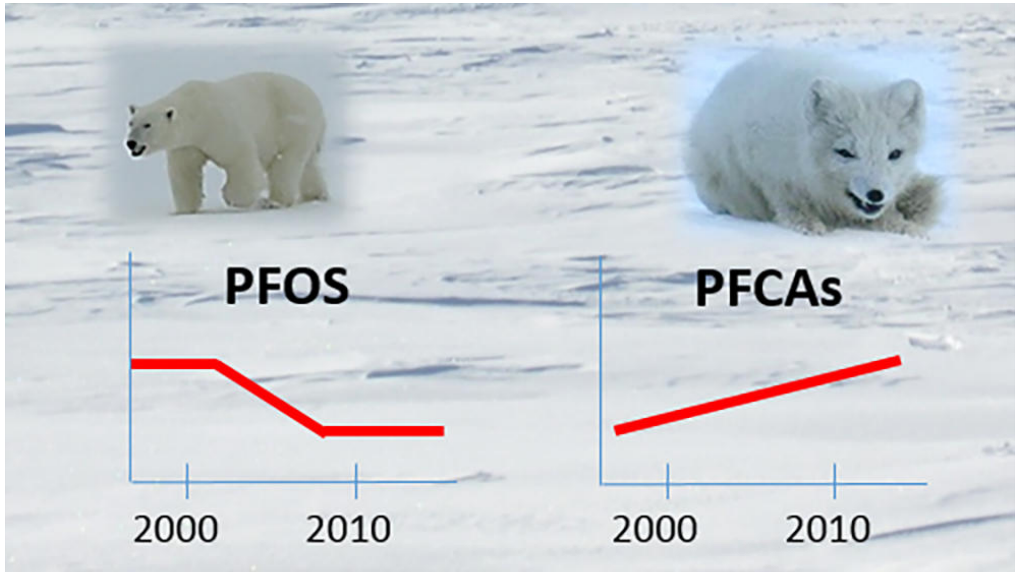
Foodweb effects in Arctic Ocean: PFOS



- Foodweb matters.

Rat~polar bear>seal>>>beluga



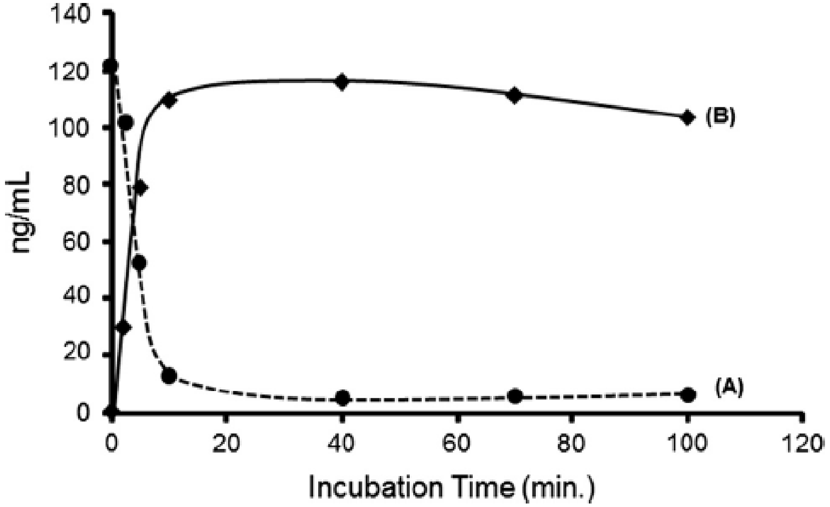


Fig. 1. Time course incubation of (A) *N*-EtFOSA depleted and (B) FOSA formed concentrations (based on *n* = 3 replicate assays for each time point) in an *in vitro* liver assay using adult male Wistar–Han rat microsomes. The initial *N*-EtFOSA concentration was 300 nM.

(Routti et al., 2017)

(Letcher et al., 2014)

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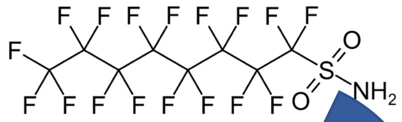
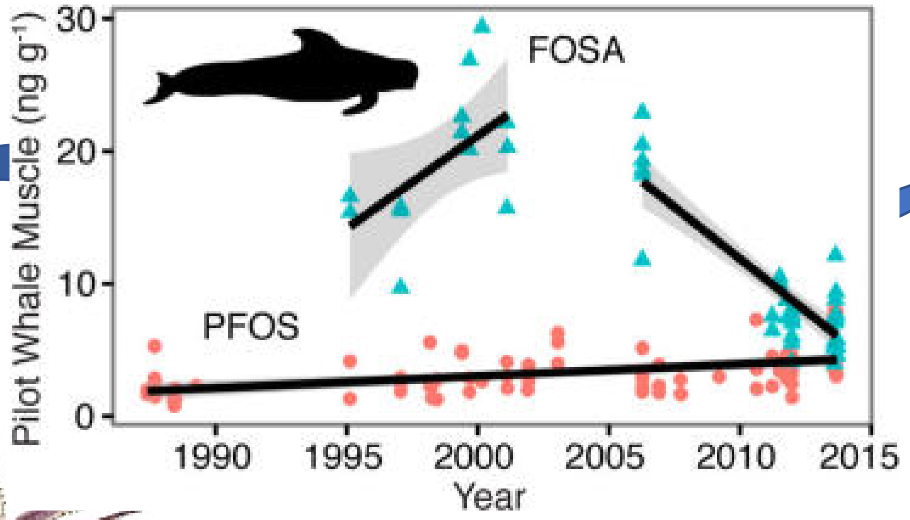
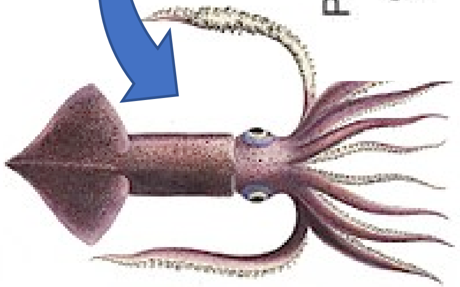
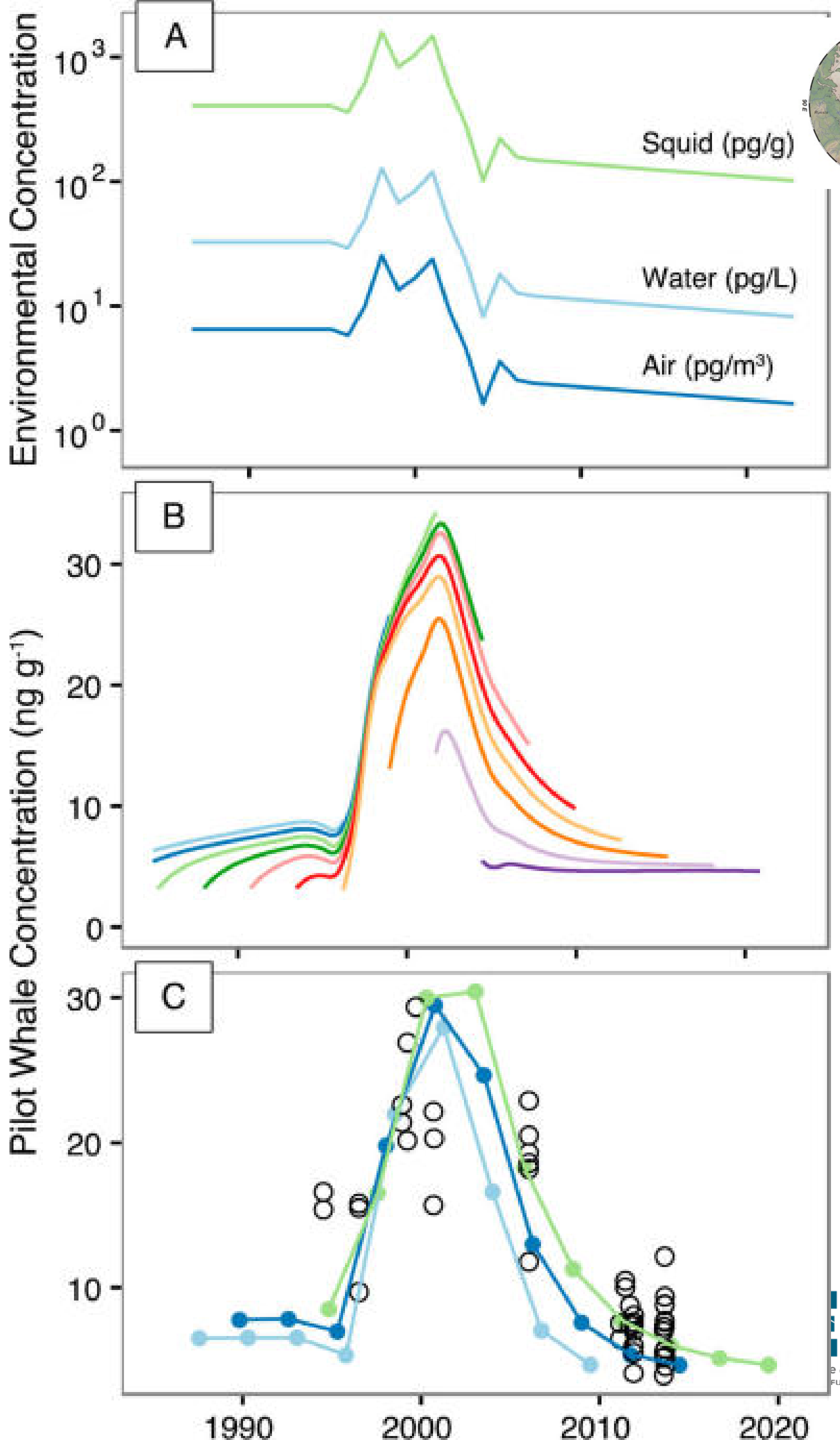
Pilot whales and FOSA

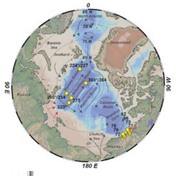
- Stay clear of metabolism







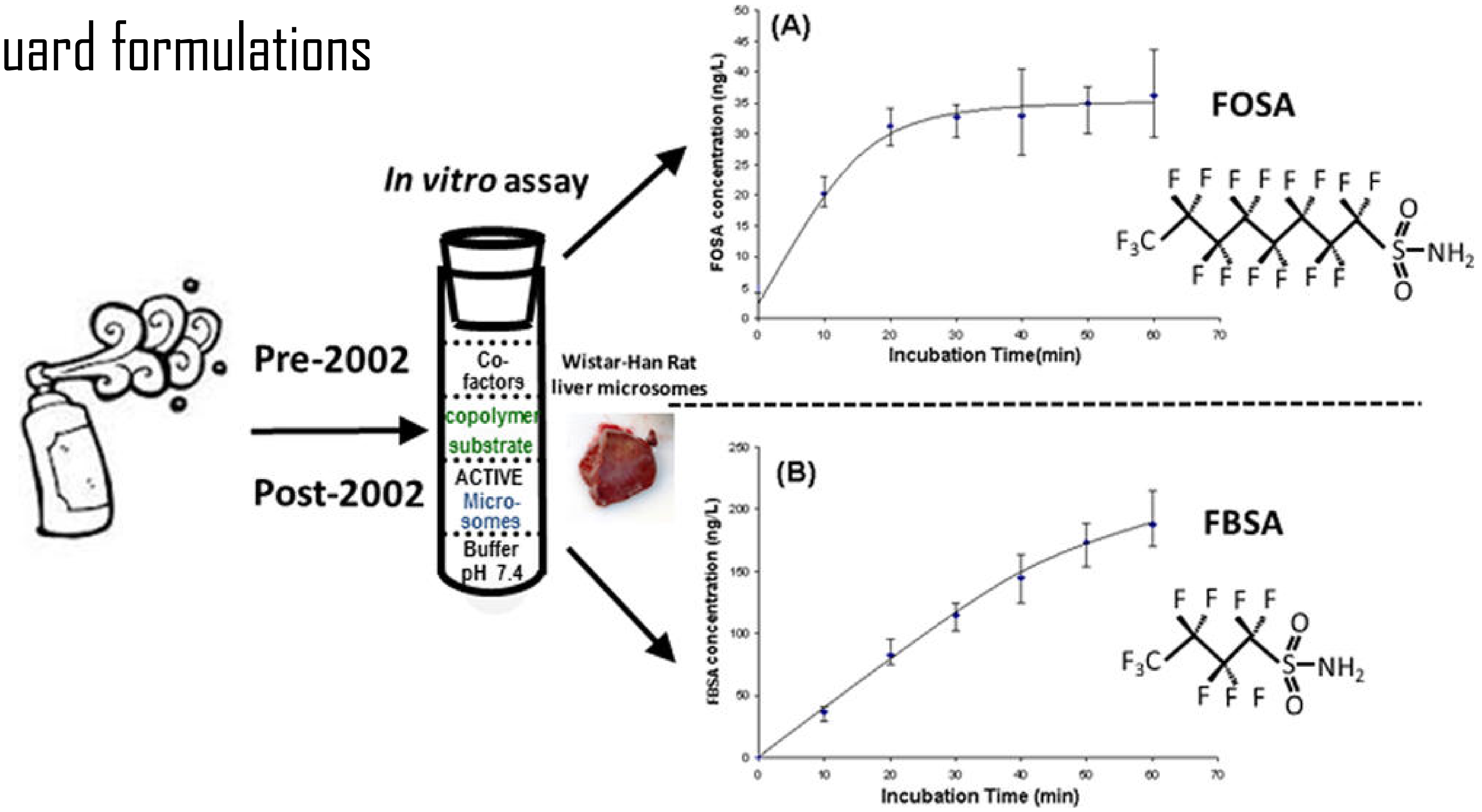




(Dassuncao et al., 2017)

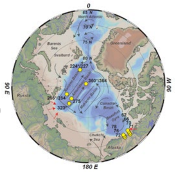
FOSA is out.. FBSA is in..

- Scotchguard formulations



(Chu and Letcher, 2014)

Fish bioaccumulation is a concern C6/ onwards



- PFAAs are well understood

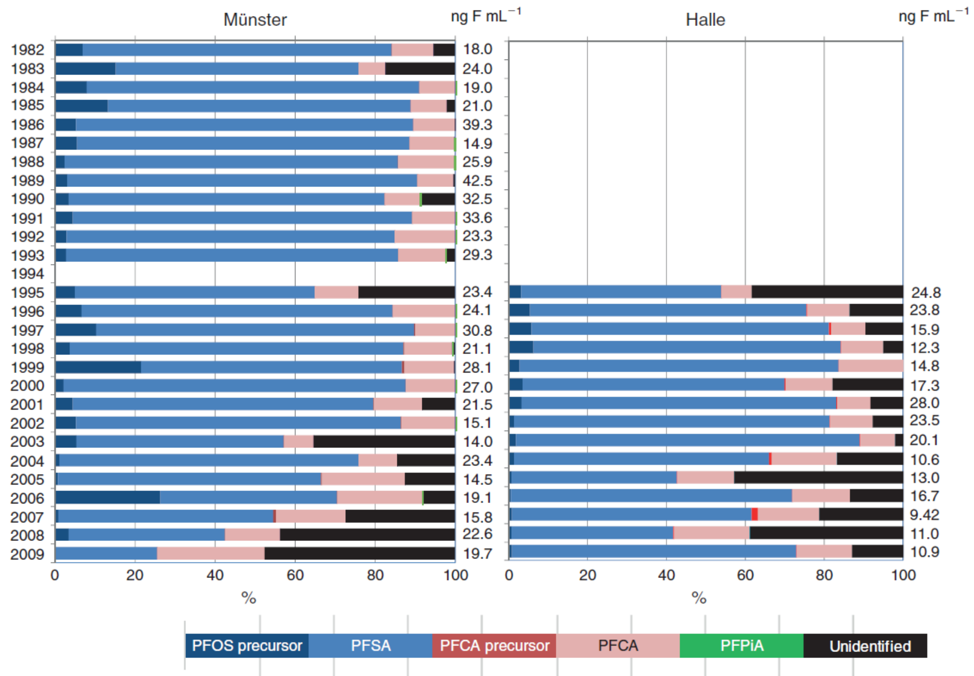
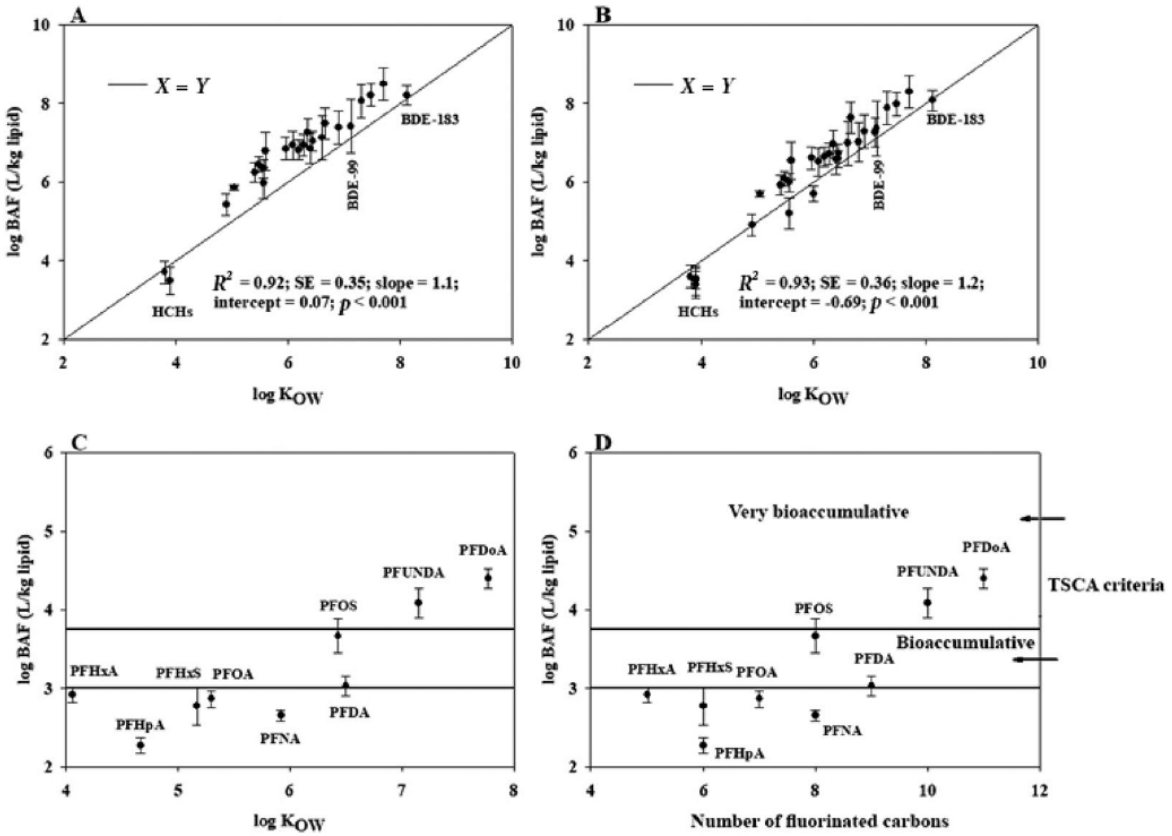


Fig. 4. Composition and concentrations (ng F mL^{-1}) of extractable organofluorine (EOF) in German blood plasma samples (perfluorooctane sulfonate, PFOS; perfluoroalkyl sulfonate, PFSA; perfluorinated carboxylates, PFCAs; perfluorinated phosphinates, PFPiAs).

(Yeung and Mabury, 2016)



(Khairy et al., 2019)

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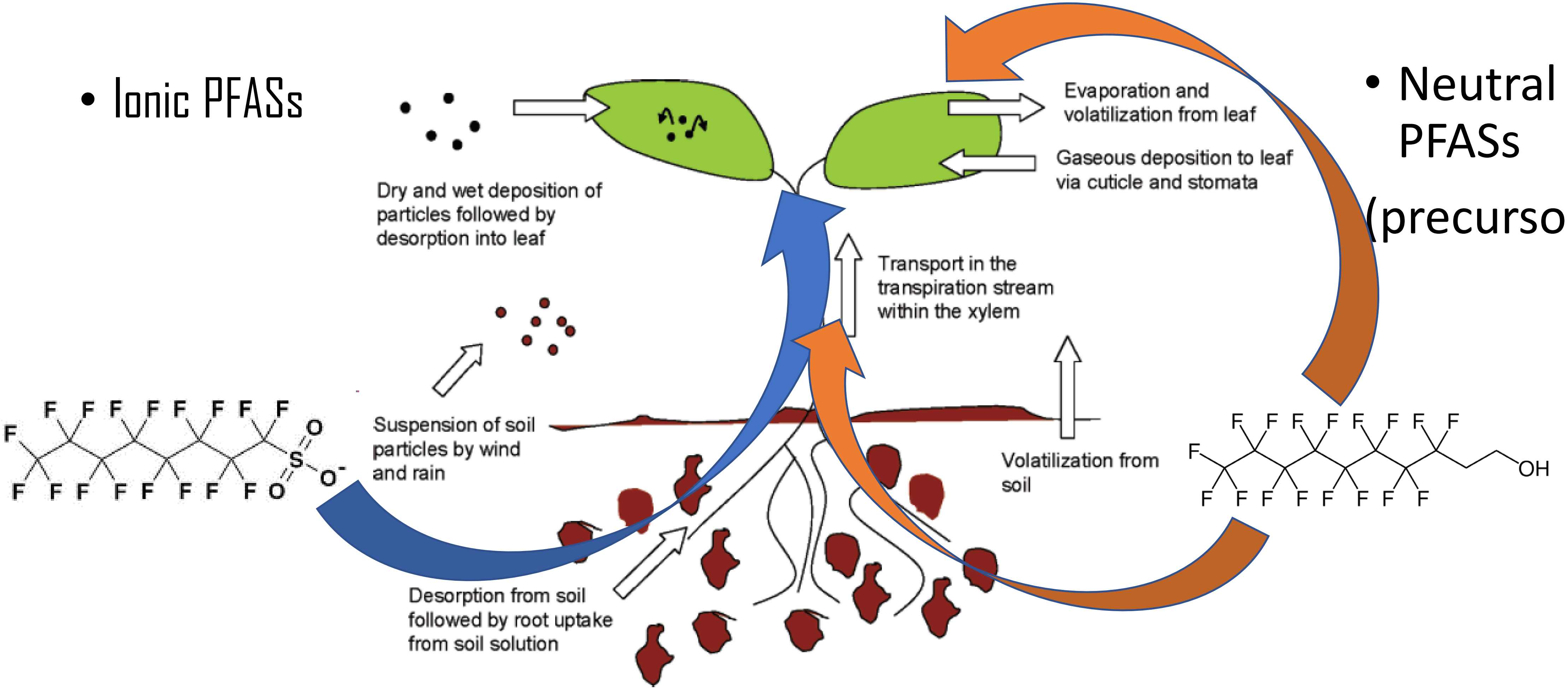
Precursors in the Arctic

- Affects likely PFOA and PFOS, among others
- Evidence for importance of precursors in Arctic abiota
- And biota..

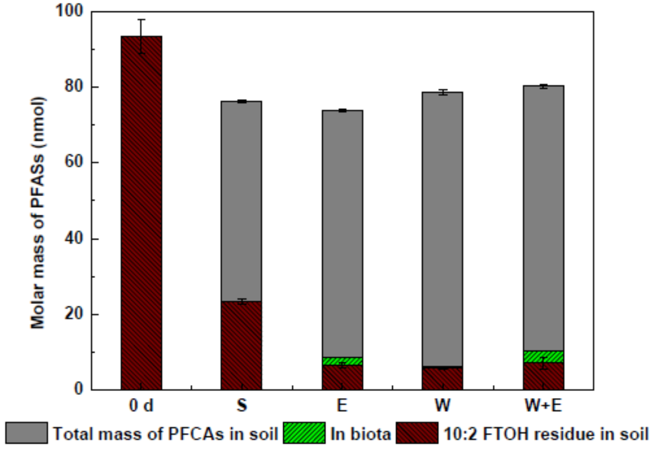
PFAS in plants = PFlAntS?

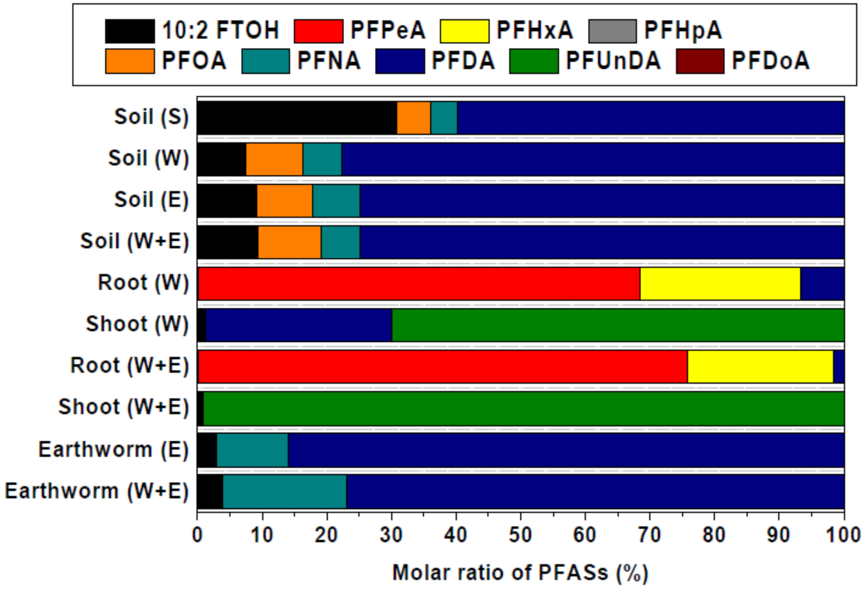
- Ionic PFASs
- Neutral PFASs (precursors)

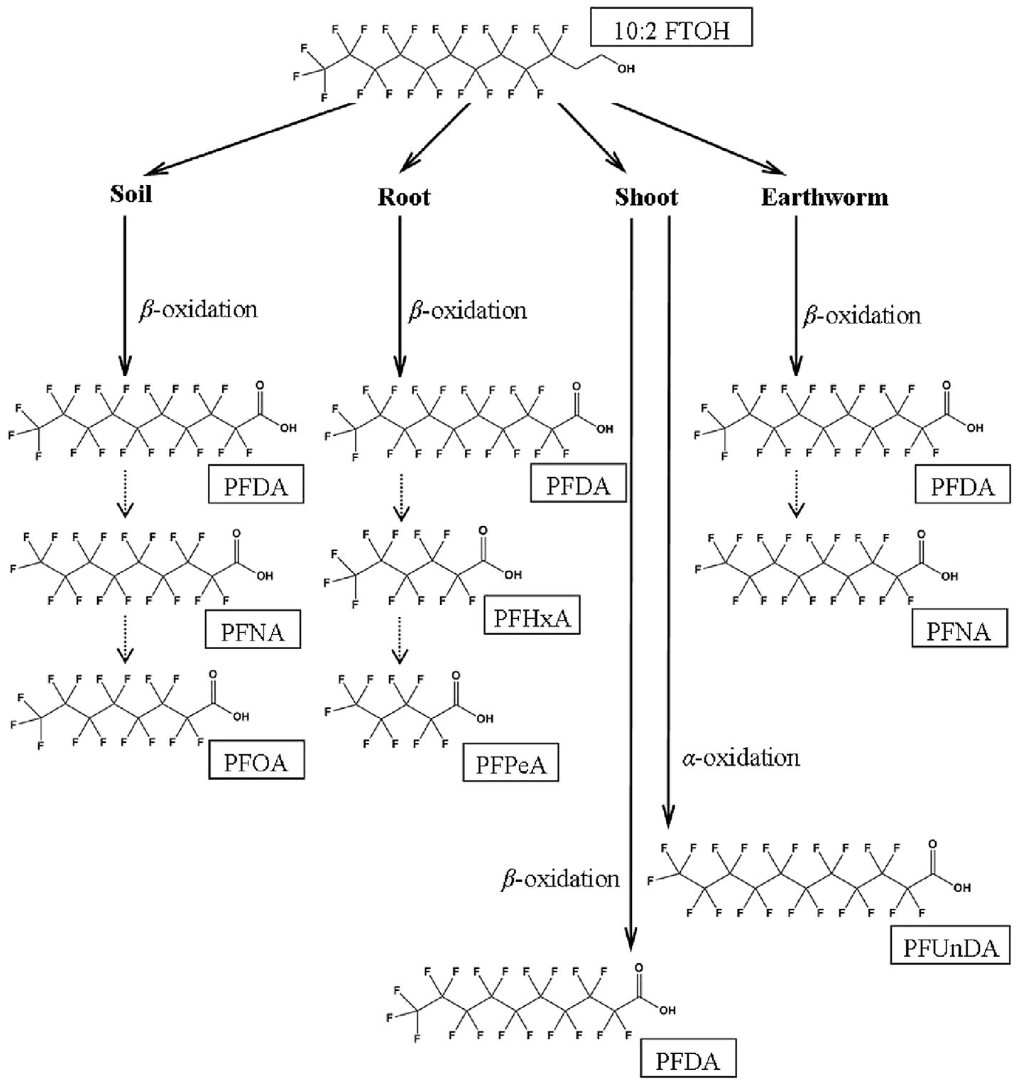


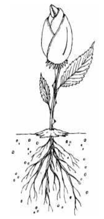
(Collins et al., 2006)

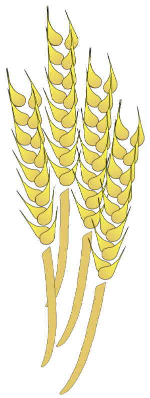
Chemists ♥ precursors – plants ⚡ them down





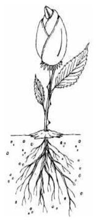






(Zhao and Zhu, 2017)

Other breakdowns



	roots	leaves
8:2 FTOH	7:3 FTCA, 7:2 sFTOH, PFOA	PFOA
N-EtFOSAA	N-EtFOSAA, PFOS	PFOS, N-EtFOSAA
N-EtFOSA	PFOS, N-EtFOSA	PFOS, PFOSA
FOSA	PFOS, FOSA	FOSA, PFOS
6:2 FTSA	6:2 FTSA, PFHpA	6:2 FTSA, PFBA

- Conversion efficiency depends on plant, precursor and time

(Wen et al., 2018; Zhang et al., 2016, Zhao, Zhou, et al., 2018, Zhao et al., 2019, Zhao, Liang, et al., 2018)

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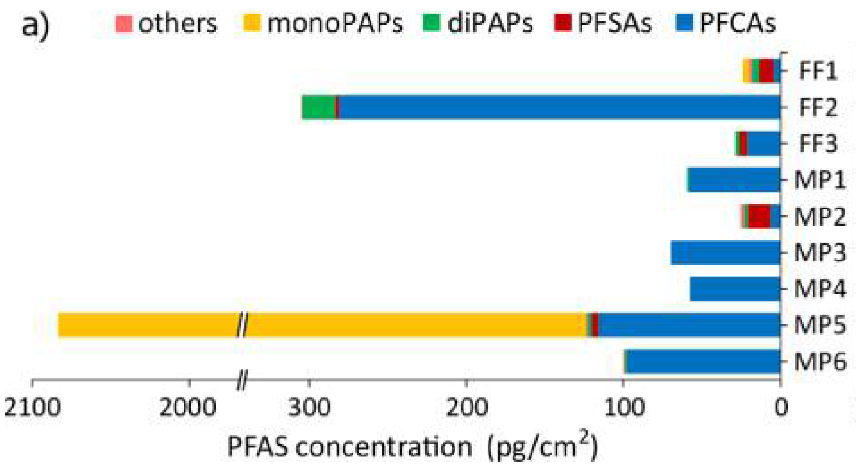
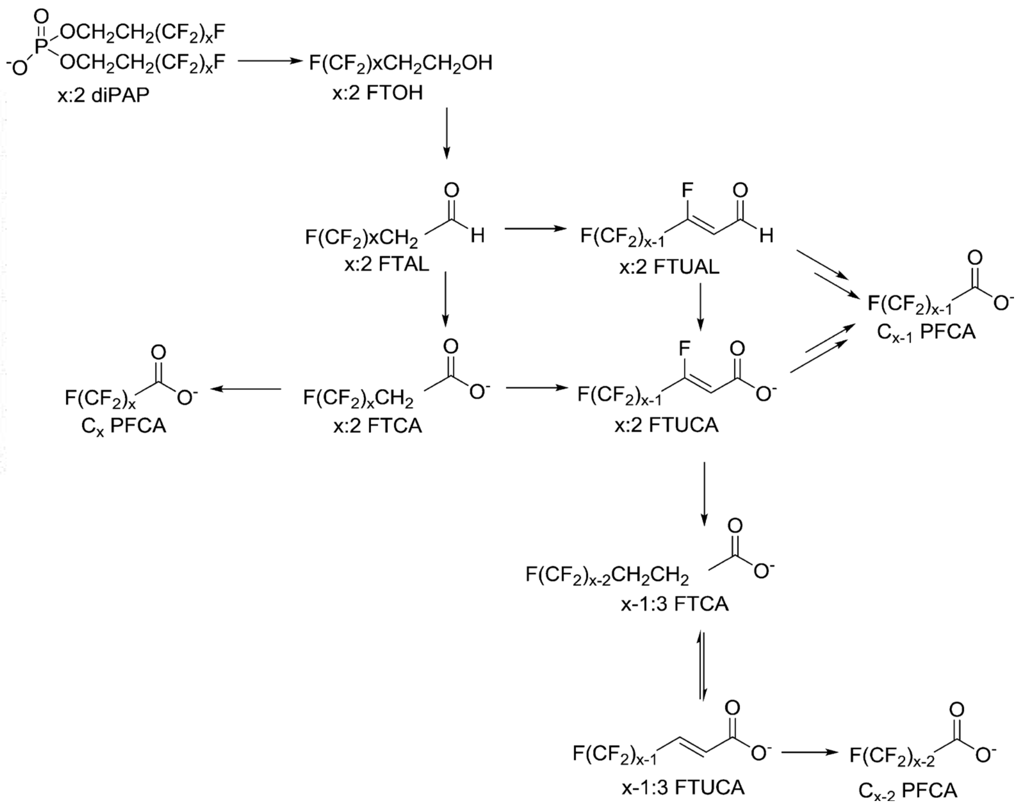
Mono- & Di-polyfluoroalkylphosphates (food packaging)

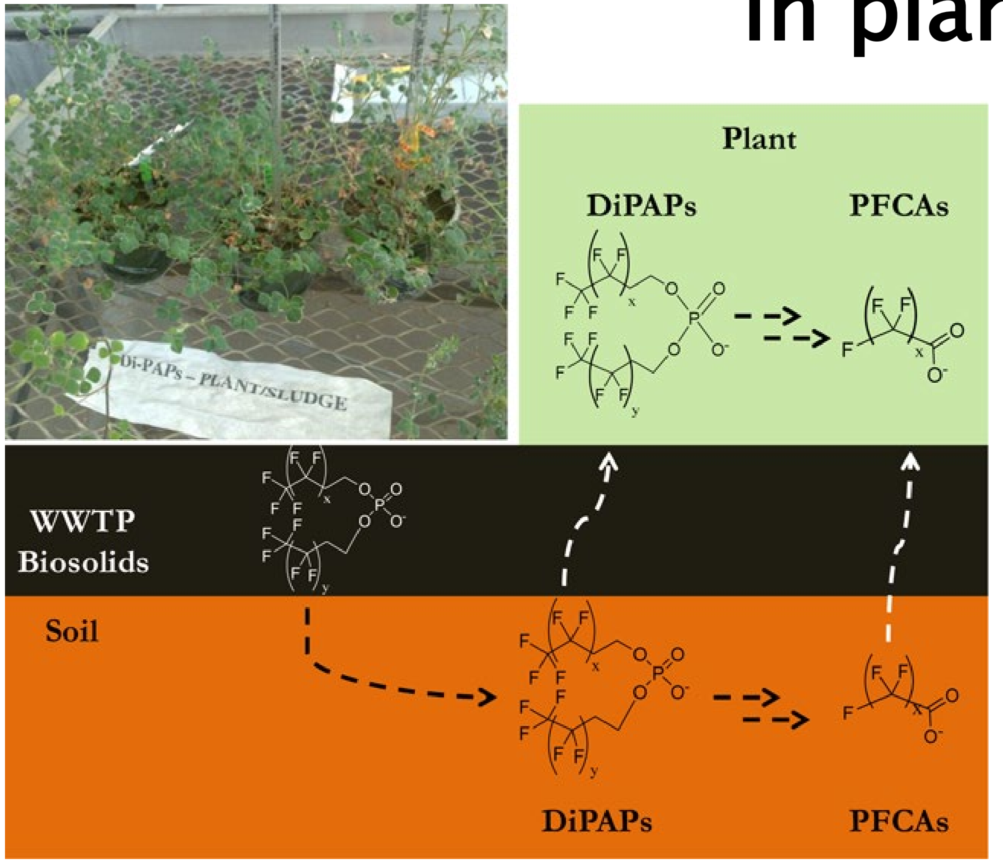


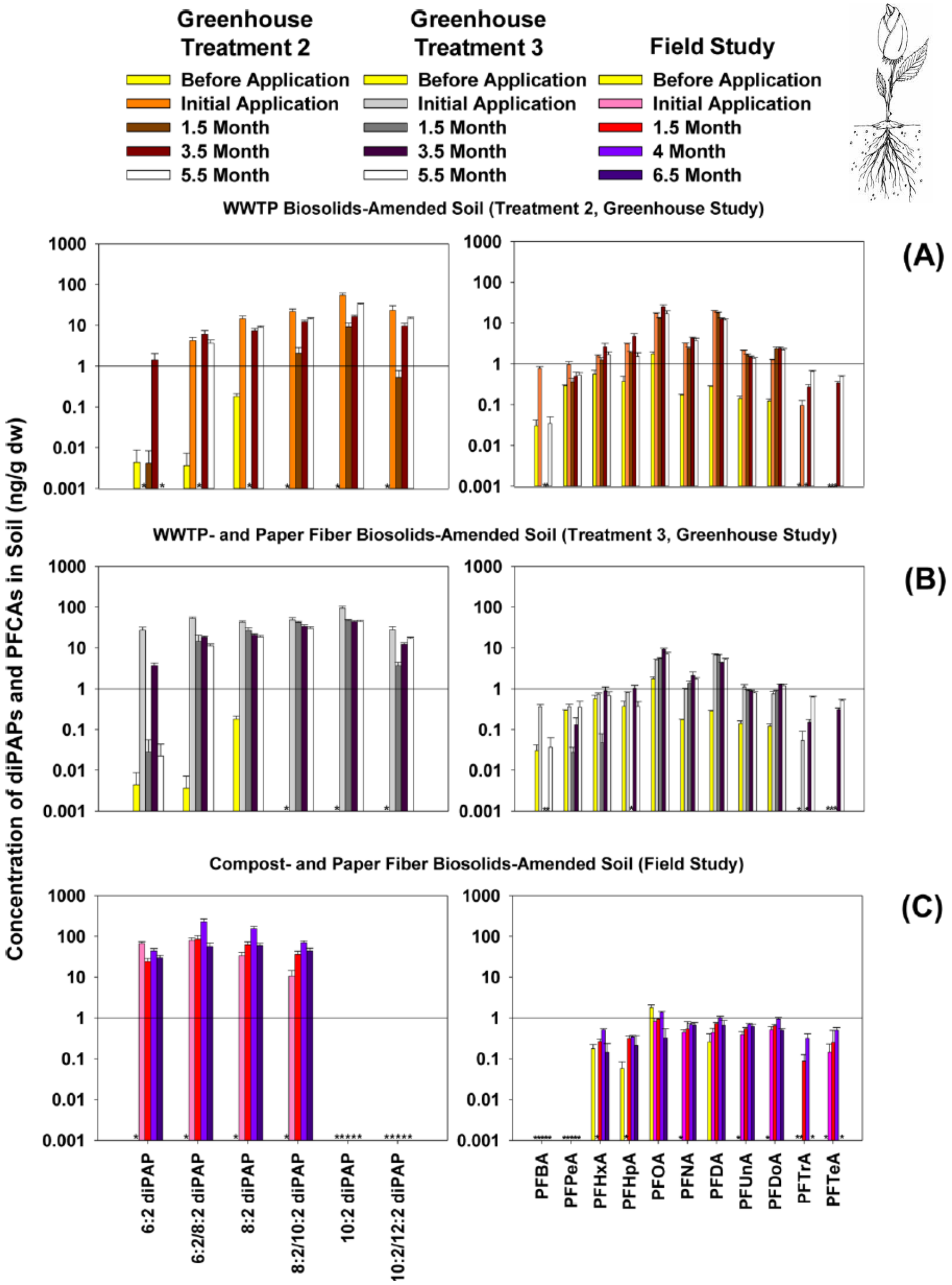
(Schultes et al, 2019)



(Rand and Mabury, 2014)

Mono– and DiPAPs in plants





(Lee et al, 2014)

Fate of spiked 6:2 DiPAP in soil/plant

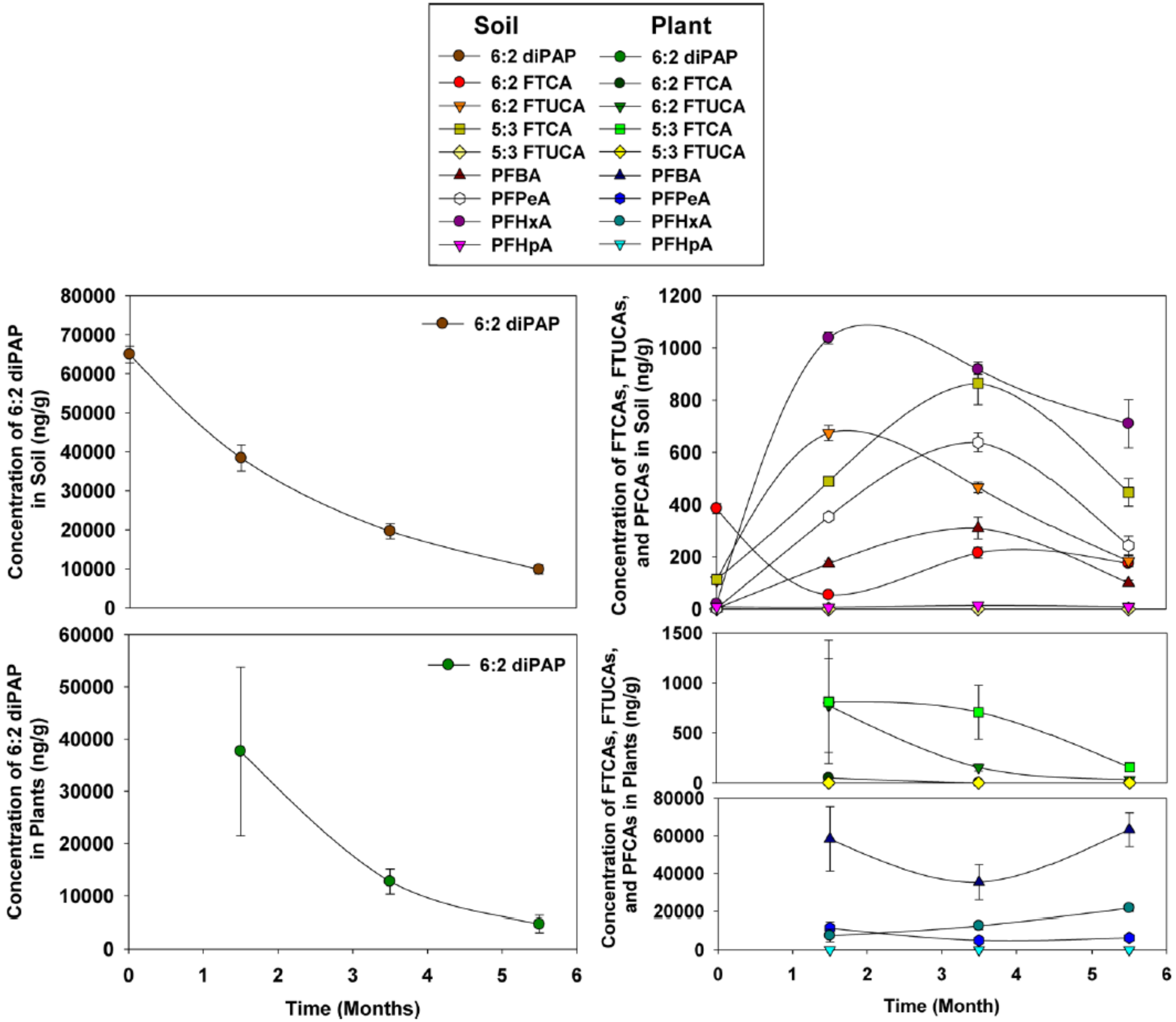
$t_{1/2}$ of 2 months

Mostly in soil

1st hydrolysis to 6:2 FTOH

Then 6:2 FTCA, 6:2 FTUCA

-> PFHxA



(Lee et al, 2014)

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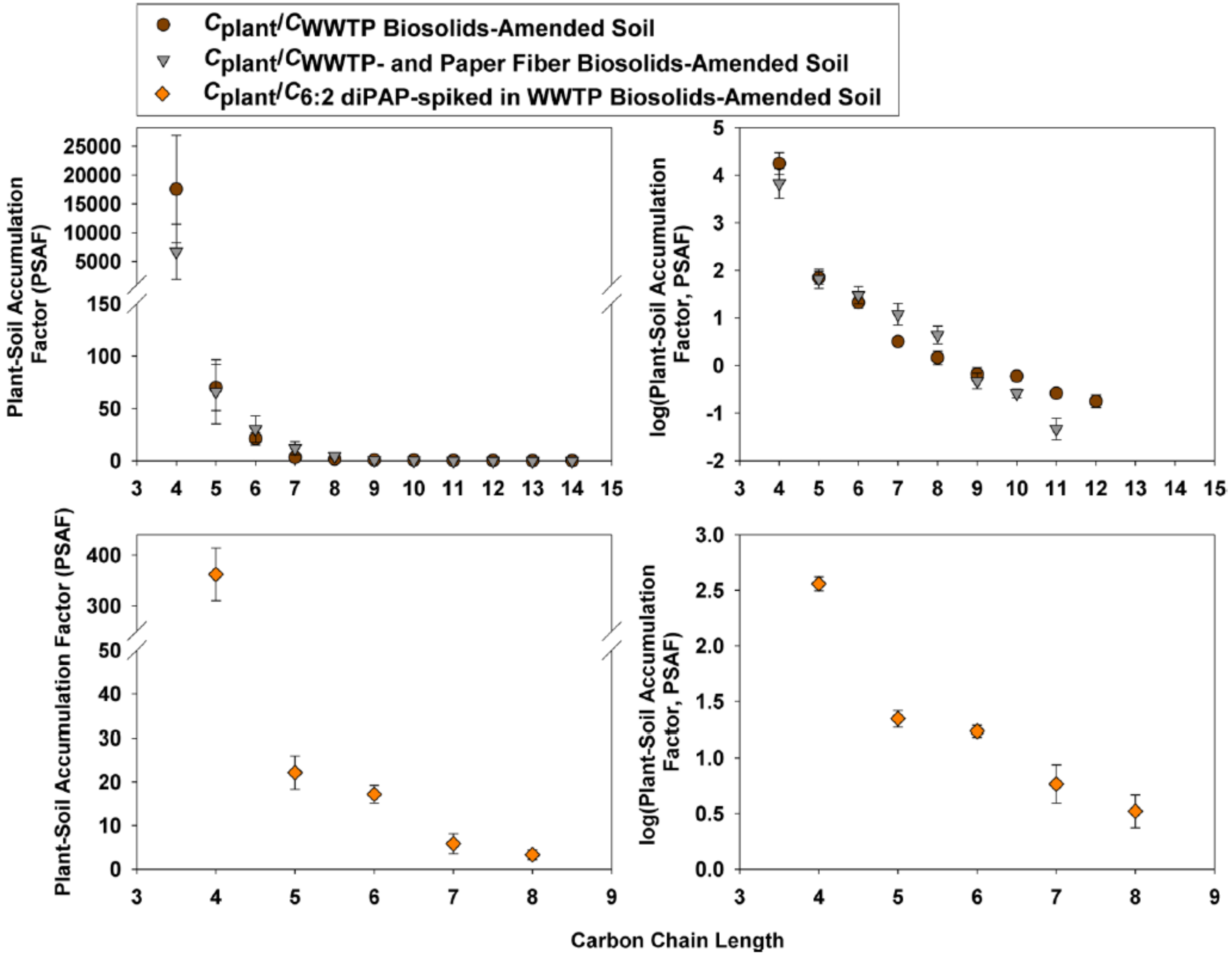
Fate of spiked 6:2 DiPAP in soil/plant

$t_{1/2}$ of 2 months

Mostly in soil

1st hydrolysis to 6:2 FTOH

Then -> PFCAs



(Lee et al, 2014)

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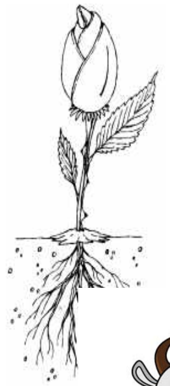
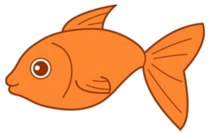
Some conclusions and thoughts

- Porewater concentrations?
- Plants return short-chain PFASs to foodwebs
- Potential opportunity for bio-removal of PFASs
- Environ. proteins and BC likely important for partitioning
- Knowledge gaps for terrestrial pathways
- Role of atmospheric precursors important (e.g., Arctic)
- Industrial vs marine signal for coastal biota
- The unknown bioaccumulating PFAS are of concern





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AFFF precursors

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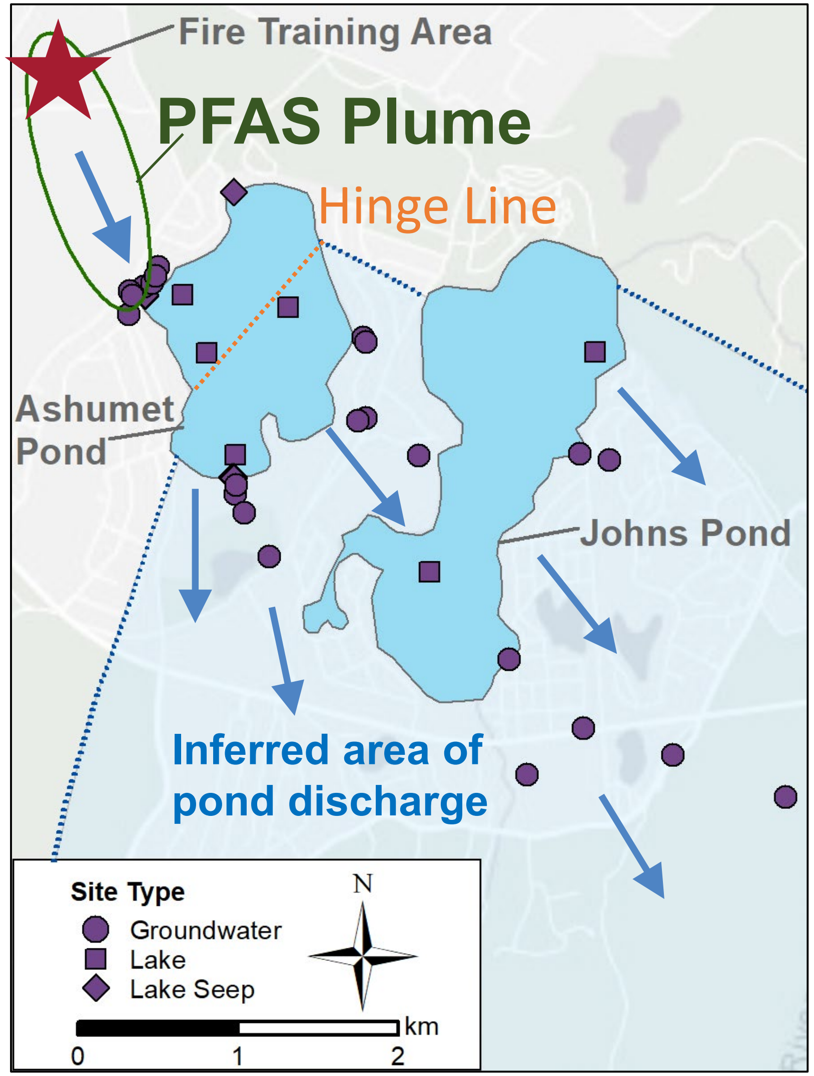
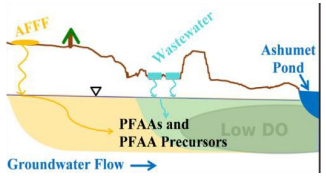
Investigating PFAS transport across groundwater/surface-water boundaries

*Interaction with a surface water body can cause significant dispersion of groundwater plumes





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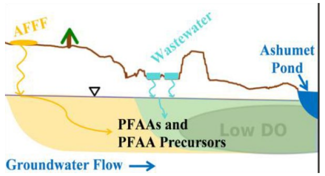
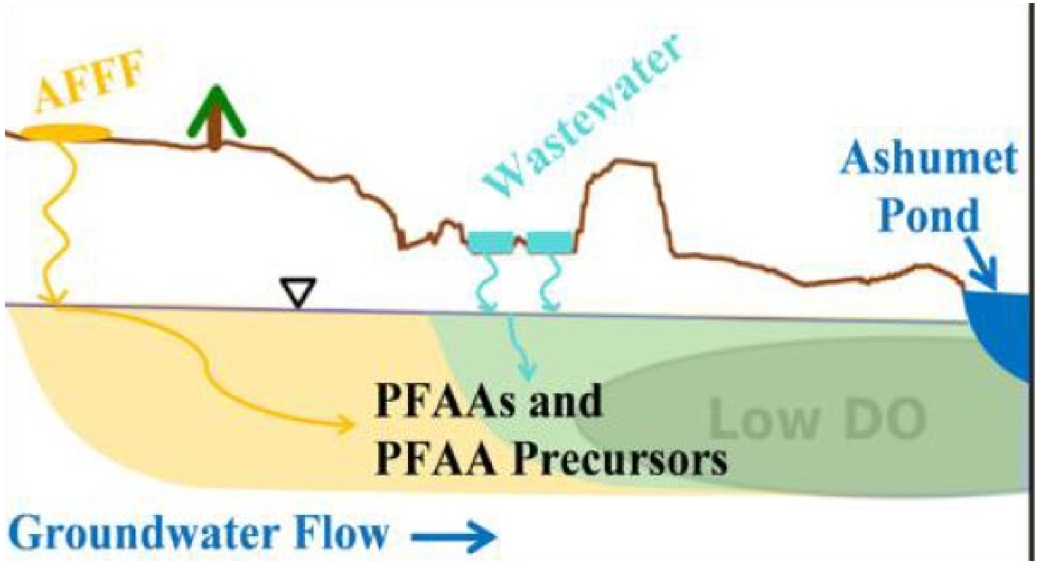
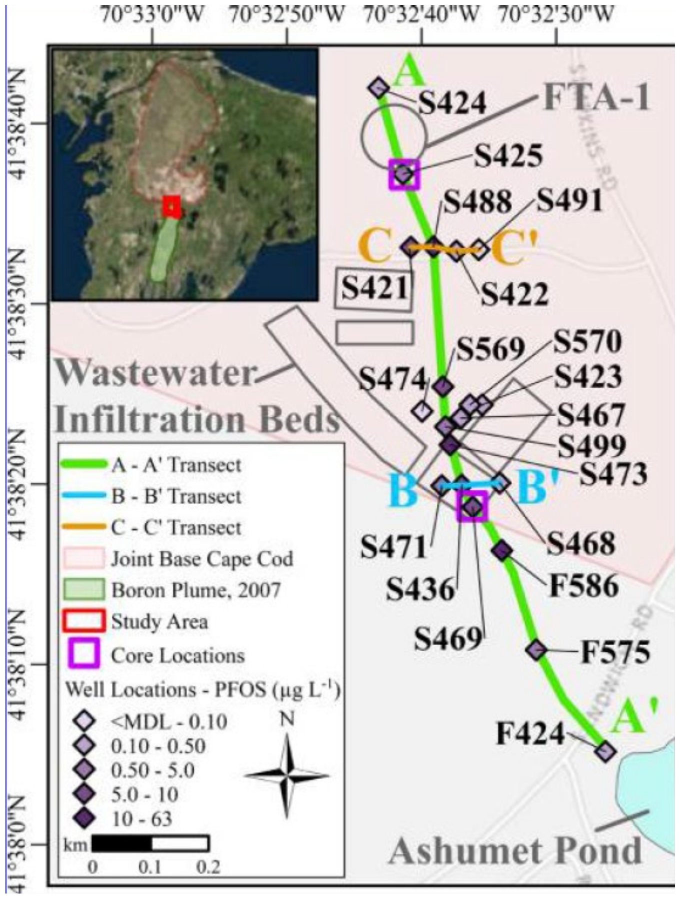
Transport and fate of AFFF precursors down under







(Weber et al., 2017)

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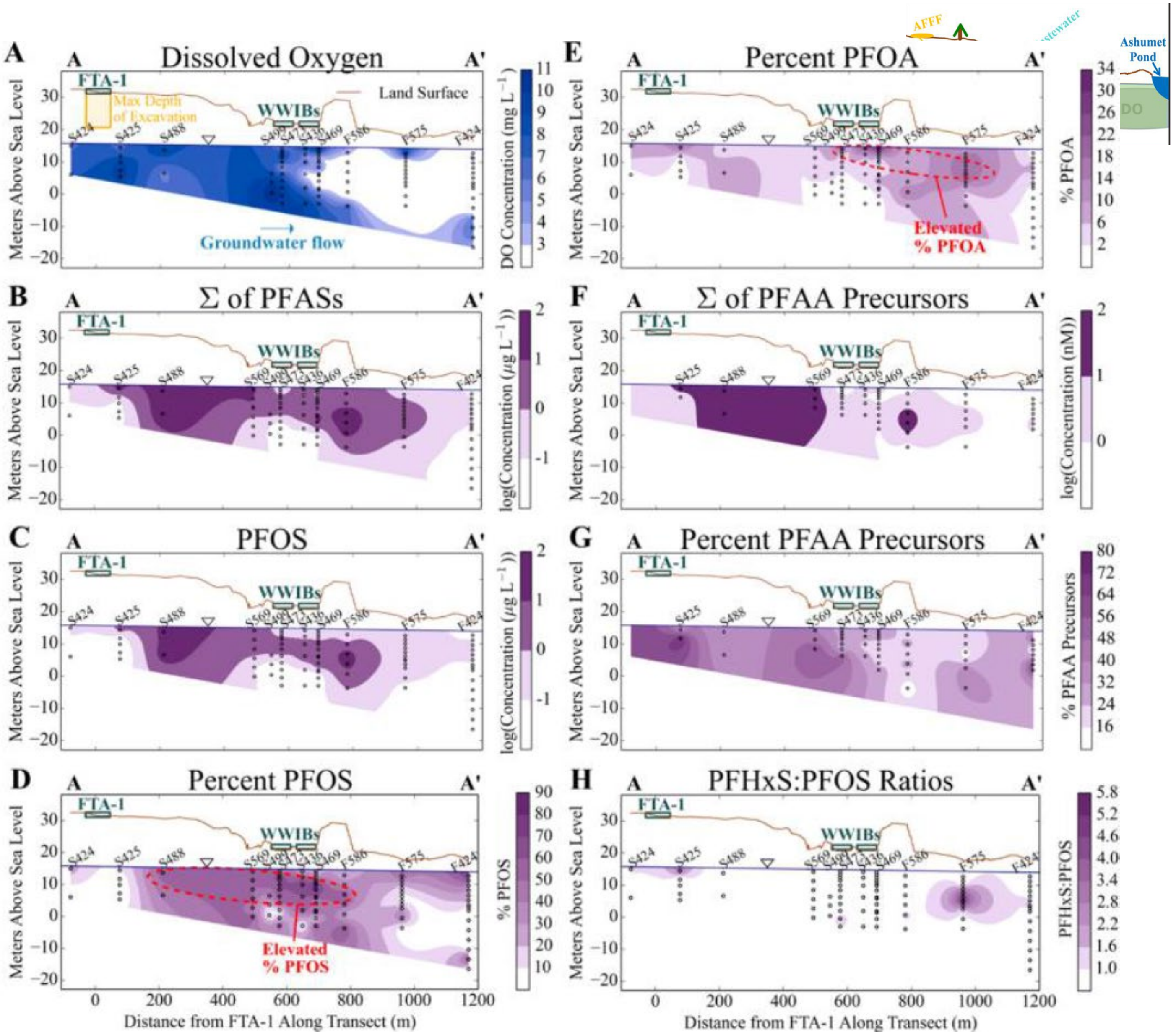
- AFFF plume from FTA-1
- 2nd source: WWTP effluent
- Precursors have not moved far
- Some evidence of transformation



(Weber et al., 2017)

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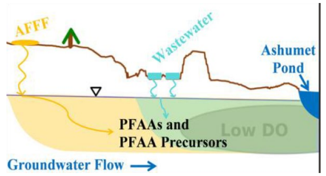


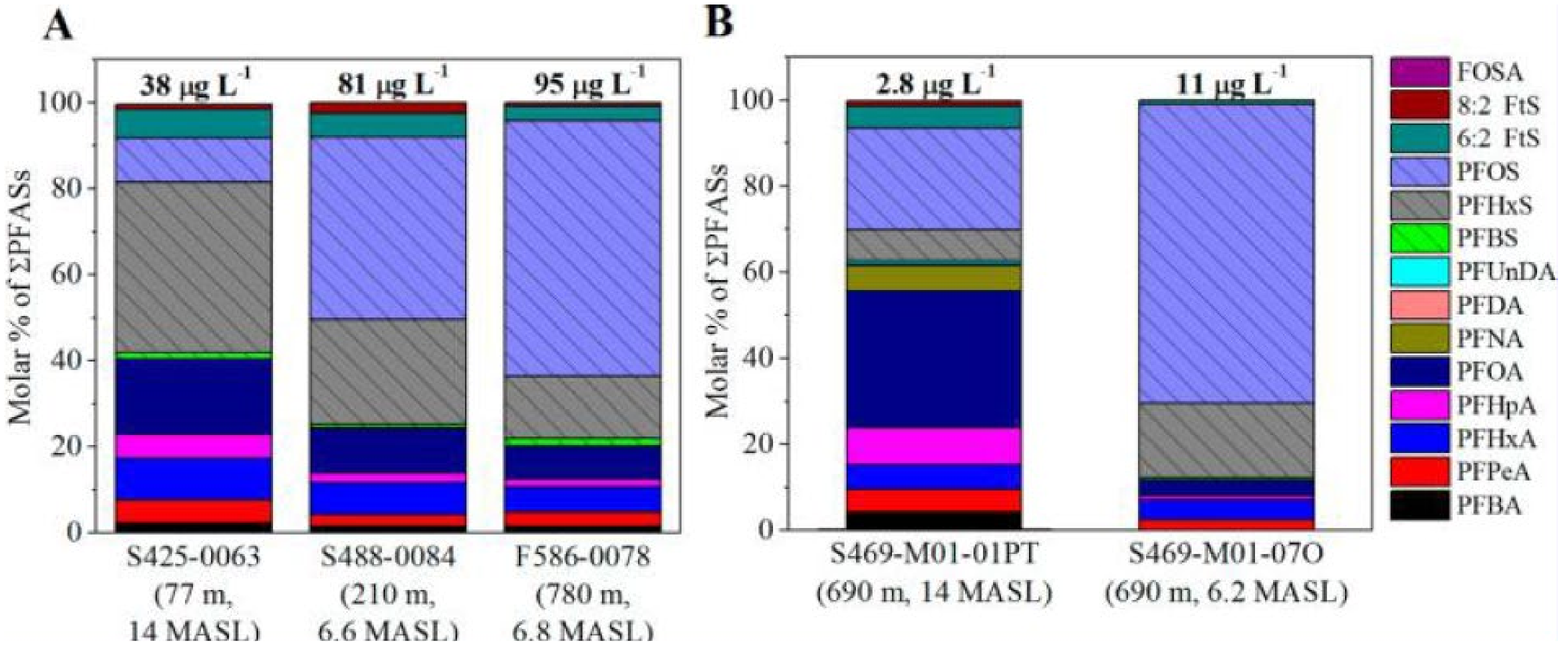
The slow conversion of precursors

In AFFF plume

- Preponderance of PFHxS, PFHxA indicative of 6:2 FTS transformation

In WWTP recharge plume

- PFNA, PFOA, also from precursors



(Weber et al., 2017)

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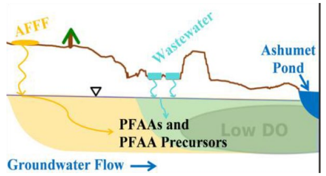


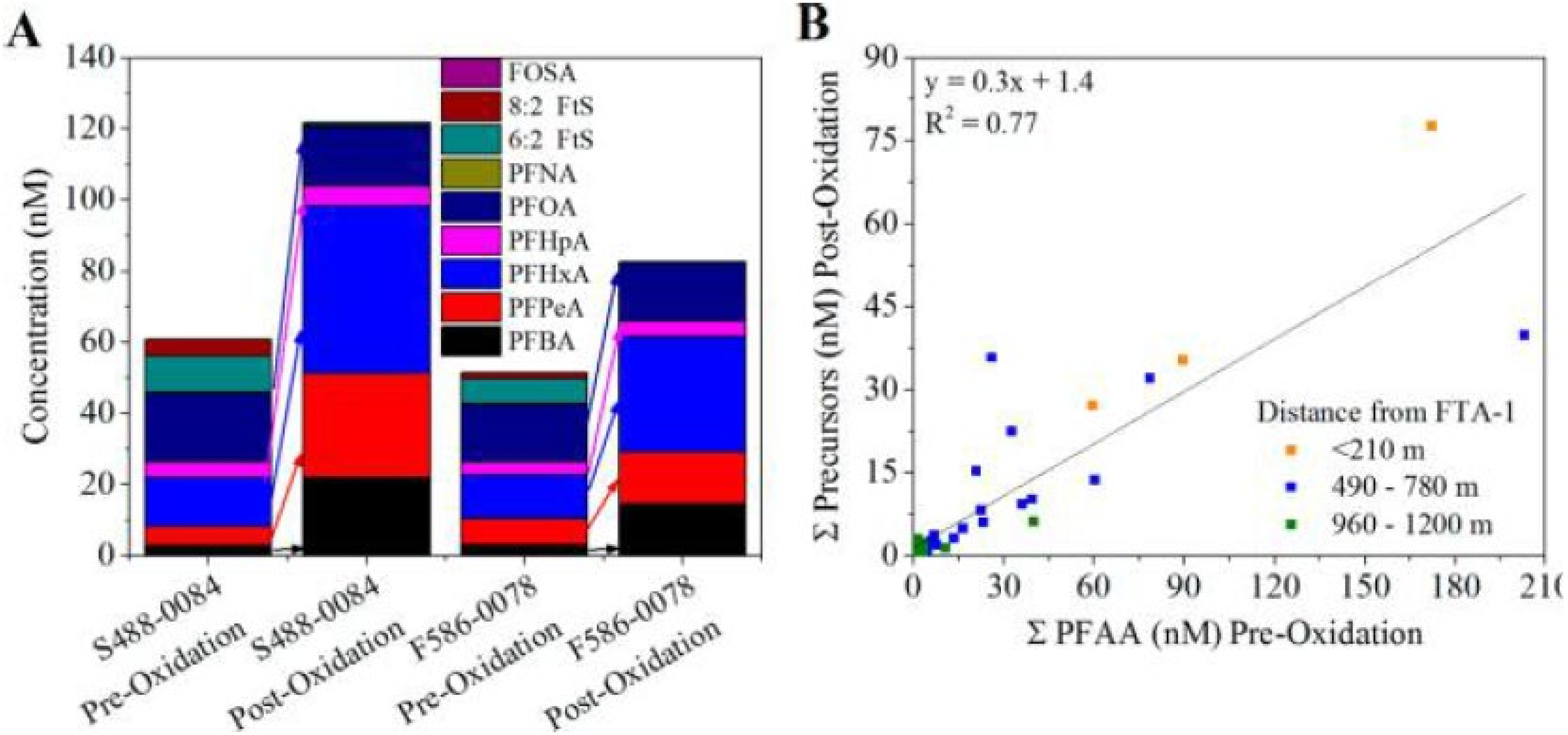
Still more to be converted

- 6:2 FTS

→ PFBA, PFPeA, and PFHxA

- 8:2 FTS

→ PFPeA, PFHxA, PFHpA, and PFOA.



(Weber et al., 2017)

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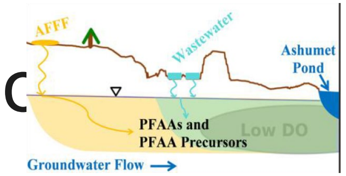
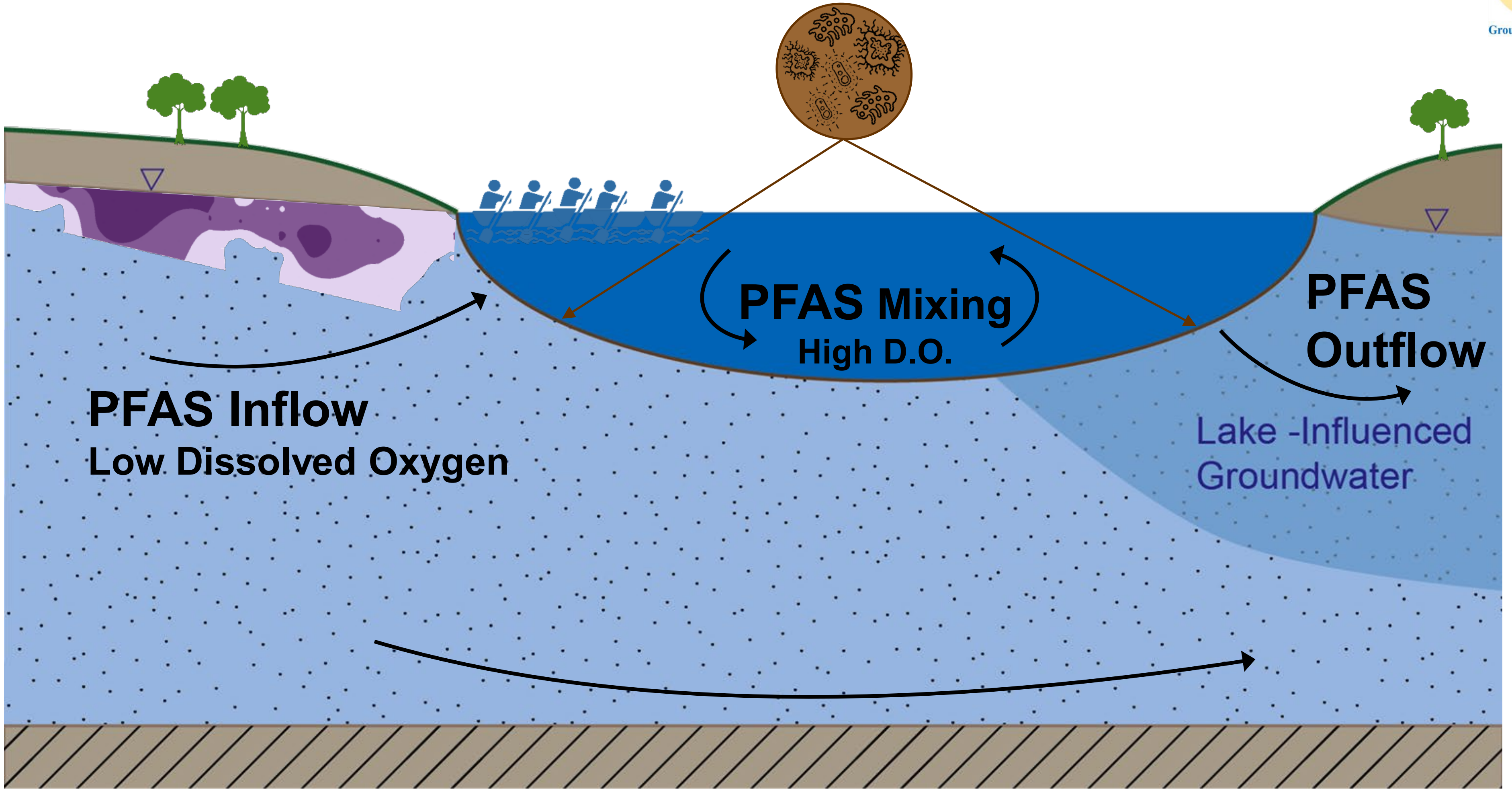
Conceptual model – ideal setup to investigate PFAA precurso





Not to Scale

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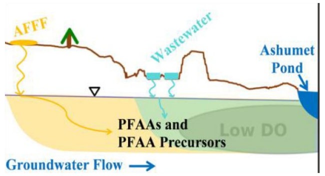
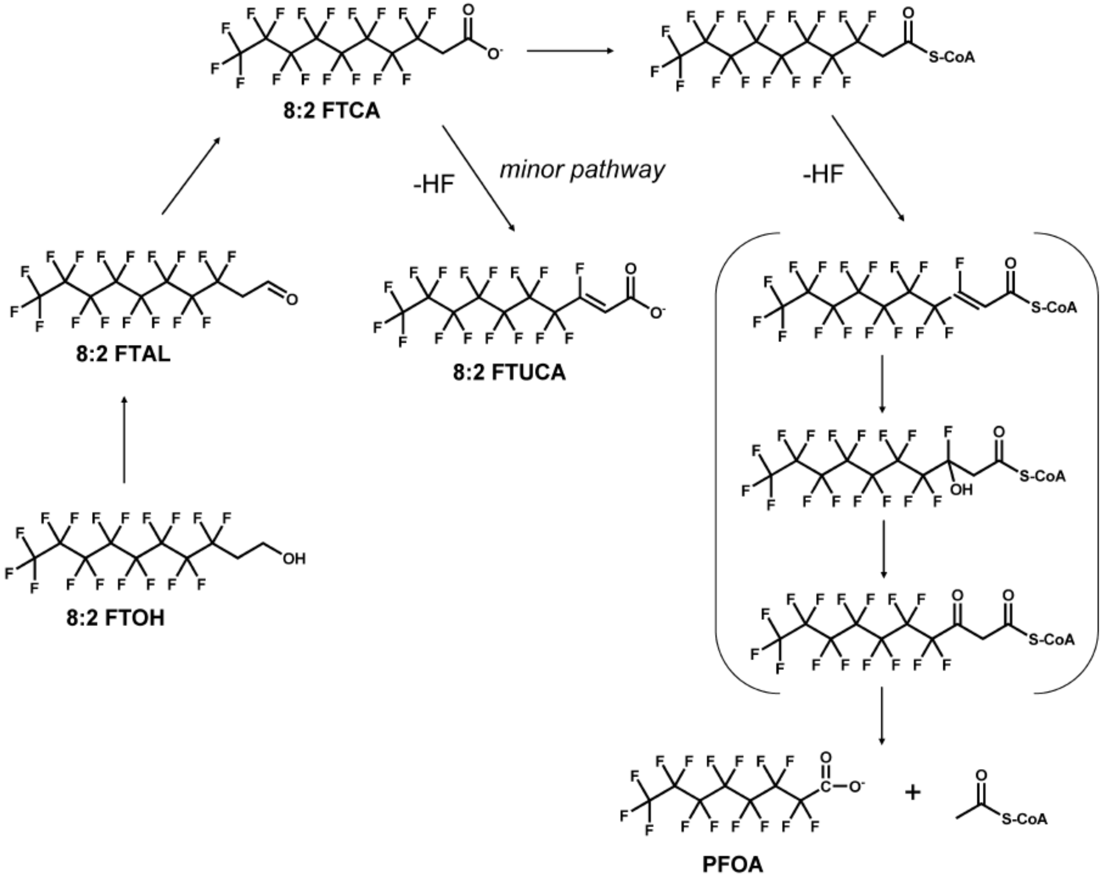
Other microbial conversion



Microbial breakdown of 8:2 FTOH



(Butt et al., 2014, after Dinglasan et al.,)

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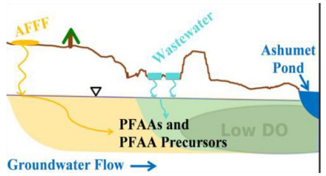
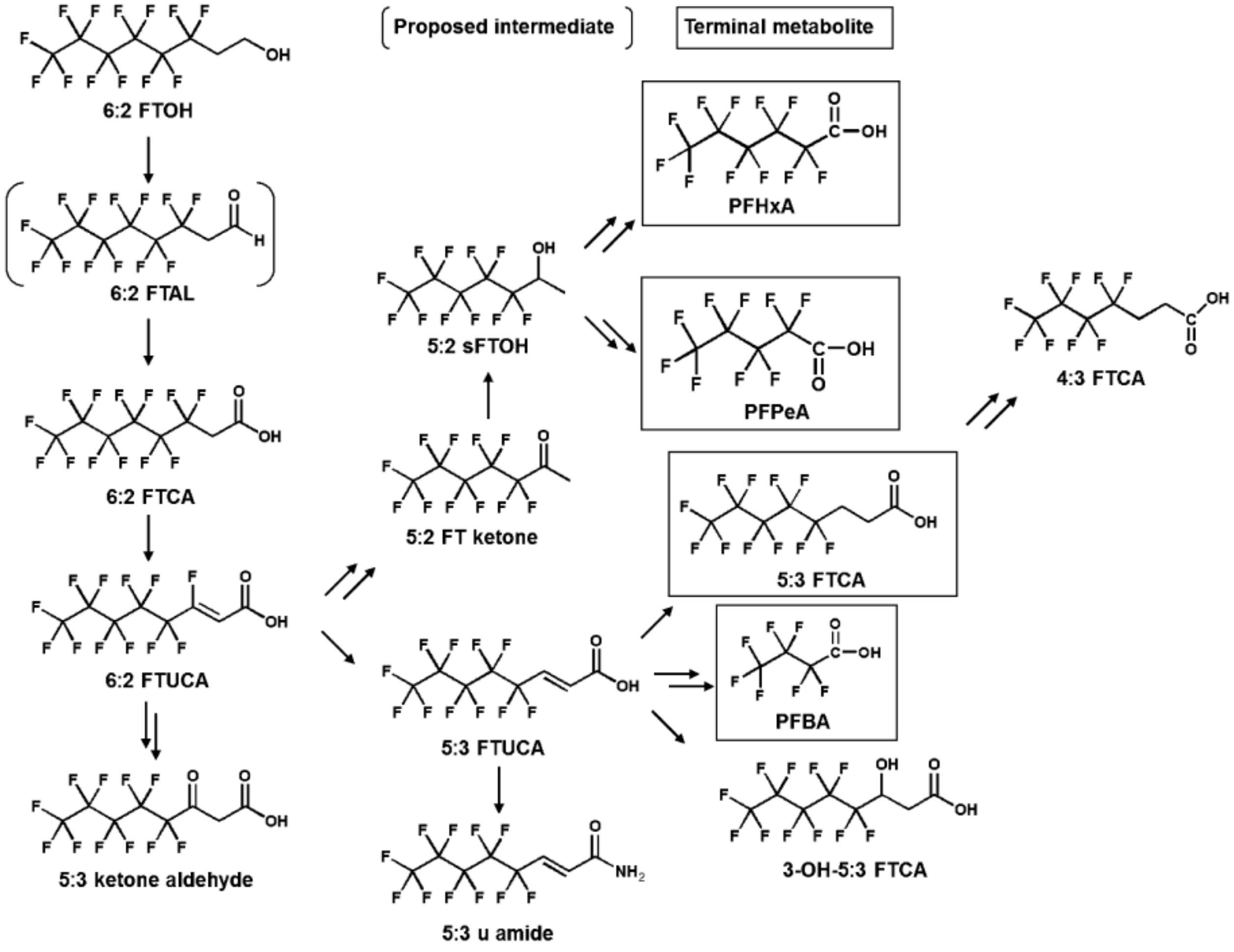


Aerobic degradation of 6:2 FTOH



(Butt et al., 2014, after Liu et al.,)

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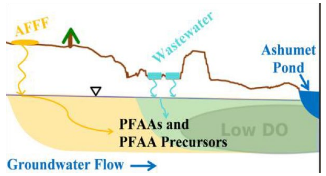
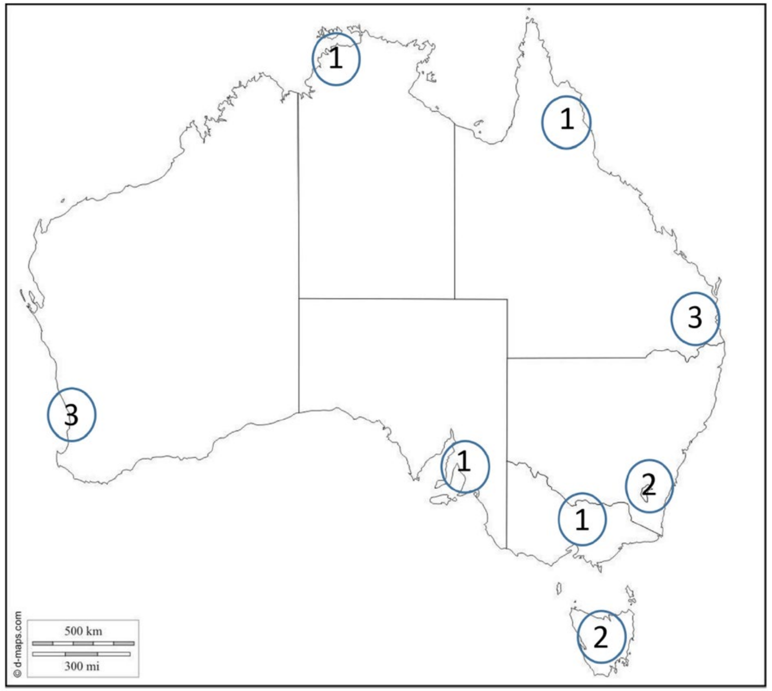
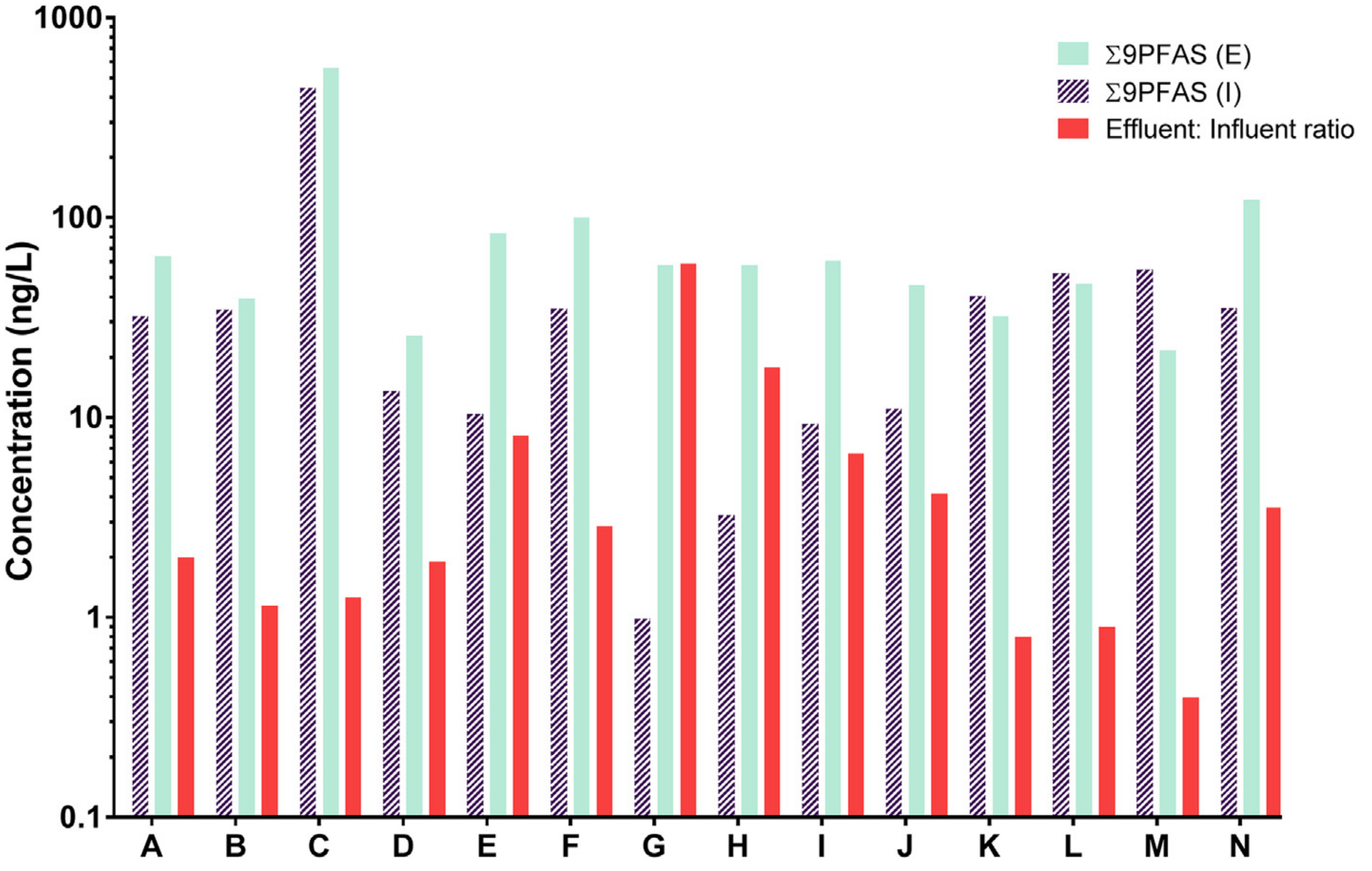
14 WWTPs sampled in 2014: Influent, effluent, biosolids



- Presence of PFASs down under
  – influent: effluent





(Gallen et al., 2018)

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WWTP down under

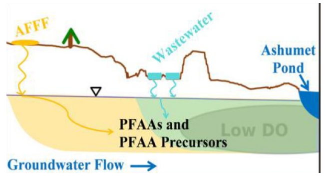


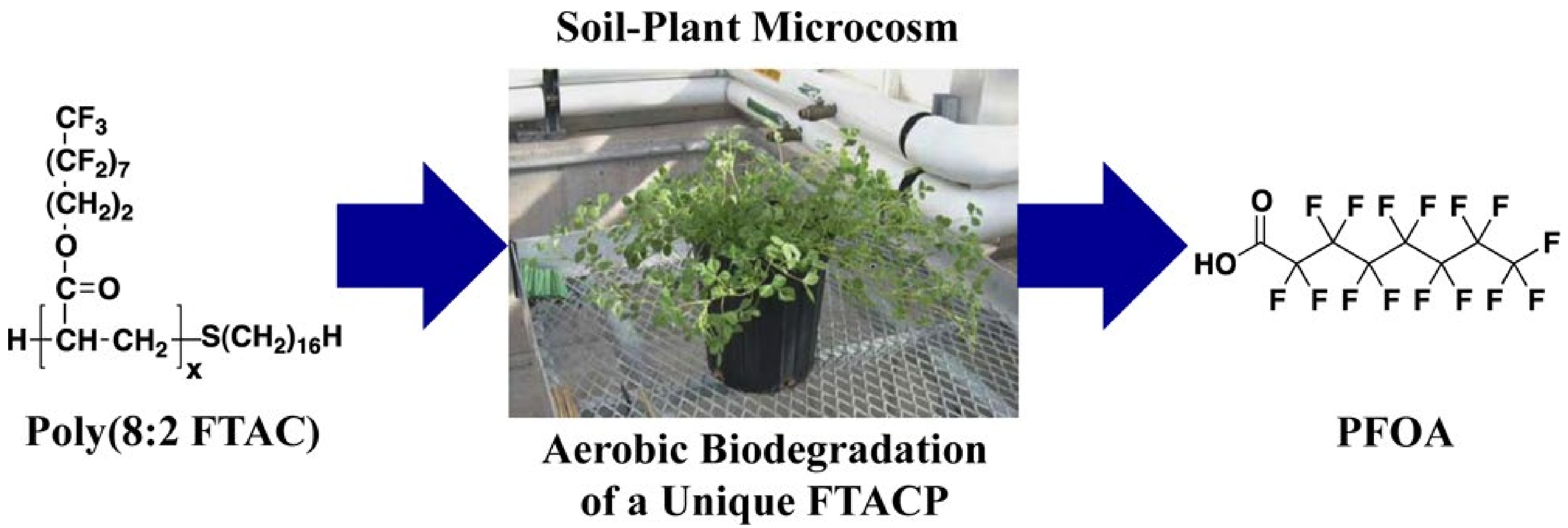
Table 1
Summary results for influent, effluent and biosolids.

	PFHxA	PFHpA	PFNA	PFDA	PFUdA	PFDoDa	PFHxS	PFOA	PFOS	Σ9PFAS
Influent										
Mean	9.5	2.5	0.64	0.36	0.18	0.03	20	4.8	17	55
SD	15	3.3	2.0	0.68	0.31	0.10	54	6.9	35	117
Median	6.9	2.0	0.00	0.00	0.00	0.00	6.0	0.79	7.2	33
% detection	71	79	14	29	36	7.1	86	50	86	100
Effluent										
Mean	18	3.6	1.3	3.0	0.49	0.11	20	22	25	94
SD	9.2	1.2	0.73	2.3	0.19	0.21	3.5	15	7.3	29
Median	13	3.0	1.1	2.8	0.33	0.00	7.0	18	6.6	57
% detection	93	100	71	86	79	14	100	93	86	100
Biosolids										
Mean	0.50	0.26	1.1	17	1.2	5.7	0.11	6.5	25	45
SD	1.1	0.57	1.2	16	1.0	4.6	0.26	8.1	31	50
Median	0.00	0.00	1.0	11	1.4	5.8	0.00	0.0	11	40
% detection	27	18	64	100	73	82	18	45	91	100

- Strongest increases for PFCAs, much less so for PFSAs
- PFDA (9x), PFOA (4x), PFUdA, PFDoDa (3x), PFNA, PFHxA (2x)

(Gallen et al., 2018)

Potential for side-chained fluoropolymer to liberate precursors



(Rankin et al., 2014)

Some summarizing thoughts

- Precursors are everywhere.
- 8:2 FTOH and 10:2 FTOH with global presence, and caused baseline contamination of Arctic (and global?) air, snow, water.
- FOSA likely implied in contamination and sharp decrease of PFOS in Arctic region, including trends in biota
- Atmosphere, plants, microbes breakdown precursors, with different endproducts
- WWTP efficient transformers of precursors; less obvious for groundwater
- Much greater indoor concentrations, and associated human exposure

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Please Hold the Date:

FLUOROS 2020

Oct 13-16, Providence (RI)

Questions?

THE UNIVERSITY OF RHODE ISLAND

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SILENT SPRING INSTITUTE
Researching the Environment and Women's Health

NIH National Institute of Environmental Health Sciences
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