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Battelle Sediment Conference
February 11-14, 2019
New Orleans, Louisiana

Bioavailability, Uptake, Bioaccumulation, and Biomagnification of Per- and Polyfluoroalkyl Substances in Sediments

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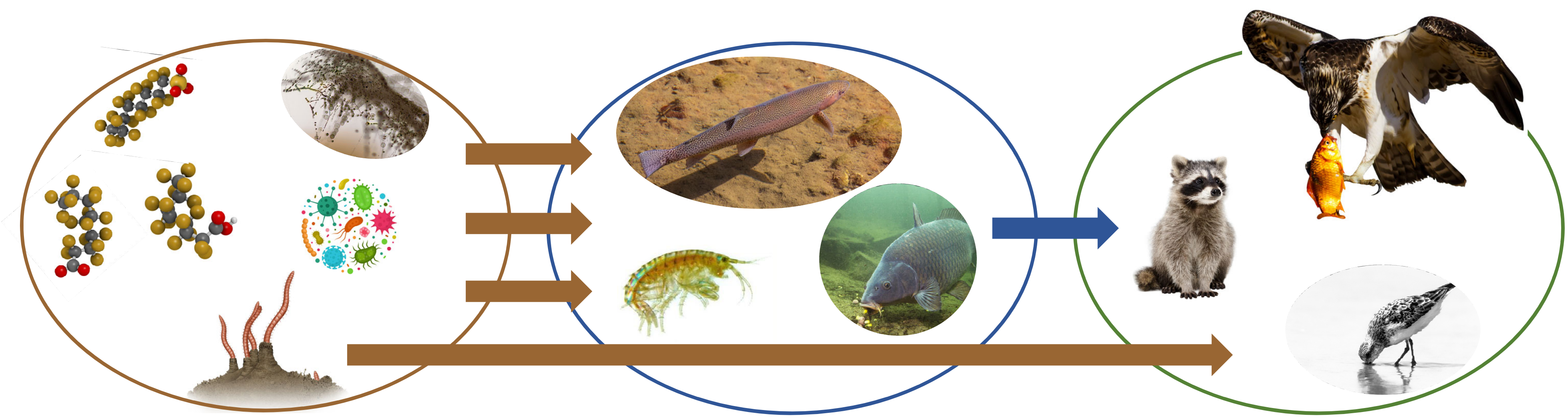
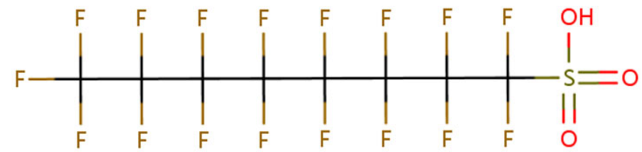


Overview

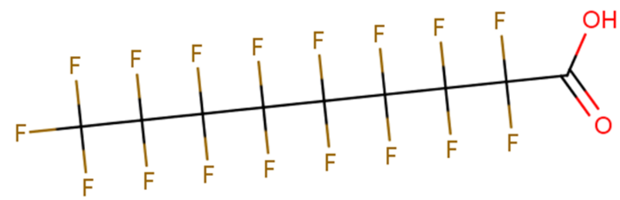
- Why are PFAS so different?
- Sediment accumulation
- Assessment challenges
- Bioconcentration by microbiota
- Bioaccumulation by macrobiota
- Biomagnification, food web transfer
- Risk assessment activities needed

Why are PFAS so Different?

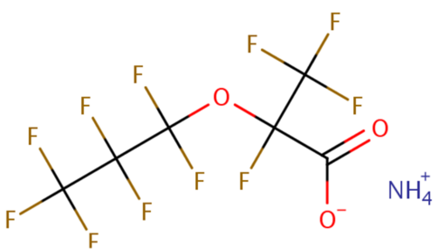
- Super-strong carbon-fluorine bond
- Thousands of different chemicals – dozens that can be identified in environmental samples
- Typically, organic chemicals accumulate in lipids/fatty tissues – PFAS bind to proteins



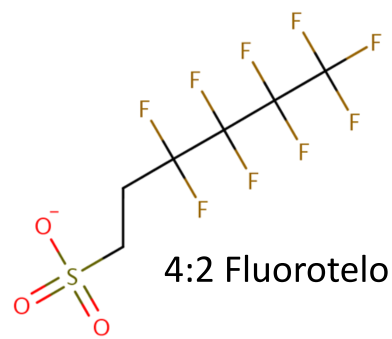
Perfluorooctanesulfonic acid (PFOS)



Perfluorononanoic acid (PFNA)



Ammonium perfluoro-2-methyl-3-oxahexanoate (GenX)



4:2 Fluorotelomer sulfonate (4:2 FtS)

Ng *et al.* (2013, 2014); Armitage *et al.* (2012, 2013); Houde *et al.* (2011) .

Structures: comptox.epa.gov

Sediment ⟷ Porewater and Surface Water

PFAS partitioning between sediment and water

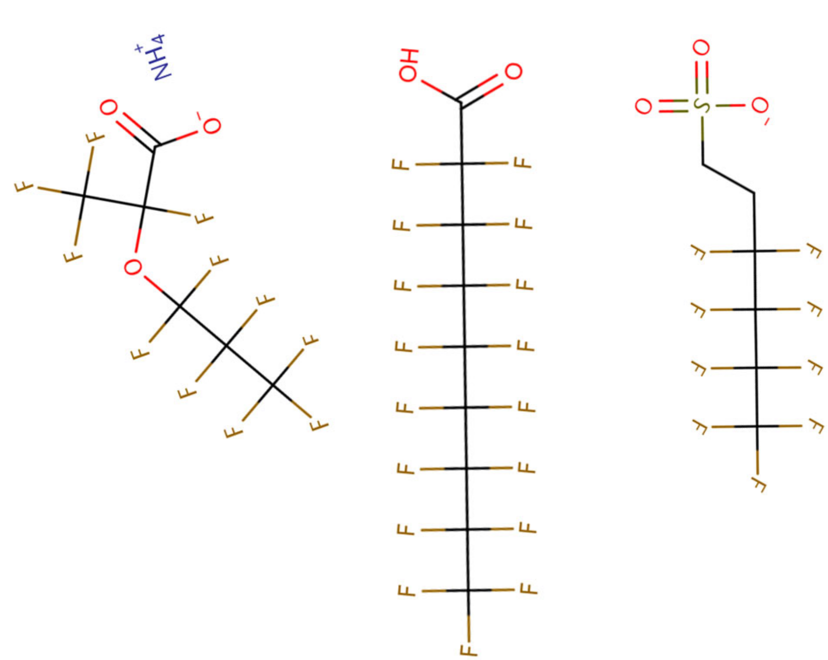
- PFAS chemicals are diverse – polarity alone is quite variable:
  - Negatively charged (anions)
  - Positively charged (cations)
  - Both positive and negative charges (zwitterions)
  - Neutral
- Many of the useful fluorinated surfactants are anions:
  - PFOS and replacements used in firefighting foams
  - PFOA and replacements used to manufacture fluoropolymers

Surfactants

Surfactants: non-polar tail, polar head

Example of a non-fluorinated surfactant sodium dodecyl sulfate (sodium lauryl sulfate):

PFAS: fluorosurfactants



Structures: comptox.epa.gov; pubchem.ncbi.nlm.nih.gov

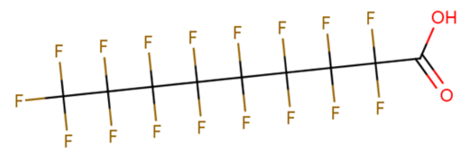
Sediment Accumulation

Sediment sorption depends on:

- PFAS chemistry: chain length, functional groups, electrical charge, pollutant load
- Water chemistry: pH, ion concentration (*e.g.*, salinity)
- Sediment chemistry: organic carbon, iron oxides, surface charge of other minerals

Sulfonic acids sorb more than carboxylic acids:
PFOS > PFOA

Longer chain: more sorption: perfluorononanoic acid (PFNA) > perfluoropentanoic acid (PFPeA)



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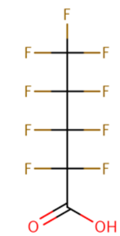

More organic carbon or iron oxides: more sorption

Liu *et al*. (2019); **Li *et al*. (2018)**; Munoz *et* al. (2017b); Valsecchi *et al*. (2017); Campo *et al*. (2016); Chen *et al*. (2016); Concawe (2016); Qi *et al*. (2016); Gao *et al*. (2015); Ahrens *et al*. (2014, 2009); Kwadijk *et al*. (2014); Ferrey et al. (2012); You *et al*. (2010); Higgins *et al*. (2007).

Images: comptox.epa.gov; usgs.gov; pubchem.ncbi.nlm.nih.gov

Sediment-Water Distribution Coefficients (K_d)

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K_d varies with chain length/functional group, type of sediment

Marine example
Martin *et al*. (2019):

Log K_d 0.1 (PFBA)

Log K_d 2.5 (PFOA)

River/lake/canal example
Kwadijk *et al*. (2010):

Log K_d 1.2 to 2.8 (PFOA)

Log K_d 2.3 to 4.0 (PFNA)

Log K_d 0.5 to 2.1 (PFBS)

Log K_d 1.7 to 3.0 (PFOS)

Salinity increases:
sorption increases

pH decreases:
sorption increases

Martin *et* al. (2019); Munoz *et al*. (2017b); Kwadijk *et al*. (2010).You *et al*. (2010).

Assessment Challenges

- Effects of mixtures and multiple stressors
- Precursor biotransformation
- Effects on biofilm composition/nutrient value
- Trophic consequences – exposure across food webs

Nilsen *et al*. (2019); Munoz *et al*. (2018); Ahrens *et al*. (2014); Sabater *et al*. (2007).

Estimating Bioaccumulation

Octanol-water partitioning constant (K_{ow}), typically used for estimating organic pollutant bioaccumulation, is not a good estimate for PFAS uptake:

- K_{ow} estimate is based on partitioning to lipid/fatty tissues
- K_{ow} cannot be measured for ionic surfactants – their surface-active nature prevents the empirical determination of K_{ow}

Bioconcentration and bioaccumulation of perfluorinated acids is related to:

- Length of the fluorinated carbon "tail"
- Whether the ionic "head" is a carboxylic acid (*e.g.*, PFOA) or a sulfonic acid (*e.g.*, PFOS) – sulfonate ion is more bioaccumulative

Munoz *et al*. (2017a); Conder *et al*. (2008).

PFAS Uptake by Biota

- Aquatic organisms mainly accumulate PFAS by absorption of free PFAS in water via respiration.
- Benthic organism accumulation depends on exposure:
  - Longer-chained PFAS more likely to be on sediment.
  - Feeding exposure to sediment-sorbed.
  - Respiration exposure to dissolved.
- Benthic *micro*biota: primary exposure is to dissolved PFAS – growing on sediment surface, but take nutrients from water.

Houde *et al*. (2011).

Benthic Microflora and Microfauna

Geotechnical Environmental Ecological Water Construction Management

Benthic microbiota are the primary producers in the sediment ecosystem

Biofilms are complex sediment microecosystems composed of polysaccharide matrices with embedded algae, bacteria, and other microbes

PFAS alter biofilm species composition

The shift in biofilm community composition can lead to changes in nutrient quality for sediment grazers

Biofilm image: Prof. S. Gerbersdorf, University of Stuttgart

Munoz *et al*. (2018); Liu *et al*. (2016); Ahrens *et al.* (2014); Sabater *et al*. (2007).

Concentration by Biofilms

Distribution of PFAS between water, sediment, and biofilms

- BCFs varied inversely with dissolved PFAS levels, pointing to concentration-dependent accumulation.
- Biofilm community characteristics may also be an influential determinant of PFAS bioconcentration – for example, carbon/nitrogen ratios.

PFAS	Log Bioconcentration Factor (BCF)
PFOA	2.1-3.0
PFNA	3.1-4.3
PFDA	2.1-4.1
PFHxS	1.2-2.2
L-PFOS	2.8-3.9
6:2 FTSA	1.0-4.1

Munoz *et al*. (2018).

Benthic Invertebrates

Accumulation at lower trophic levels leads to contamination in higher organisms

- Amphipods, copepods
- Gastropods
- Worms
- Insect larvae

Bioaccumulation by Benthic Invertebrates

Bioaccumulation in oligochaete *Lumbriculus variegatus* (Lasier *et al*. 2011):

- Perfluoroalkyl sulfonates ≥ 4 carbons
- Perfluoroalkyl carboxylates ≥ 7 carbons

Higgins *et al*. 2007: Higher BSAF values, but similar trends. Also found N-EtFOSAA (a PFOS precursor) accumulation and biotransformation in *L. variegatus* tissues – to PFOS and other PFOS precursors.

PFAS	Biota-Sediment Accumulation Factor (Lasier *et al*. 2011)
PFHpA	0.18 ± 0.18
PFOA	0.07 ± 0.03
PFNA	0.20 ± 0.12
PFDA	0.24 ± 0.12
PFUnDA	0.30 ± 0.12
PFDoDA	0.34 ± 0.14
PFTrDA	0.62 ± 0.24
PFTeDA	0.62 ± 0.24
PFBS	**0.31 ± 0.13**
PFOS	**0.59 ± 0.20**

Lasier *et al*. (2011); Higgins *et al*. (2007).

Bioaccumulation by Benthic Invertebrates

Bioaccumulation in midge larvae *Chironomus riparius* (Bertin *et al*. 2014):

- PFOS;
- Long-chain PFCAs, >10 carbons: PFUnA, PFTrDA, PFDoA, and PFTeDA);
- Precursor (FOSA); and
- Fluorotelomer (6:2 FTSA).

Bertin *et al*. 2018 and Wen *et al*. (2016): bioconcentration in *C. riparius* dependent on sediment concentration

PFAS	Biota-Sediment Accumulation Factor (Bertin *et al*. 2014)
PFUnDA	0.020
PFDoDA	0.004
PFTrDA	0.042
PFTeDA	0.004
PFOS	**0.023**
FOSA	**0.018**
6:2 FTSA	**0.098**

Bertin *et al*. (2018, 2014); Wen *et al*. (2016).

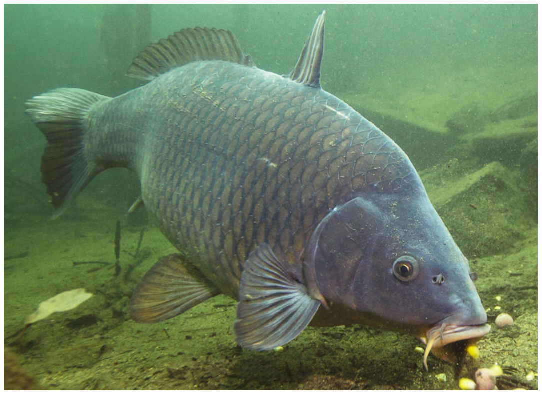
Trophic Transfer and Magnification

Different ecosystems contribute to variations in PFAS exposure and bioaccumulation

- Estuary: wide variations in freshwater discharge, salinity, sediment resuspension; smaller particle size, higher organic content
- Ocean: larger particles, more uniform salinity
- Bayou: most stagnant, longer exposure times
- River: larger particle size/smaller surface area, lower organic content, more oxygenated, higher flow
  Rate moves dissolved PFAS
  downstream of point sources

Trophic Transfer and Magnification

Bottom-feeding fish: Fang *et al*. (2016): Carp (*C. carpio*) exposed to perfluoroalkyl acids



- BAFs from free PFAAs in water: 3.85 to 97,000 L/kg
- BAFs that include contribution of suspended particulate matter: 3.61 to 600 L/kg
- Suspended particulate matter is an important source of exposure to long-chain PFAS

Trophic Transfer and Magnification

Geotechnical Environmental Ecological Water Construction Management

Bottom-feeding bird: Larson *et al*. (2018): For aquatic-dependent birds, *"Sediment-associated PFAS, rather than water-associated PFAS, were the source of the highest predicted PFAS exposures, and are likely to be very important for understanding and managing AFFF [firefighting foam] site-specific ecological risks."*

Risk Assessment Activities Needed

- Understanding the relationship between sediment biota and PFAS chain length, functional group, and degree of fluorination.
- Understanding the uptake and excretion rates of PFAS by different organisms, including competitive uptake and selective bioaccumulation at different trophic levels.
- Understanding the biotransformation of polyfluoroalkyl substances to perfluoroalkyl substances (the precursor issue).

Summary

- Thousands of PFAS with unknown bioaccumulation factors.
- K_{ow} is not a useful estimate of PFAS bioaccumulation.
- Studies of sediment grazers do not always find trends in accumulation based on PFAS chemistry – trends may be based on sediment PFAS concentrations, which depend on the chemical interactions between sediment, water, and PFAS.

Questions?

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