PFAS Transport in the Vadose Zone: Implications for Managed Aquifer Recharge

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University of Arizona

Webinar
Groundwater Protection Council

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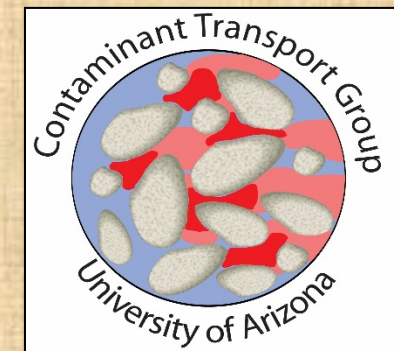

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Superfund Research Program
The University of Arizona

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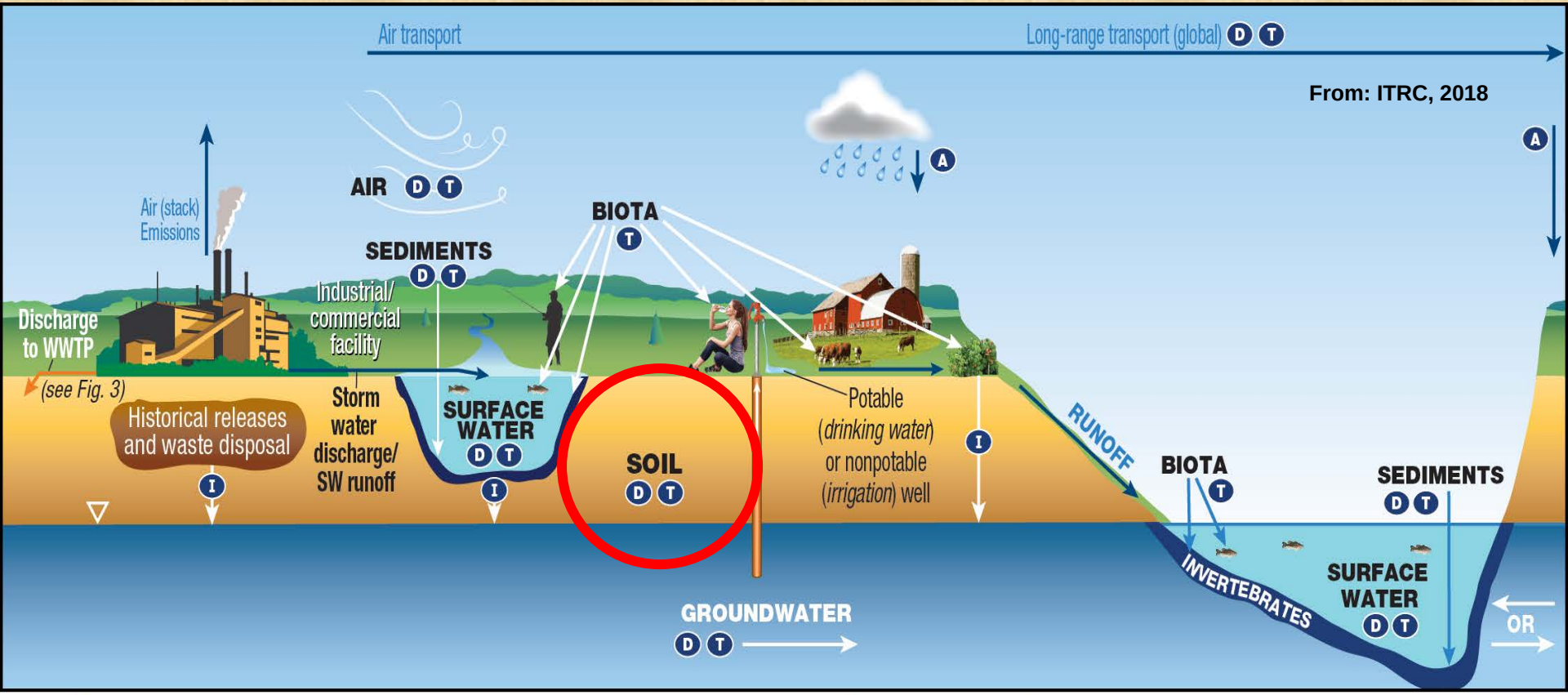
PFAS Transport & Fate in the Environment

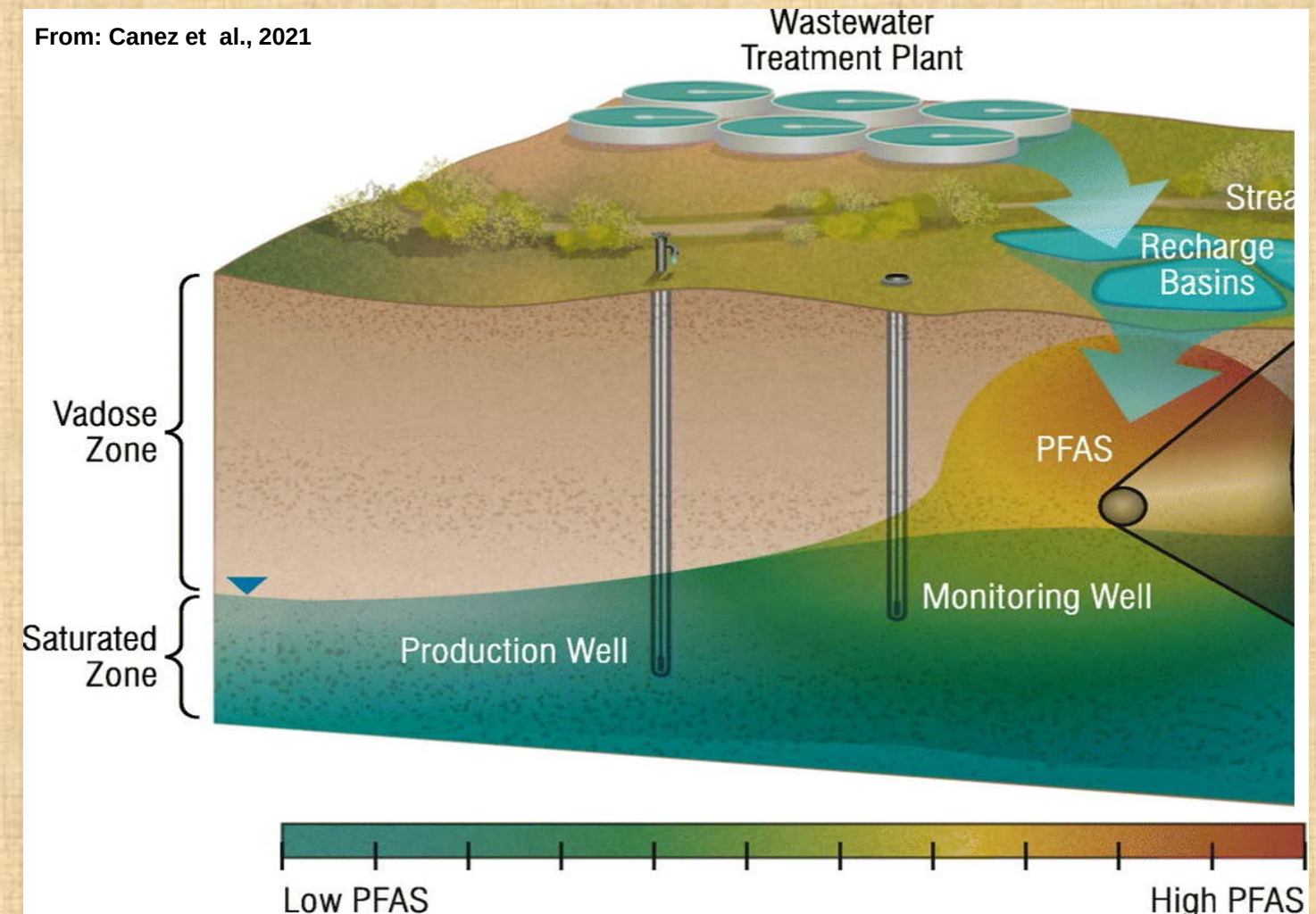
>>> Numerous Sources of Release to:

- -atmosphere
- -surface water
- -soil
- -groundwater



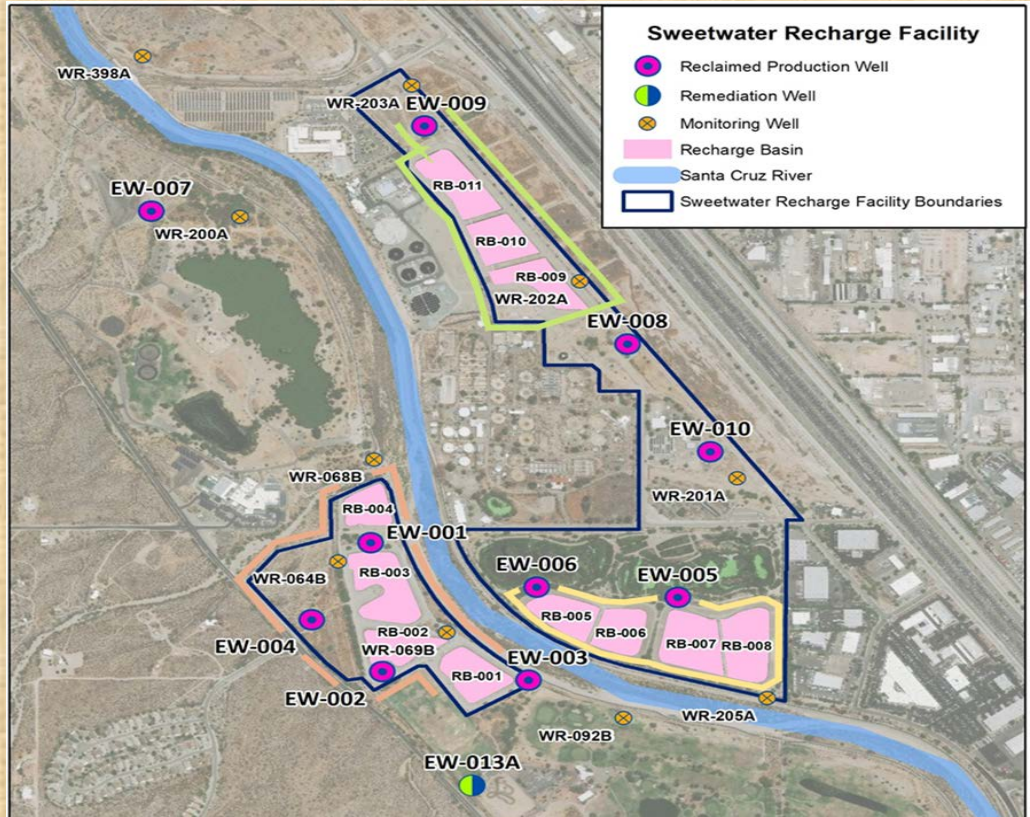
PFAS at MAR Facilities

Potential Impact: Leaching of PFAS from recharge basin, through vadose zone, to groundwater



Case Study: Sweetwater Recharge Facility

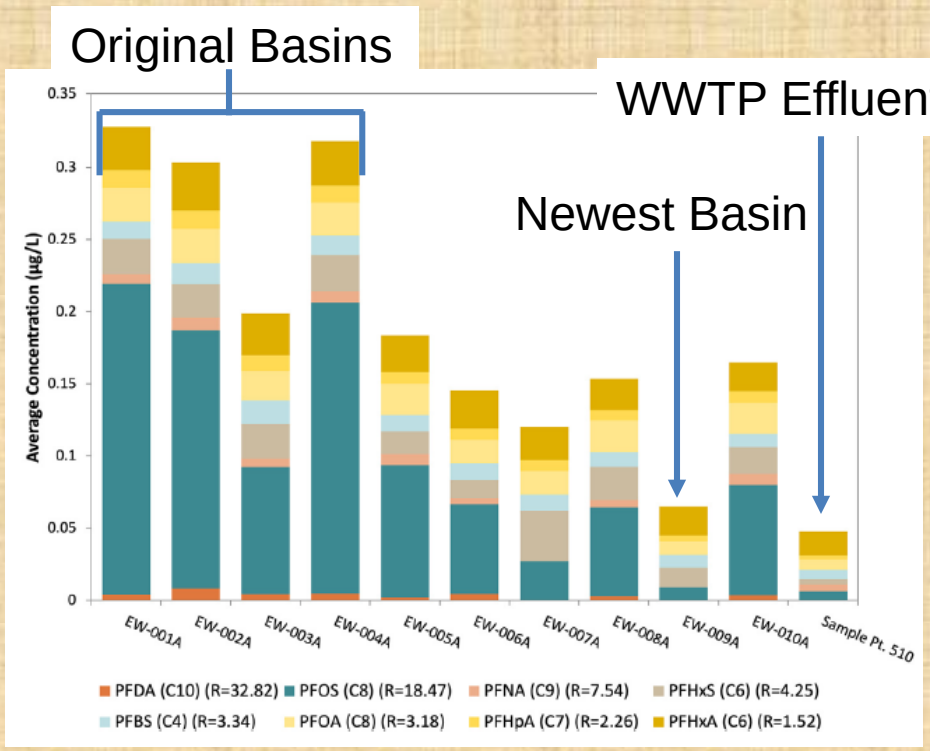
- Recharge of treated municipal wastewater
- Permitted to recharge 1.6 M m^3/year
- Started operations in 1989
- Received treated wastewater from original WWTP until 2014
- New WWTP in operation from 2014
- Old WWTP=secondary treatment
- New WWTP=tertiary treatment
- Depth to GW: 42-48 m
- Estimated transit time to GW: 30 d
  [high variability]



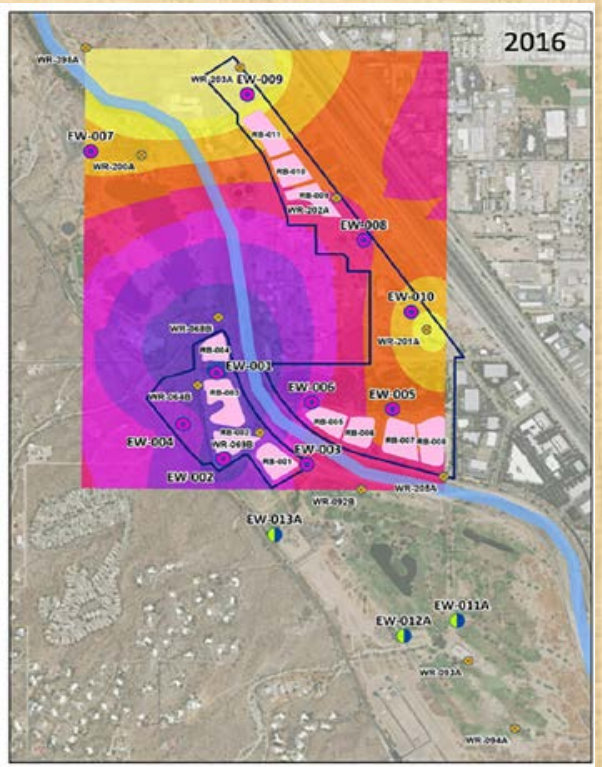
From: Canez et al., 2021

Case Study: Sweetwater Recharge Facility

- PFOS is present in highest concentrations
- Groundwater concentrations are higher than current WWTP effluent concentrations
- Groundwater concentrations are highest for the original basins, which received the most wastewater from the original plant
- Groundwater concentrations are lowest for newest basin, which has received wastewater only from the new WWTP

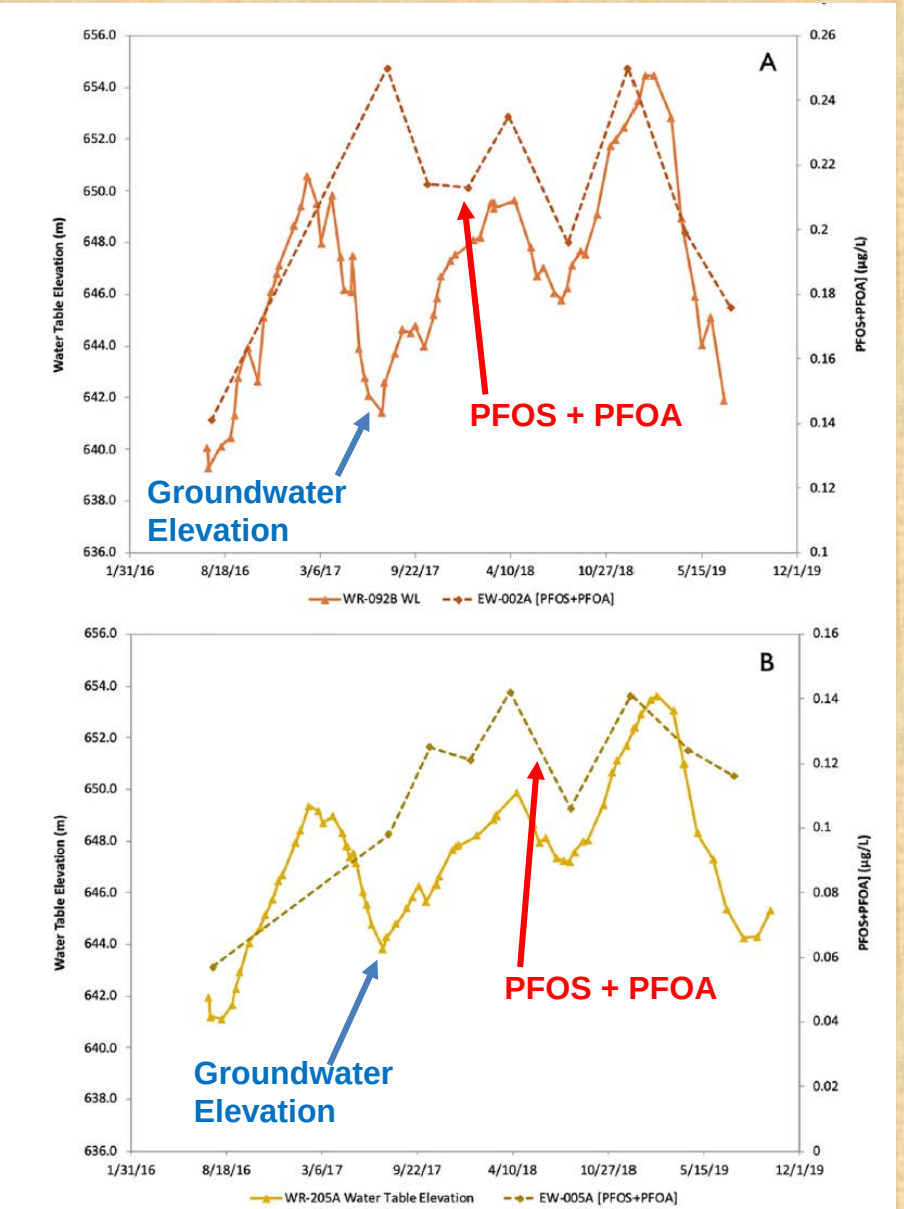


PFOS



From: Canez et al., 2021

Case Study: Sweetwater Recharge Facility



Groundwater Level Fluctuations

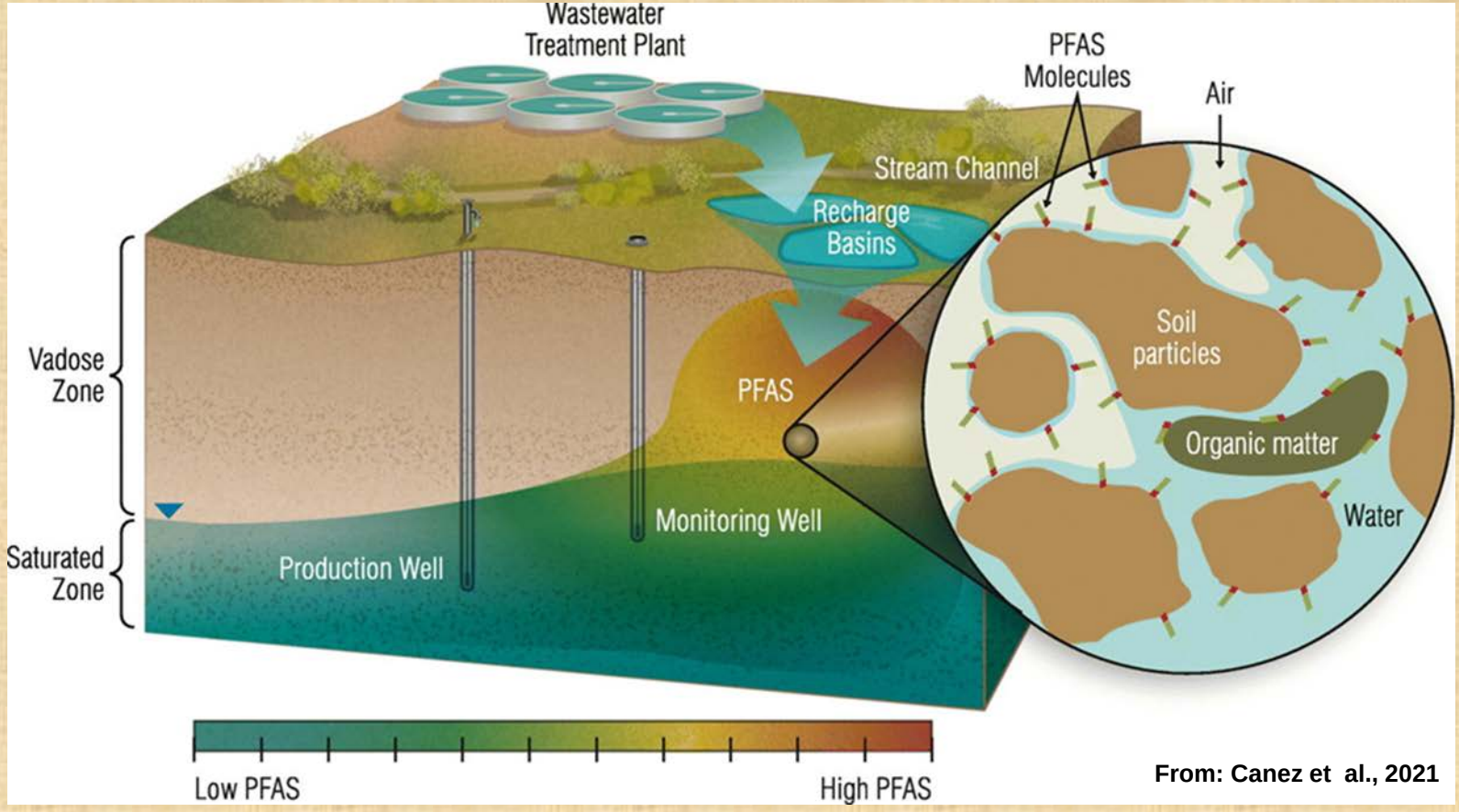
- Correlations between groundwater levels and PFAS concentrations for several wells
- Possible indication of leaching of PFAS present in vadose zone
- On-going study to investigate PFAS distribution and migration within recharge basin

From: Canez et al., 2021

PFAS at MAR Facilities

Assessing Impacts and Evaluating Mitigation Actions:

Understand the processes influencing PFAS retention and leaching in the vadose zone

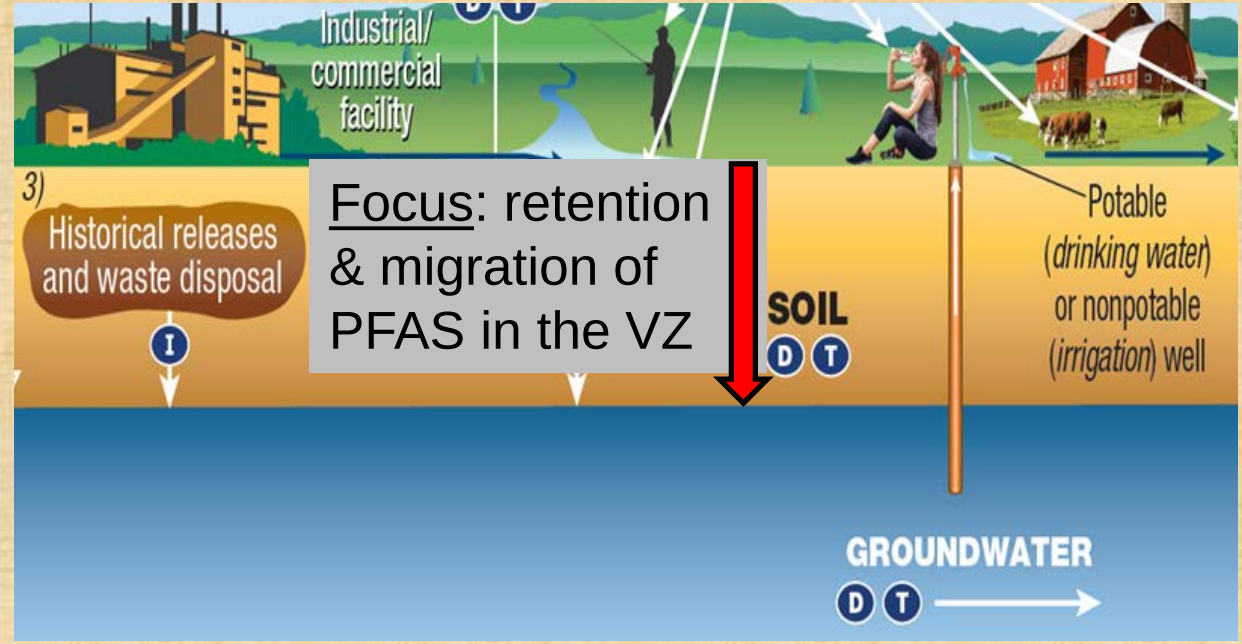


From: Canez et al., 2021

PFAS in Soil & Vadose Zone

Critical questions to address:

- How are PFAS retained in the vadose zone? [retention processes]
- How long are PFAS retained in the vadose zone? [magnitude of retention & leaching potential]
- What is the magnitude of mass discharge to groundwater? [leaching rates]



PFAS migration in the vadose zone is a function of several factors:

- Source type
  - e.g., AFFF sites vs biosolids/wastewater-application sites
  - types of PFAS and relevant concentration ranges
- Site conditions
  - Soil properties (sorptive constituents, air-water interfacial area)
  - Physical and geochemical heterogeneity
  - Potential precursor presence and transformation
  - Presence of other contaminants
- Precipitation/Evapotranspiration/Infiltration
  - Infiltration-recharge dynamics
- **Transport & Retention processes**
  - **Solid-phase sorption**
  - **Adsorption at air-water interfaces**
  - **Impact of infiltration-recharge dynamics on retention and transport**

Simplified Retention Analysis

- Retardation Factor for aqueous-phase transport of PFAS influenced by solid-phase adsorption and air-water interfacial adsorption:

Porewater Sorbed Adsorbed at air-water interface

$$R = 1 + K_d \rho_b/\theta_w + K_i A_i/\theta_w$$

K_d = solid-phase adsorption coefficient

K_i = air-water interfacial adsorption coefficient

A_i = air-water interfacial area

ρ_b = bulk density of porous medium

θ_w = volumetric water content

- Retention is a function of:
  - Properties of PFAS
  - Properties & conditions of the soil

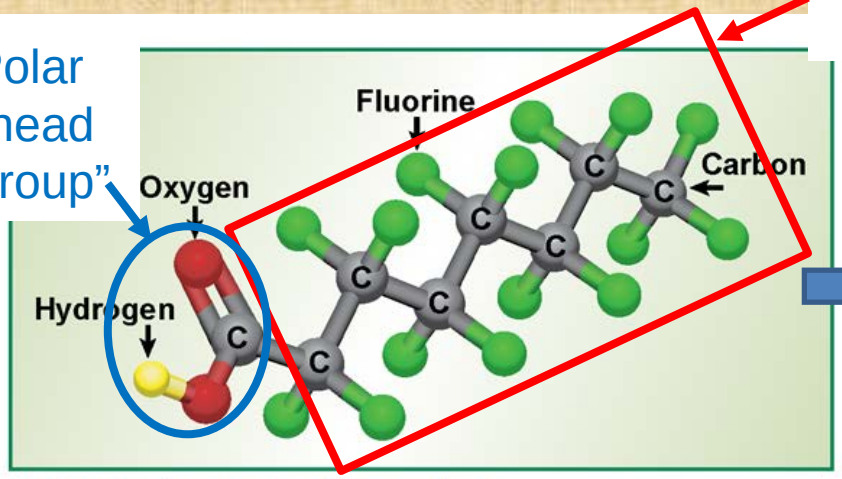
PFAS Properties

Most PFAS are **amphiphilic** (contain both nonpolar & polar regions)

***behave as surfactants

Nonpolar "tail"– fluorinated carbon chain rather than hydrogenated

Polar "head group"



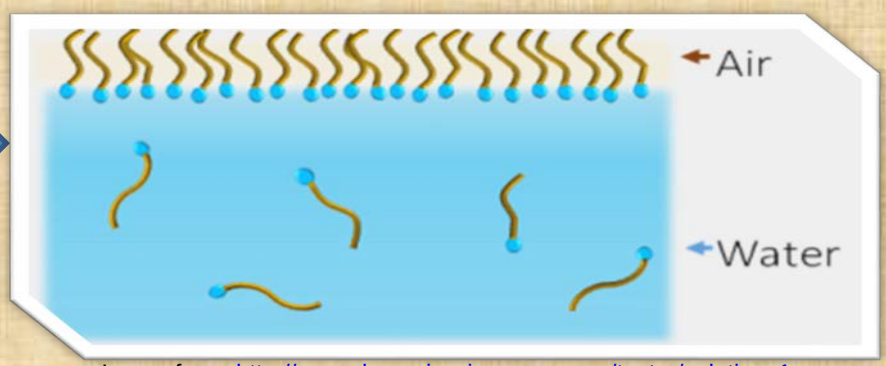


Image from: http://www.dynamicscience.com.au/tester/solutions1/chemistry/foodchemistry/emulsions.htm

PFAS chains are both hydrophobic and oleophobic

*Provides water and oil repellency

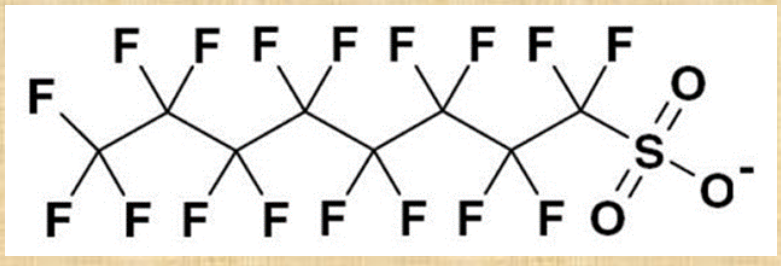
>>special attributes that make PFAS useful for many applications

>>causes transport to be complex

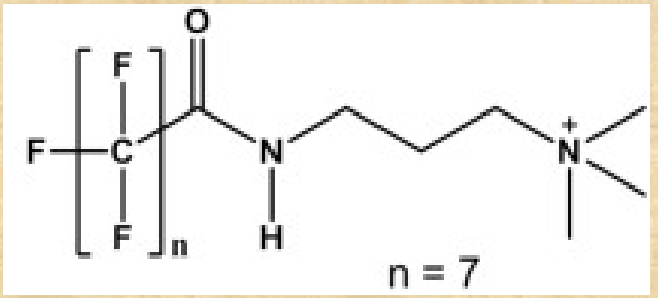
PFAS Structures

- PFAS have different types of **surfactant headgroups**

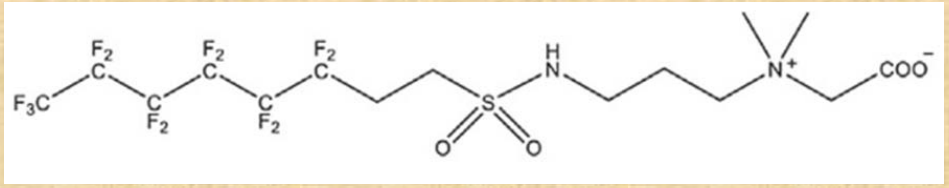
Anionic



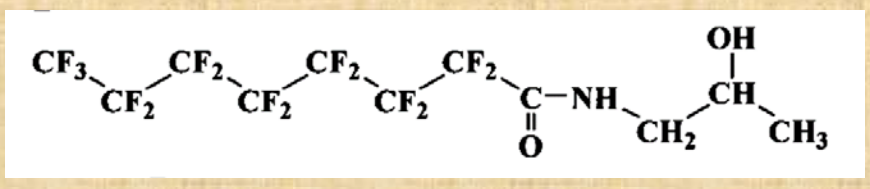
Cationic



Zwitterionic



Nonionic



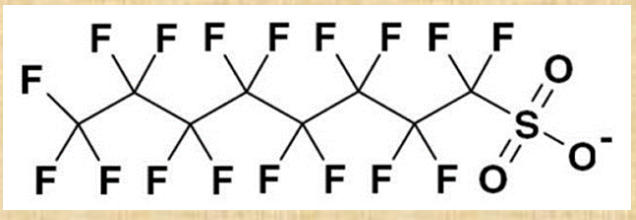
PFAS Structures

PFAS have different **tail structures**

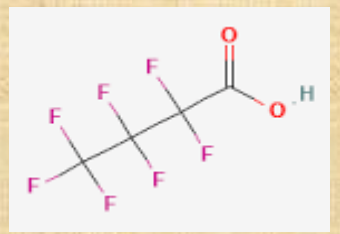
- Per vs Poly
- Straight-chained vs branched
- Different chain lengths

Perfluoroalkyls

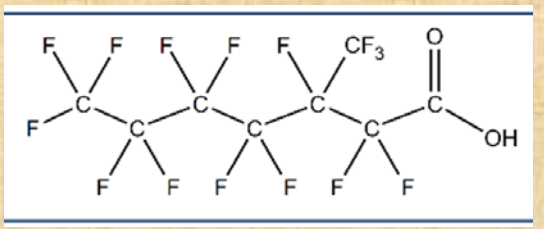
Straight long-chained



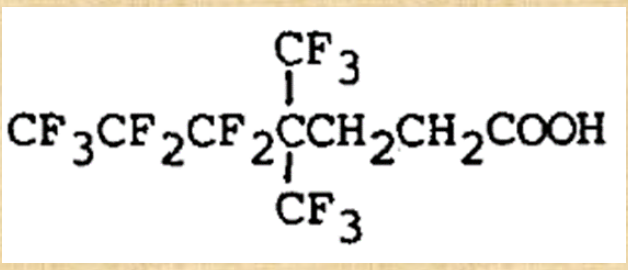
Straight short-chained

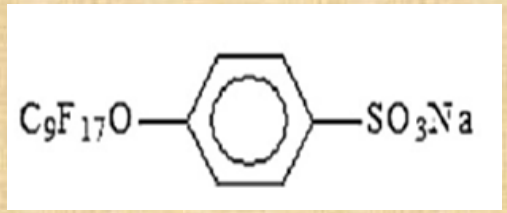


Branched



Polyfluoroalkyls





QUESTIONS

Solid-phase Sorption

- Sorption of PFAS by soil, sediment, and aquifer material (geomedia) is complex
- Function of PFAS molecular structure and the geochemical properties of the geomedia

Geomedia are geochemically heterogeneous

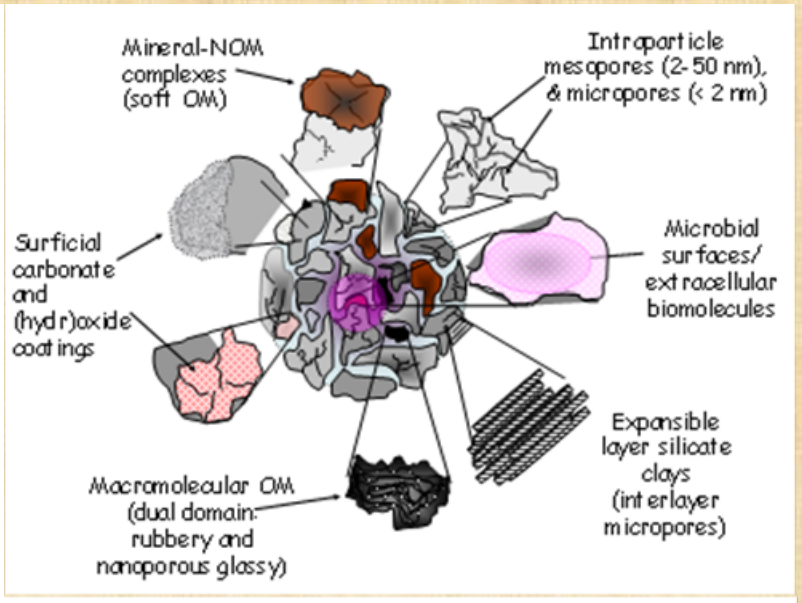
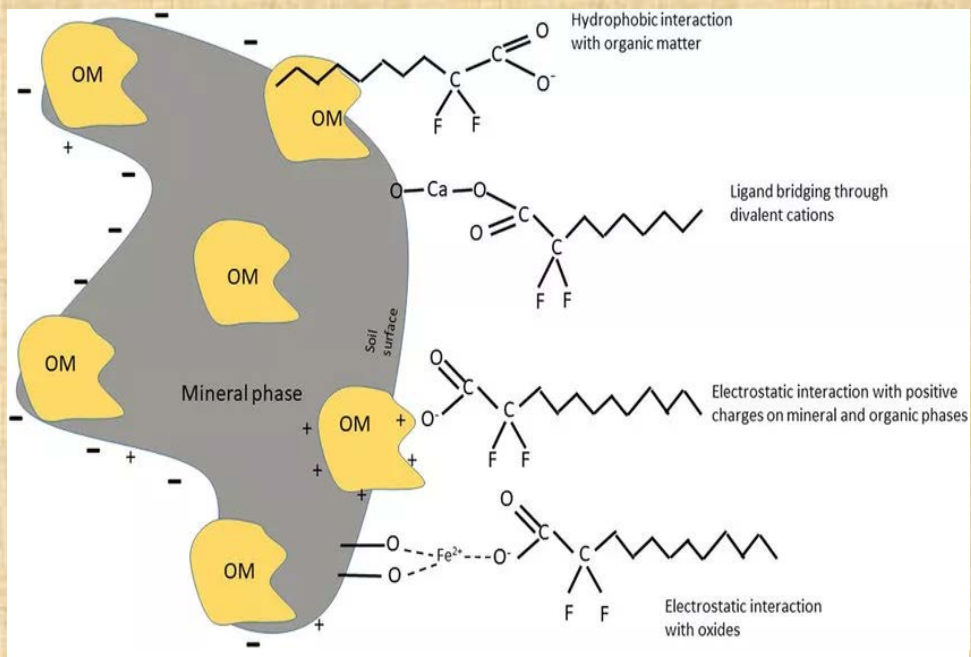


Figure A. Biogeochemically-reactive solid-water interfaces present in natural and waste-impacted geomedia (from Chorover and Brusseau, 2008)

Multiple Sorption Mechanisms



From: Li, Y., Oliver, D.P., Kookana, R.S., 2018. Sci. Total Environ. 628/629, 110-120

QSPR Analysis

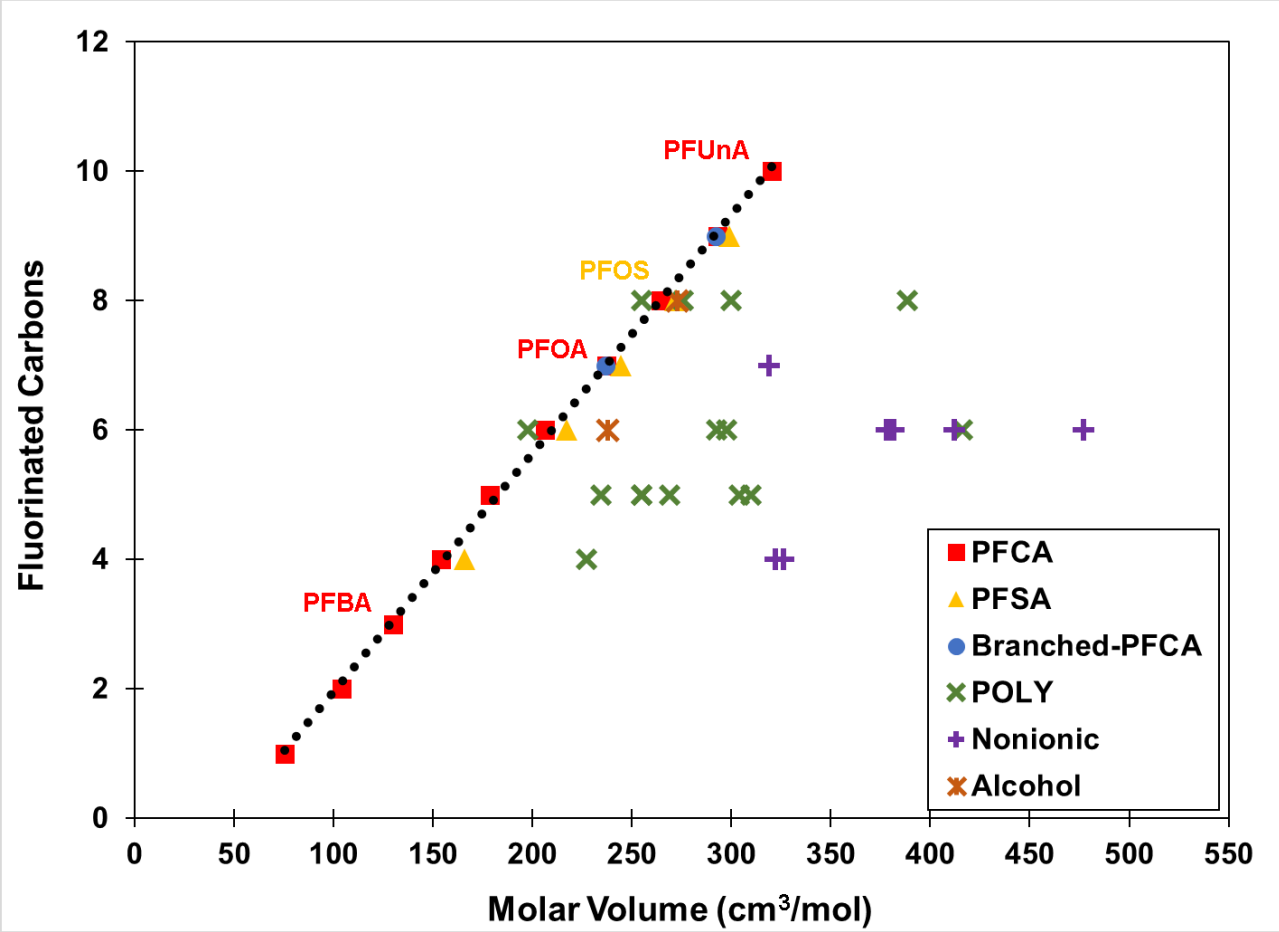
- QSPR = quantitative-structure/property-relationship analysis
- Empirical approach to estimating properties and parameters based on molecular descriptors
- Example common descriptors
  - Number of Carbon atoms
  - Number of Fluorinated Carbons
  - Molar Volume--- represents volume in solution occupied by molecule
- Use QSPR to characterize partitioning behavior

QSPR Analysis

- Fluorinated carbons- commonly used
  - Works for PFCAs & PFSAs
  - Not for more complex PFAS structures

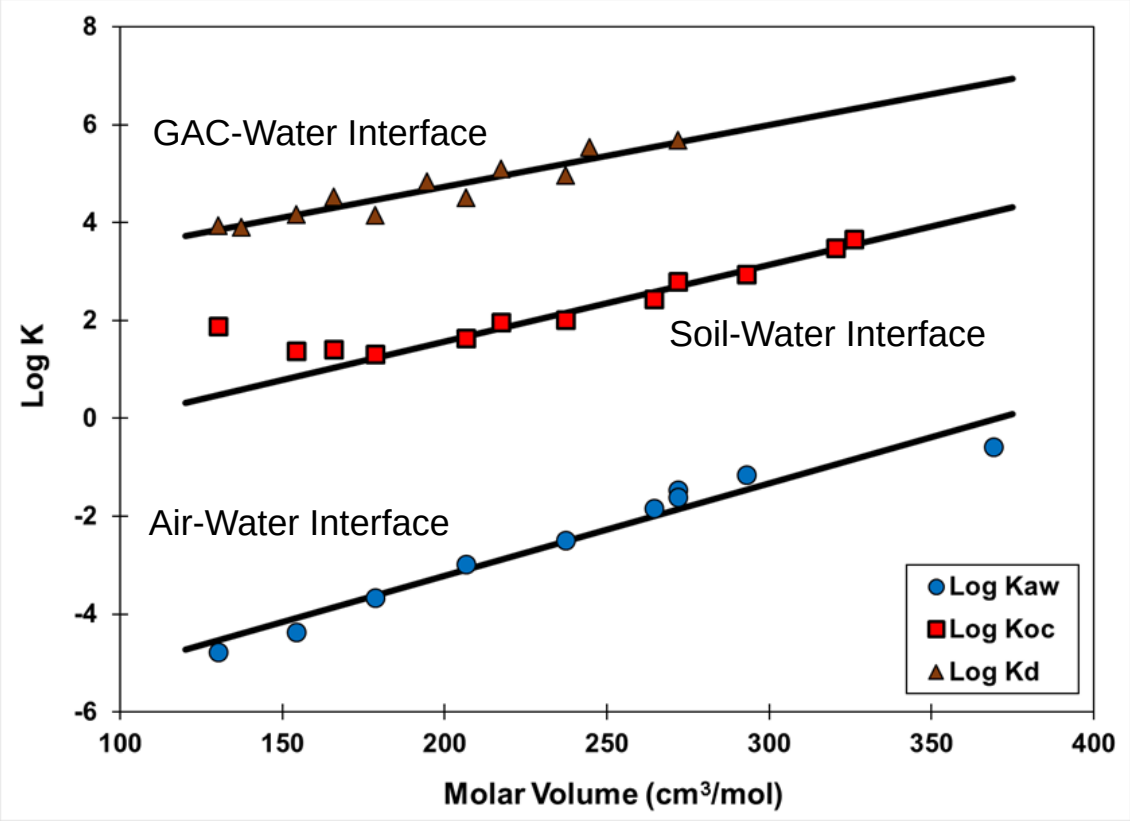
- Molar Volume is more representative

Molar volume can be determined from molar-mass/density



Air-Water Interface = model interface for investigating PFAS molecular structure impacts on partitioning [most physically & geochemically homogeneous]

Uniform log-linear relationship indicative that hydrophobic interaction serves as primary driving force for partitioning



From: Brusseau, 2019

Quantifying PFAS Sorption

- Standard approach for characterizing sorption of hydrophobic organic contaminants-

$$K_d = K_{oc} \times f_{oc}$$

K_d = equilibrium sorption coefficient
K_{oc} = organic-carbon normalized sorption coefficient
f_{oc} = fraction of organic carbon

- **Question**: is this approach representative for PFAS?

Meta-Analysis of PFAS Sorption

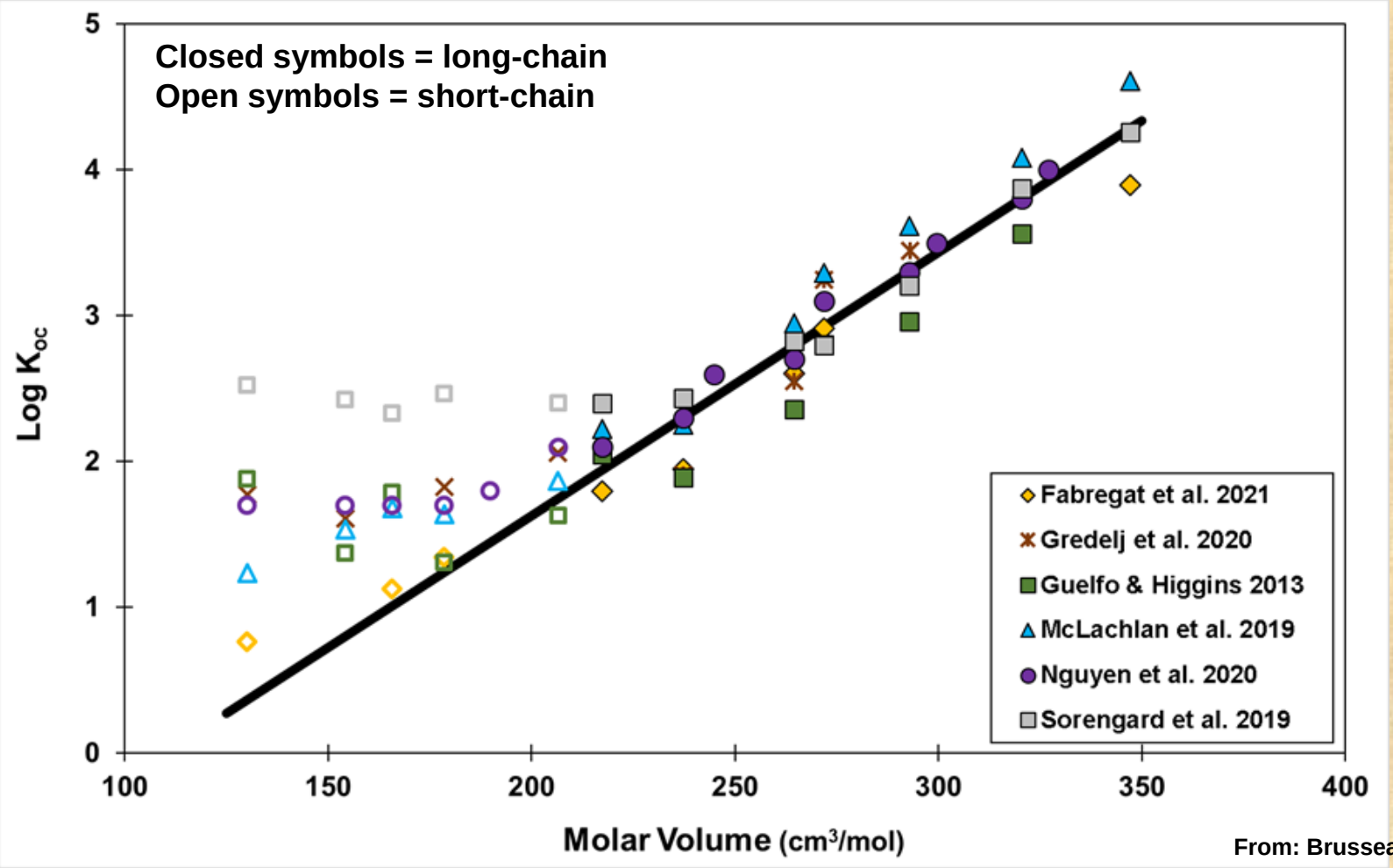
- Integrated QSPR analysis of the differential sorption of short-chain versus long-chain anionic PFAS

- QSPR = quantitative-structure/property-relationship analysis, an empirical approach to characterize partitioning/adsorption behavior

- 11 Studies:
  - Total of 65 soils & freshwater sediments
  - wide range of organic carbon, silt+clay, pH
  - 16 PFAS (9 perfluorocarboxylic acids & 7 perfluorosulfonic acids)

- Log K_{oc} values for short-chain PFAS deviate from regression representing long-chain PFAS

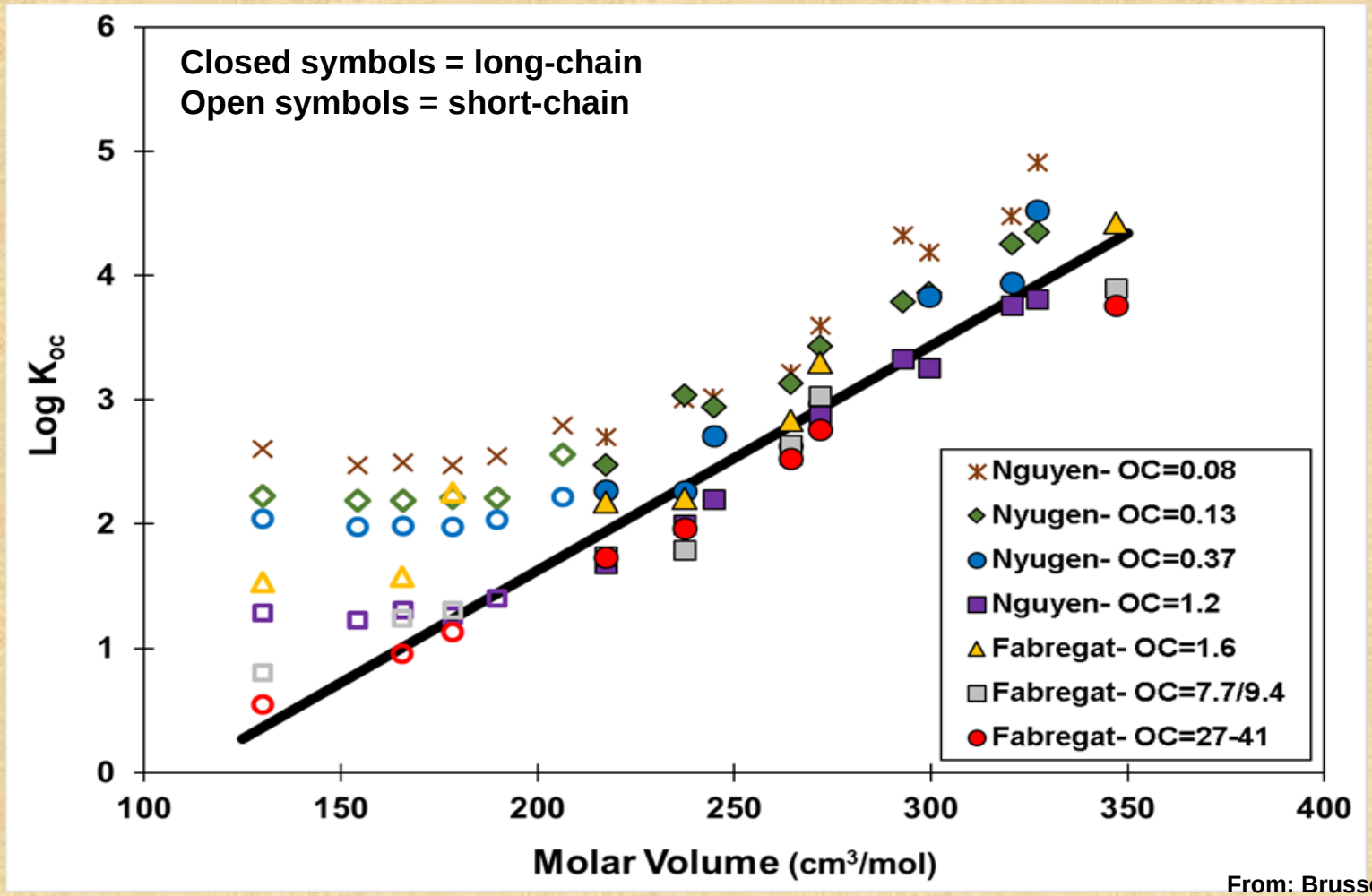
- "Enhanced" sorption of short-chain PFAS



From: Brusseau, 2023c

- Deviations for short-chain PFAS are greater for lower organic-carbon contents

- Sorption of short-chain PFAS mediated by additional soil components (clay minerals, metal-oxides)



From: Brusseau, 2023c

- Deviations for short-chain PFAS are greater for high silt+clay content

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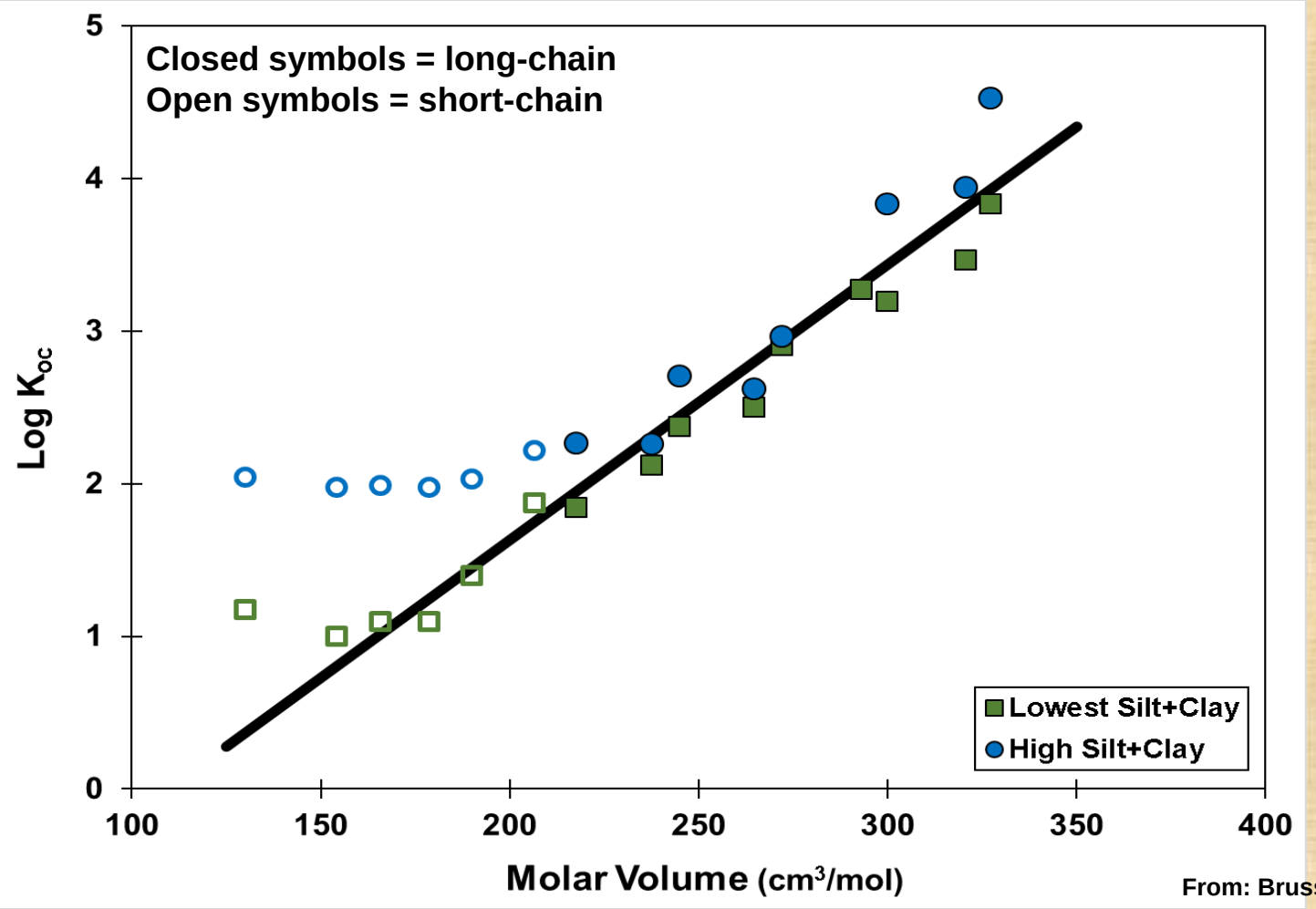
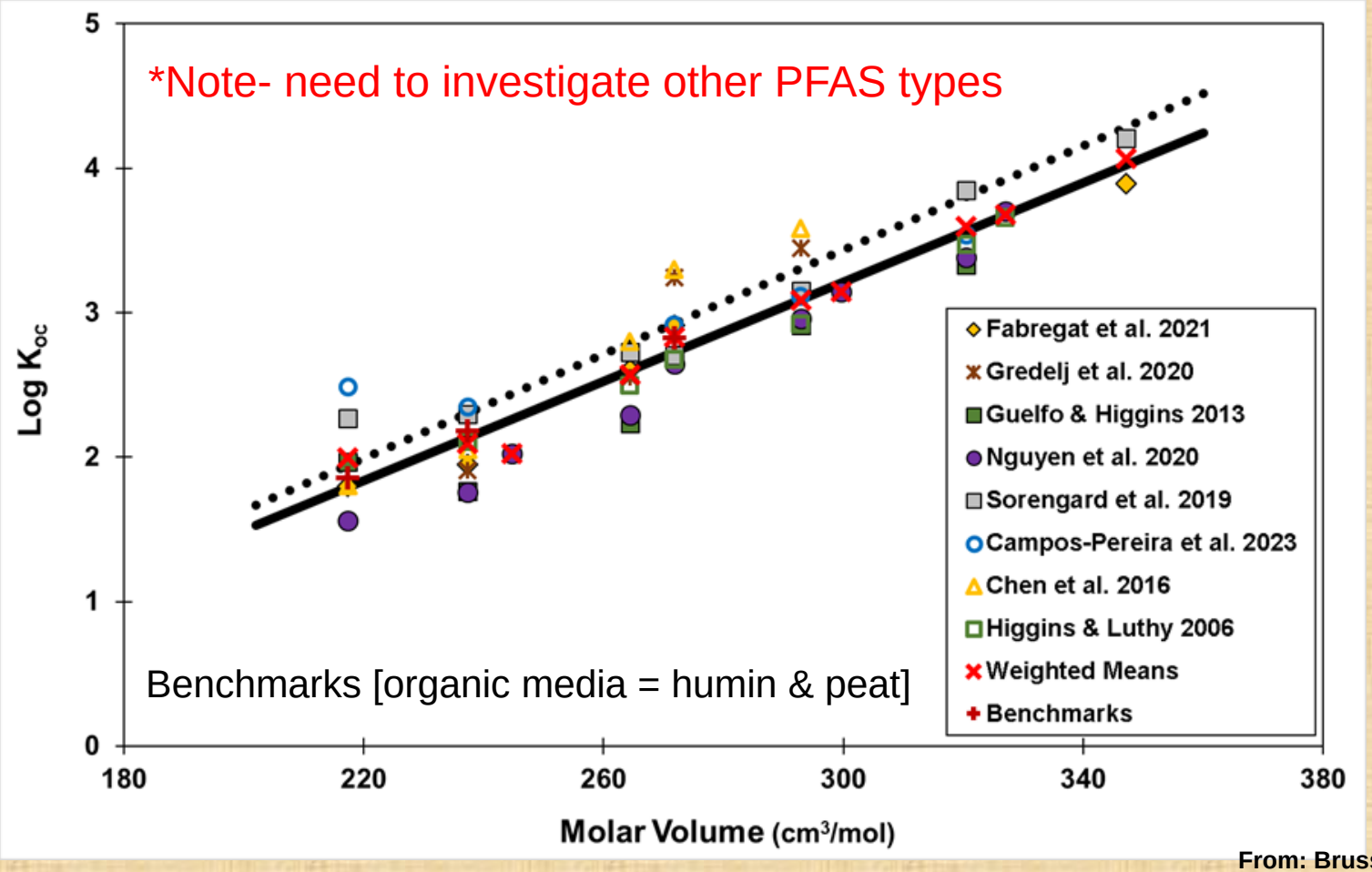


From: Brusseau, 2023c

Long-chain PFAS for soils & sediments with organic-carbon content > 1%

K_{oc} approach may be reasonable for long-chain PFAS for soils with OC>1%

Not for short-chain PFAS



From: Brusseau, 2023c

Surfactant Behavior of PFAS

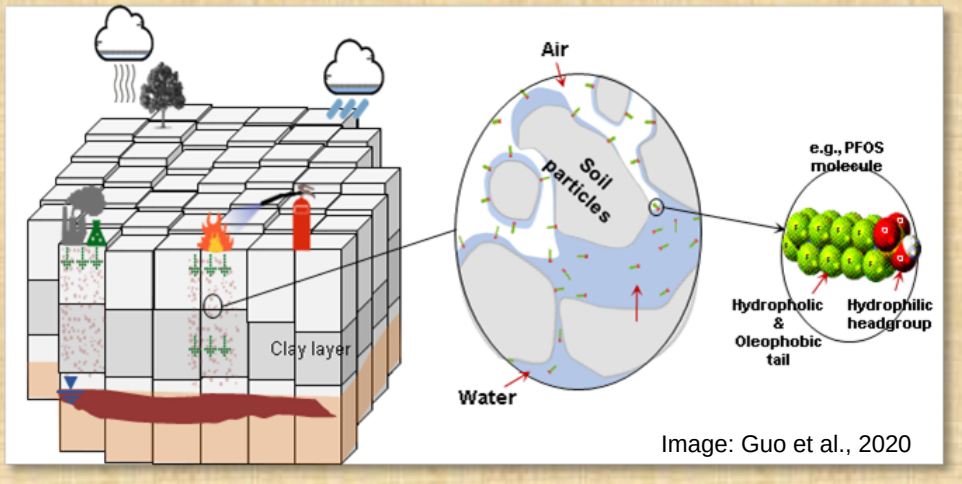


Image: Guo et al., 2020

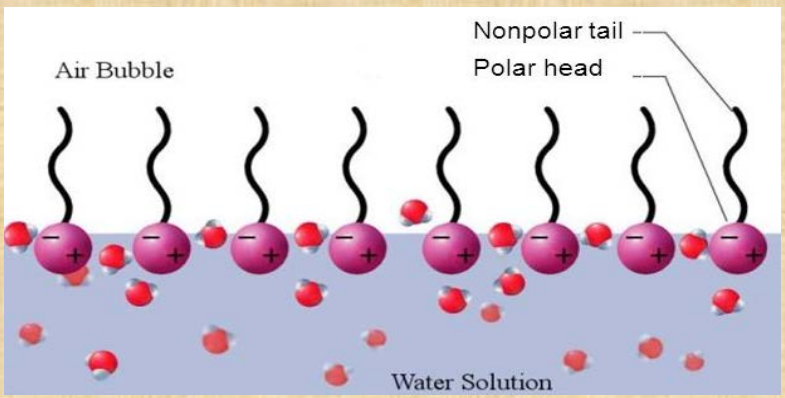
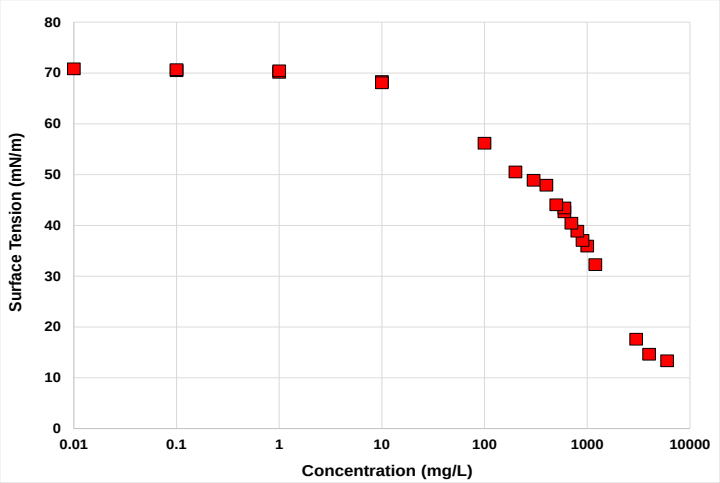


Image: https://en.wikipedia.org/wiki/Langmuir%E2%80%93Blodgett_film

- Adsorption at the air-water interface
- Ramifications:
  - Potential to cause surfactant-induced flow
  - Increased retention and retardation for transport

Surfactant-Induced Flow

Adsorption of PFAS at air-water interface reduces surface tension (σ)



>>> Unsaturated Porous Media

Changes in surface tension impact capillary pressure: $h = \frac{\sigma}{\sigma_0} h_0$

This causes water flow

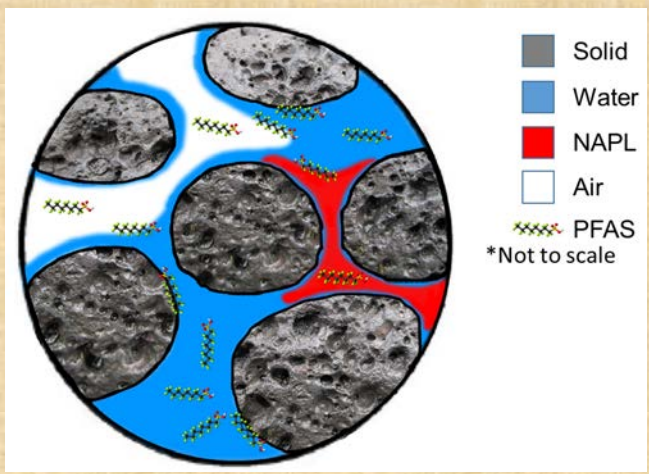
- **Surfactant-induced flow leads to:**
  - Transient flow
  - Impacts to solute transport
  - Changes in local water saturation
  - Changes in the magnitude of air-water interfacial area
  - Impact on the magnitude of retention by air-water interfacial adsorption

>>> Complex, interconnected flow and transport behavior

Fluid-Fluid Interfacial Retention

- Transport in source zones is influenced by additional retention processes: >>>> ***this adds complexity***
  - **Adsorption at air-water interfaces in vadose zones**
  - **Adsorption at NAPL-water interfaces in NAPL source zones**

    [NAPL = chlorinated solvents, fuels]



From: Brusseau, 2018

Comprehensive Retention Model for PFAS

Phase	*Source Zone*[a]	*Plume*[b]
Aqueous[c]		
Sorbed by solid phase		
Vapor		
Adsorbed at air-water interface		
Adsorbed at air-NAPL interface		
Adsorbed at NAPL-water interface		
Absorbed by NAPL		

Relevant for vast majority of PFAS at essentially all sites

Relevant for many critical PFAS of concern at many sites

Relevant for select PFAS at some sites

Not relevant

From: Brusseau et al., 2019b

Simplified Retention Analysis

- Retardation Factor for aqueous-phase transport of PFAS influenced by solid-phase adsorption and air-water interfacial adsorption:

$$R = 1 + K_d \rho_b/\theta_w + K_i A_i/\theta_w$$

Magnitude of AWIA

K_d = solid-phase adsorption coefficient

K_i = air-water interfacial adsorption coefficient

A_i = air-water interfacial area

ρ_b = bulk density of porous medium

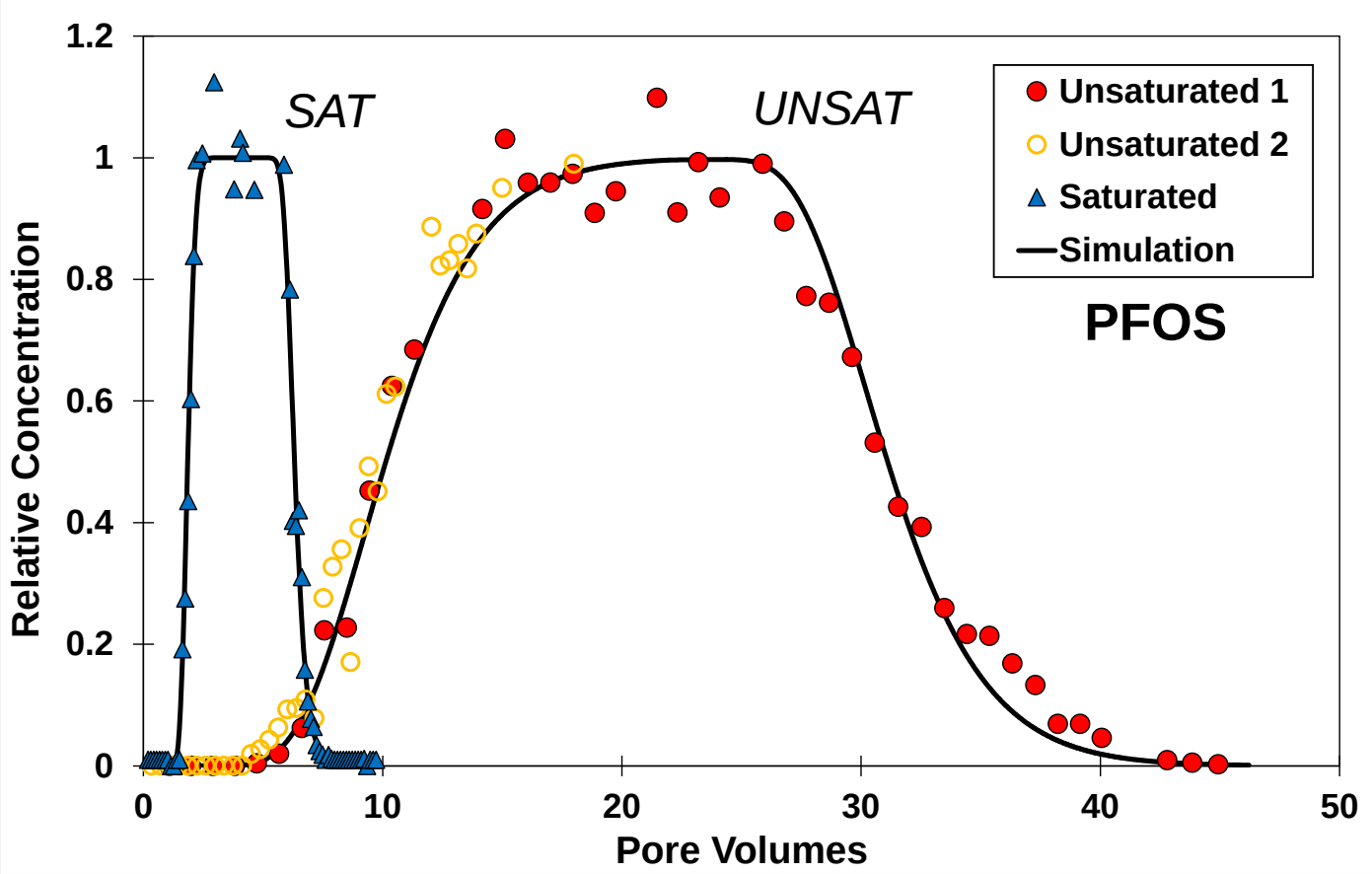
θ_w = volumetric water content

PFAS Transport Experiments

- PFAS transport: **unsaturated** conditions

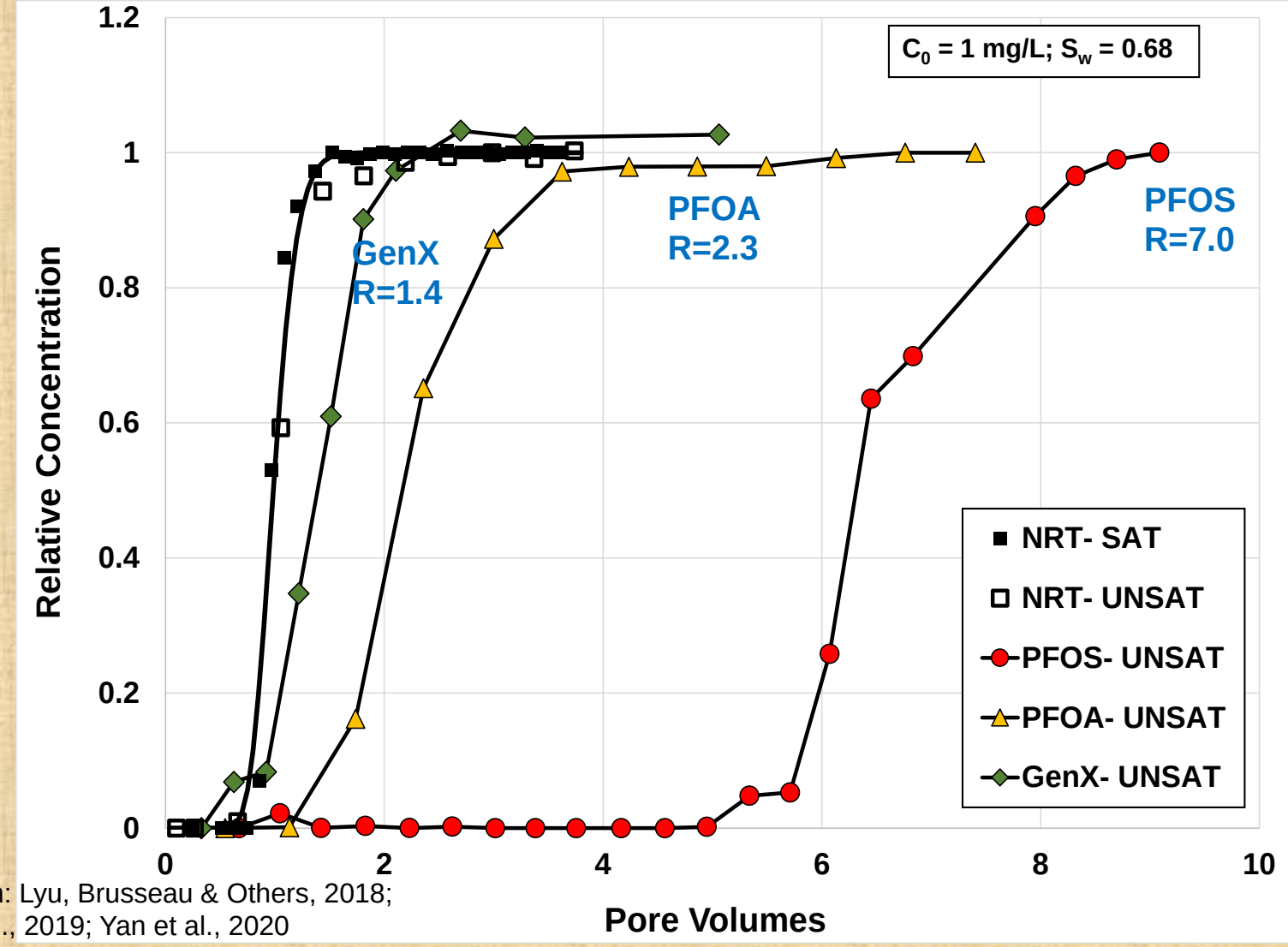
****Greater retardation for transport in unsaturated conditions; a result of adsorption at the air-water interface**

From: Brusseau et al. 2019, 2021



R and PFAS Molecular Structure

- **Retardation is larger for longer-chain PFAS**



Compiled from: Lyu, Brusseau & Others, 2018; Brusseau et al., 2019; Yan et al., 2020

Retention of PFAS

- Retardation Factor for aqueous-phase transport of PFAS influenced by solid-phase adsorption and air-water interfacial adsorption:

$$R = 1 + K_d \rho_b/\theta_w + \boxed{K_i A_i/\theta_w}$$

Magnitude of AWIA

K_d = solid-phase adsorption coefficient

K_i = air-water interfacial adsorption coefficient

A_i = air-water interfacial area

ρ_b = bulk density of porous medium

θ_w = volumetric water content

- **Air-water interfacial adsorption coefficient (K_i) is a function of:**

  - PFAS molecular structure

  - PFAS concentration (nonlinearity)

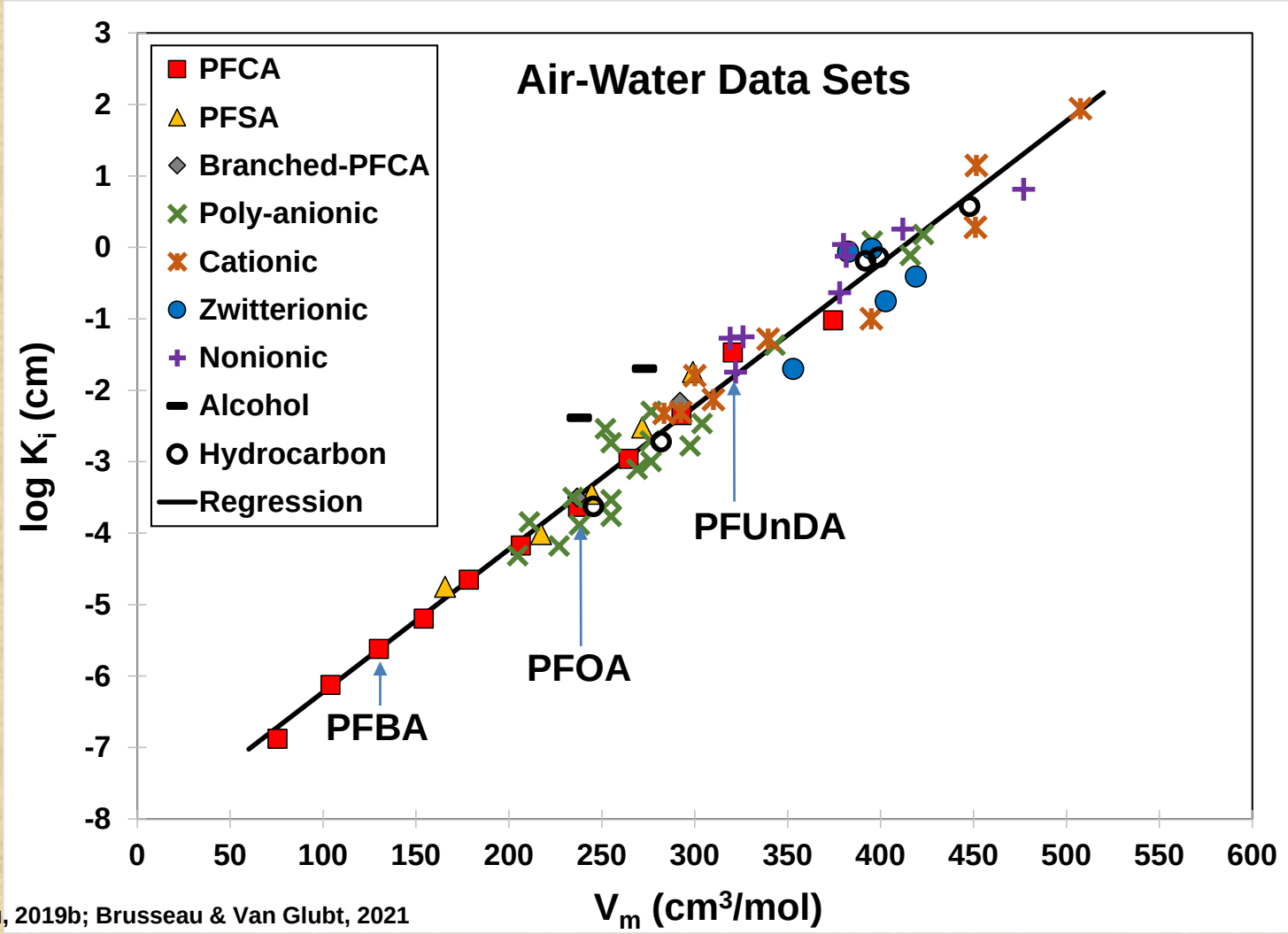
  - Solution composition

- **QSPR Meta-Analysis:**
  - 61 individual PFAS
  - All PFAS structure types
  - Hydrocarbon surfactants for comparison

K_i for PFAS

- K_i is larger for larger PFAS

Predictions representative for most PFAS structure types

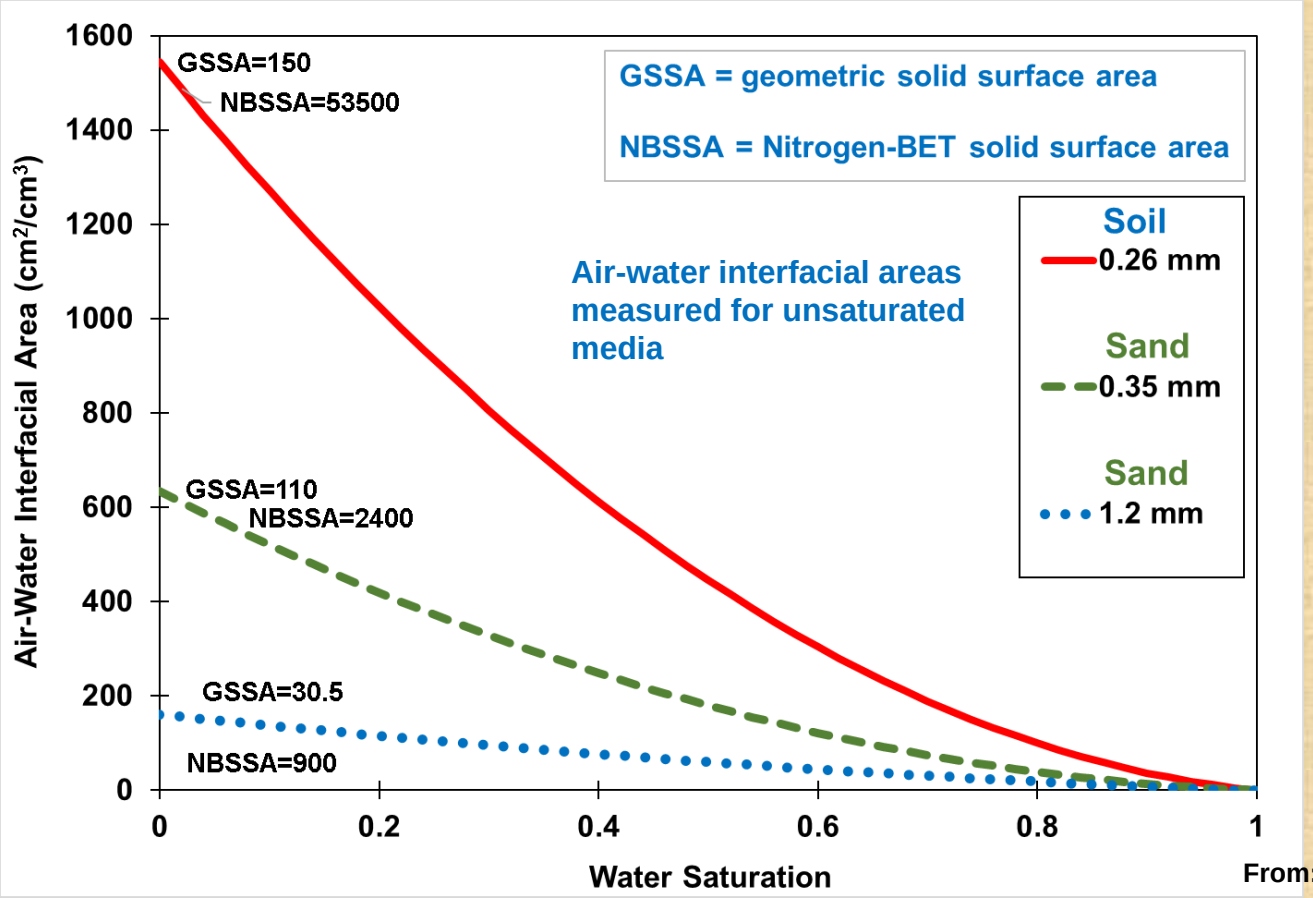


From: Brusseau, 2019b; Brusseau & Van Glubt, 2021

Air-water interfacial area (A_i) is a function of:

- Soil properties- Interfacial area is larger for media with smaller grains and larger solid-surface areas

- Water saturation- Interfacial area increases nonlinearly as wetting-fluid conten decreases



From: Brusseau, 2023a

Questions for Field-scale Applications:

- Is surfactant-induced flow relevant?
  - Unlikely to be significant for lower concentration ranges present at many sites
  - May be relevant under high-concentration conditions
- How to determine the $\mathbf{K_i}$?
  - QSPR estimation model appears reasonable for many PFAS
- How to determine the $\mathbf{A_i}$?
  - One of the most difficult parameters to characterize and quantify
  - Prediction models based on soil properties have been developed but need testing for a range of soils

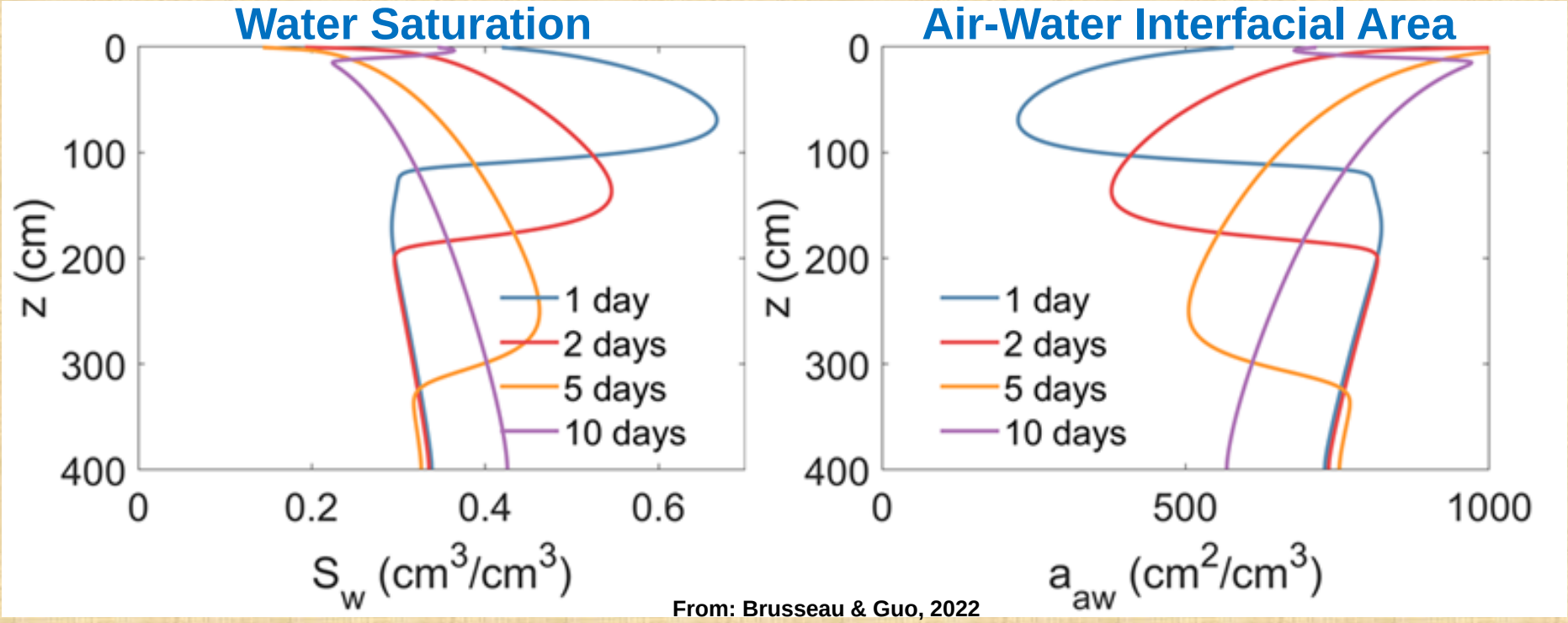
QUESTIONS

Dynamic Infiltration & Recharge Impacts

Simulation of a single precipitation-infiltration-redistribution event

-PFAS present in vadose zone after 30-year operation of FTA
-No PFAS input during 10-day precipitation event

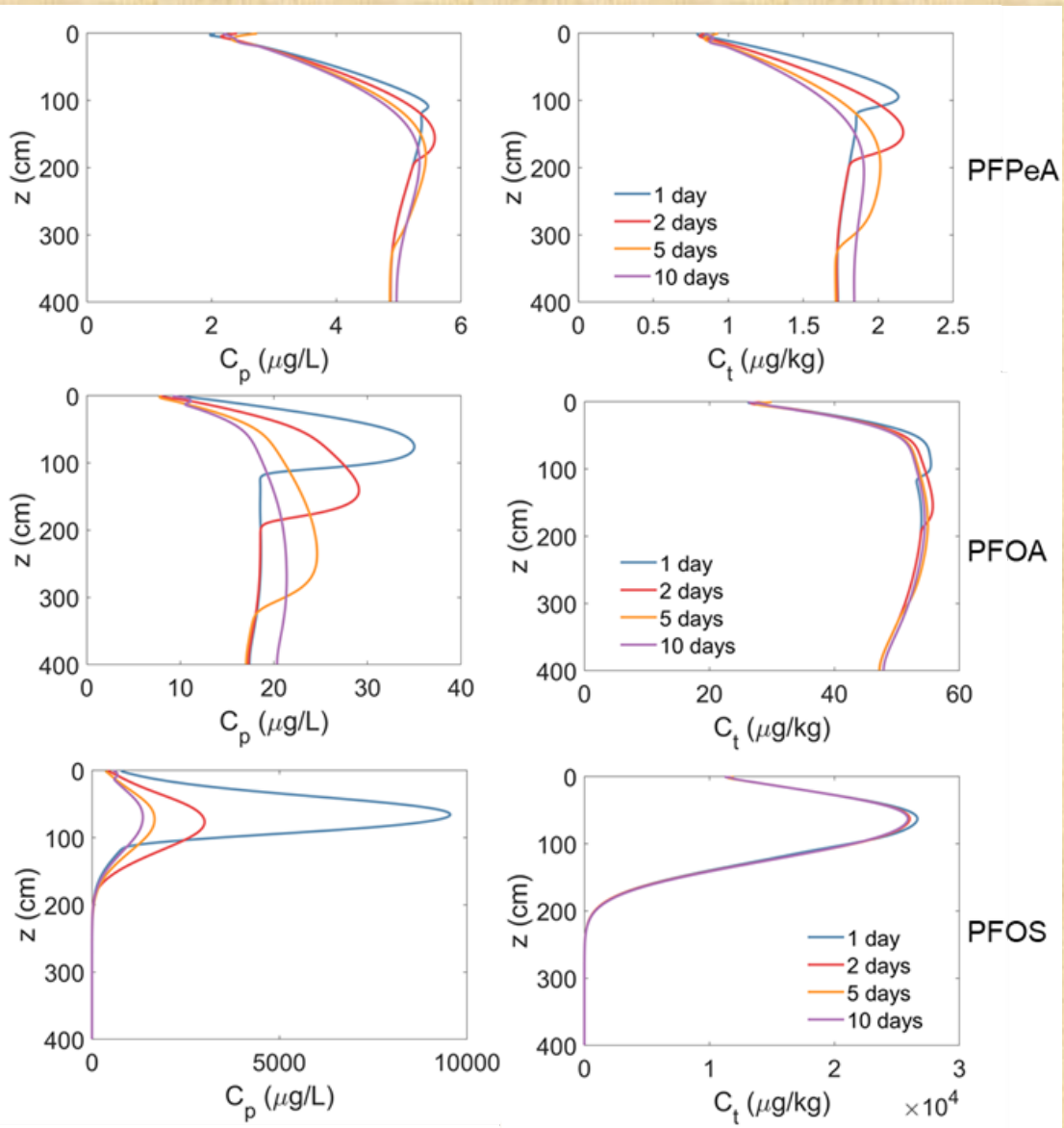
Changes in air-water interfacial area due to changes in water saturation



From: Brusseau & Guo, 2022

PFAS Retention Dynamics

Porewater Concentration **Total Soil Concentration**



- Porewater concentrations increase temporarily
- The change is greatest for PFOS (highest interfacial activity)
- Leaching is observed (change in total soil concentration)
- Leaching is greatest for PFPeA (lowest retention)
- Leaching is minimal for PFOS (highest retention)

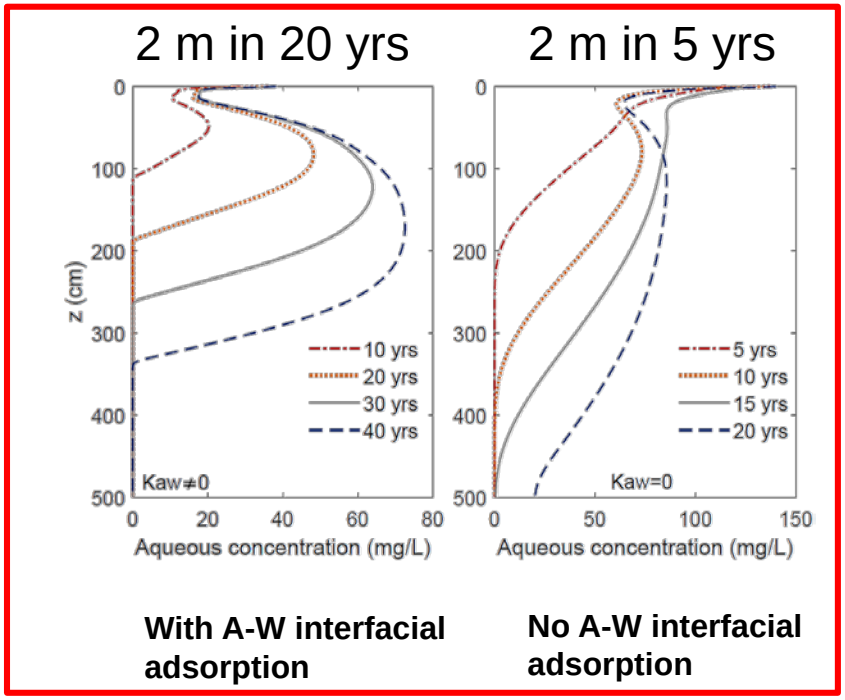
From: Brusseau & Guo, 2022

Long-term PFAS Migration in Vadose Zone

Temporal evolution of vertical profiles of PFOS (Vinton soil) at a FTA

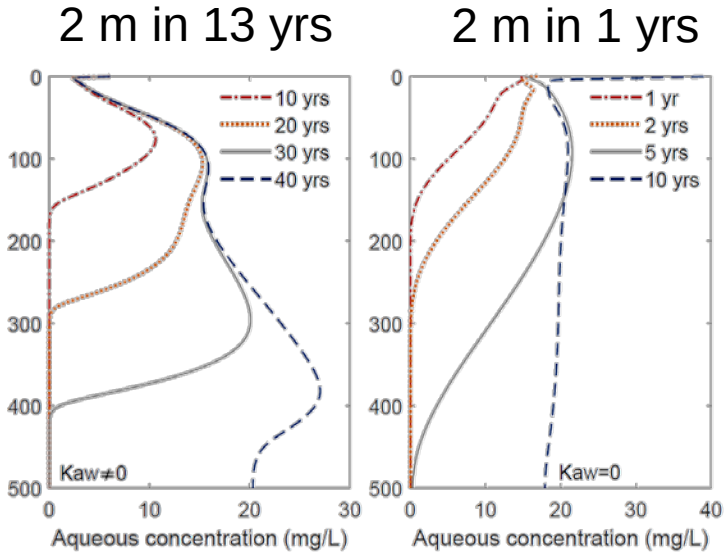
- **Higher recharge rate = shorter transit times**
- **Air-water interfacial adsorption significantly increases retention and decreases migration rate in the vadose zone**

AZ climate



With A-W interfacial adsorption | **No A-W interfacial adsorption**

NJ climate



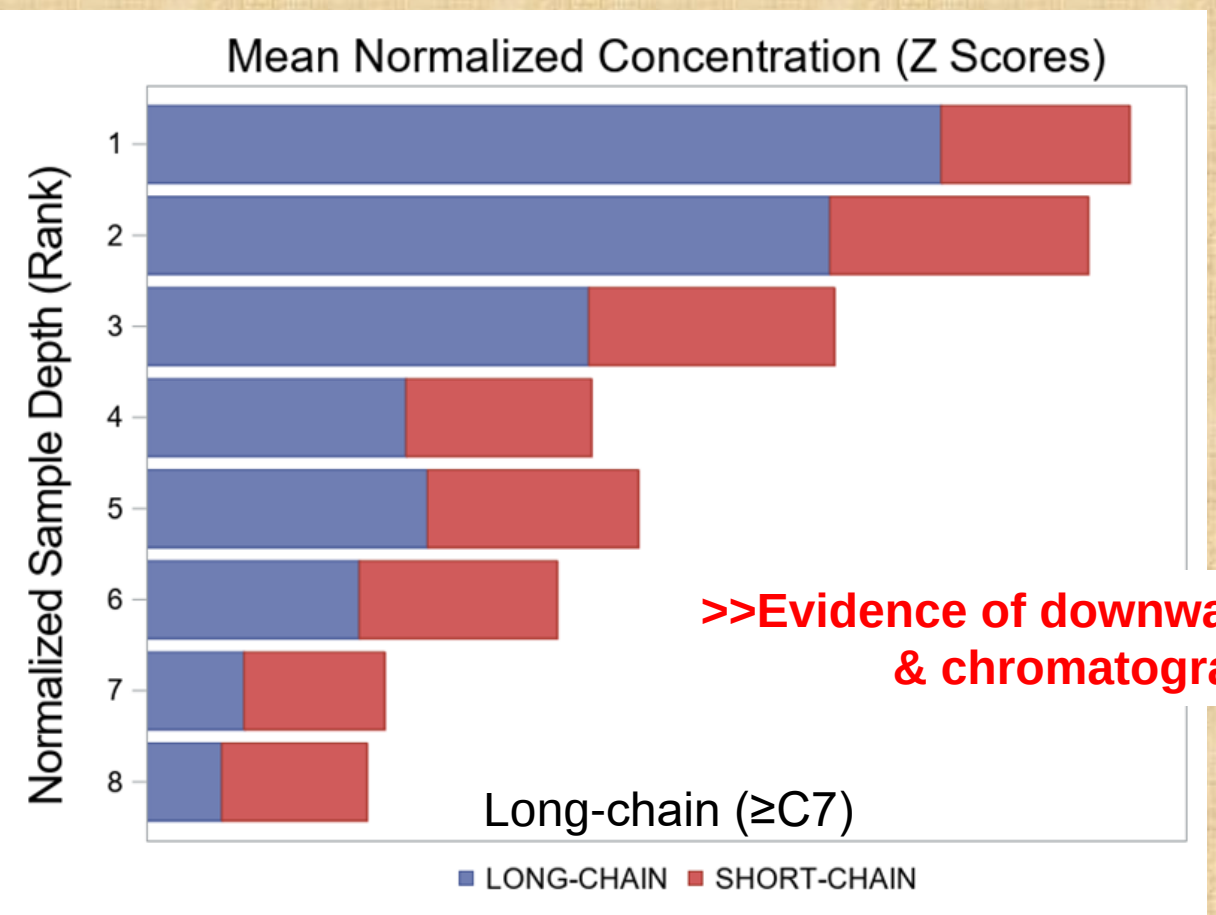
With A-W interfacial adsorption | **No A-W interfacial adsorption**

From: Guo, Zeng, and Brusseau, 2020

Long-term PFAS Distribution in Vadose Zone

Field Study of PFAS Vadose-zone concentrations

Depth distribution of **total PFAS** in soil as a function of chain length



- The data represent 124 boreholes across 30 AFFF sites for which at least 8 depth-discrete samples were collected for each borehole.

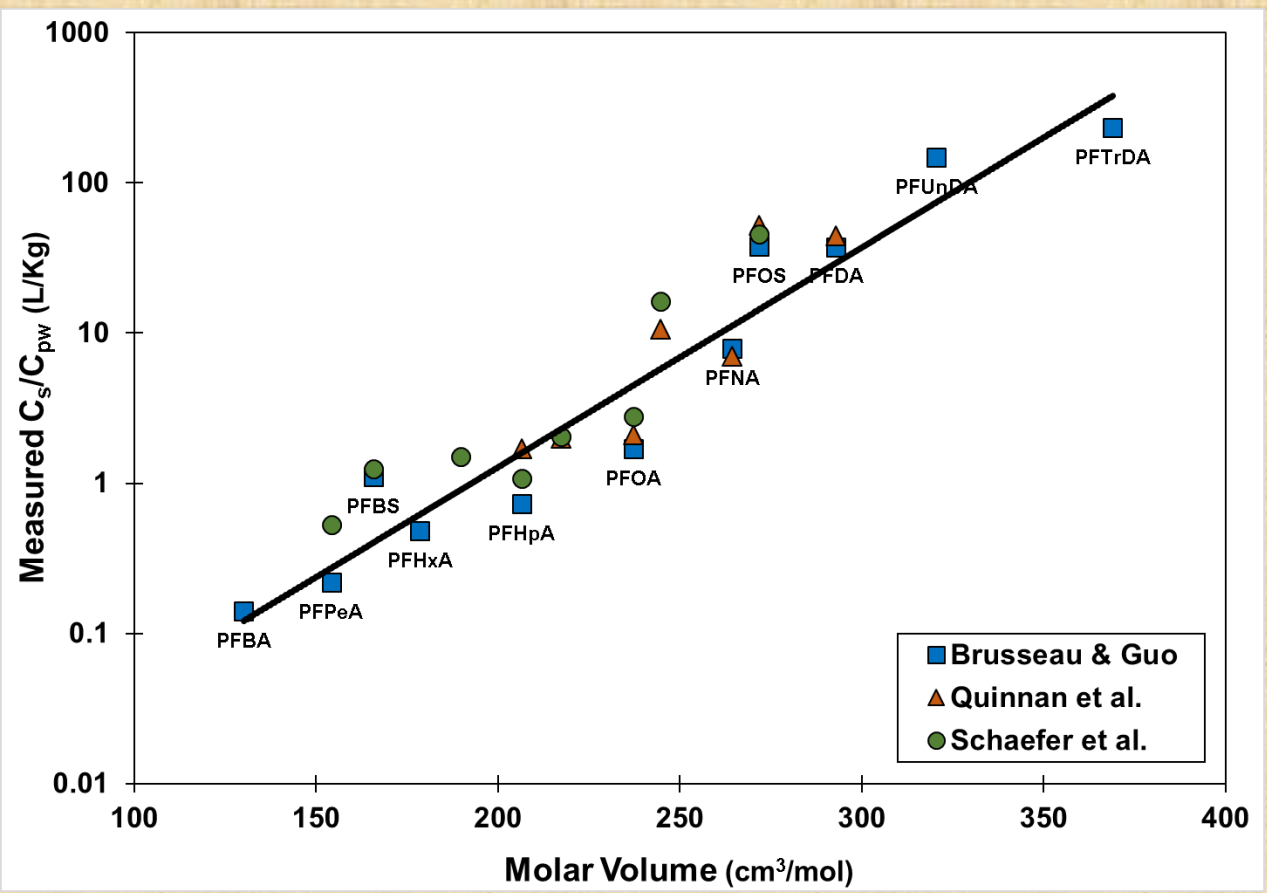
- Depth interval spans from ground surface to top of saturated zone (gw).

>>Evidence of downward migration & chromatographic separation

From: Brusseau, Anderson, & Guo, 2020

Soil vs Porewater Concentration Distributions

Field Studies of PFAS soil vs porewater concentrations



C_s = Soil Concentration

C_{pw} = Porewater Conc

$$\frac{C_s}{C_{pw}} = \frac{\theta_w}{\rho_b} R_d$$

$$R_d = \left(1 + K_d \frac{\rho_b}{\theta_w} + K_{aw} \frac{a_{aw}}{\theta_w}\right)$$

>>Evidence that PFAS distributes between soil and porewater as anticipated for these three systems

From: Brusseau, 2023d

Other Factors

- Physical heterogeneity & preferential flow
  - May reduce retention and led to enhanced transport
- Geochemical heterogeneity
  - Complicate sorption processes
- PFAS mixtures
  - Impact of co-solute interaction on retention
- Co-contaminants
  - Impact on PFAS retention
- Precursors and non-characterized PFAS
  - Potential impacts

Summary

- Retention and leaching in the vadose zone is complex-influenced by multiple processes
- Adsorption at the air-water interface can be significant
  - Determining air-water interfacial areas at the field scale is difficult
- Solid-phase sorption can be complex
  - K_{oc} approach may not be representative
- Models are being developed to simulate PFAS transport
  - Applications for:
    - Quantifying leaching and mass discharge to groundwater
    - Determining soil screening levels
    - Evaluating mitigation and remediation actions

Thank You

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