PFAS in Wastewater Residuals: What we know!

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nebra

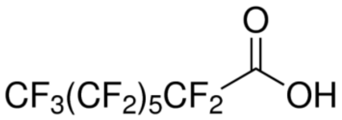
North East Biosolids & Residuals Association

Topics to be Covered

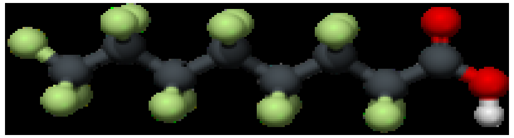
- Background
- PFAS Chemistry
- Risk to Public Health from Land Application
  - Occurrence and concentrations in wastewater and residuals
  - Occurrence and concentrations in the soil resulting from land application
  - Mobility/leaching
- Regulatory Reactions
- Perspectives on PFAS Risk from Wastewater Residuals

Background (general info)

- Per- and polyfluoroalkyl substances (PFAS)



Perfluorooctanoic Acid (PFOA)



PFOA

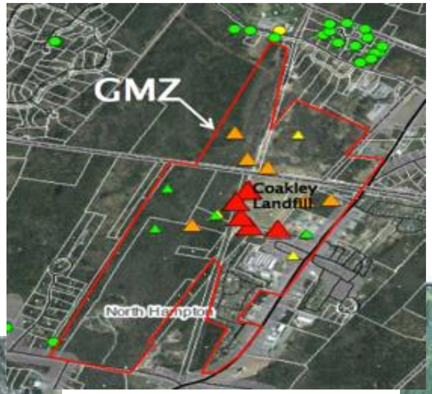
- Large group of chemicals with many subgroups
- Man-made highly fluorinated <u>alkyl</u> (C2-C16) chemicals with unique properties
- Hydrophobic and Lipophobic
- High affinity for proteins
- No natural counterparts

Background (Why we're talking about PFAS)

- Mobile and ubiquitous (arctic, human blood & serum)
- Detected in groundwater in a number of states
- Found in groundwater near land application sites
- Legislatures and state environmental agencies expressing increased concern about PFAS
  - Establishing regulatory limits
  - Attempting to identify sources other than industry (landfills and wastewater residuals)
- PFAS have been found in residuals and land applied soils **<u>not</u>** impacted by industrial sources.

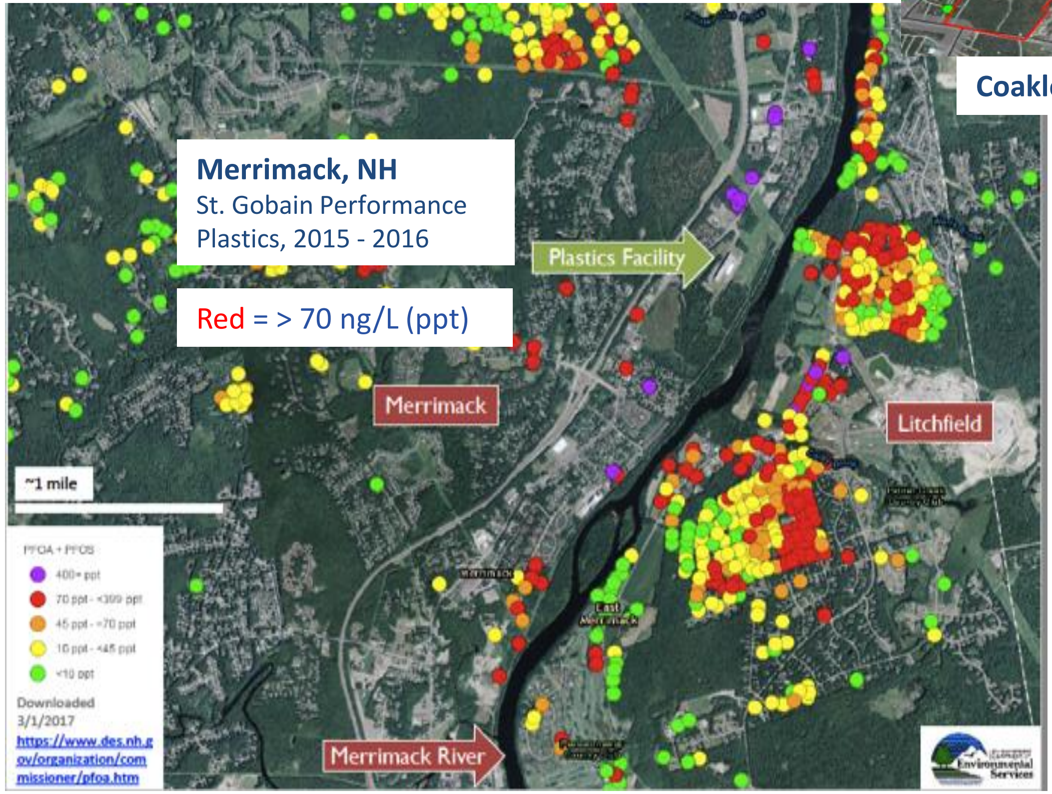
Water well testing around known industrial & landfill sites

Red = elevated
Green / Blue = low



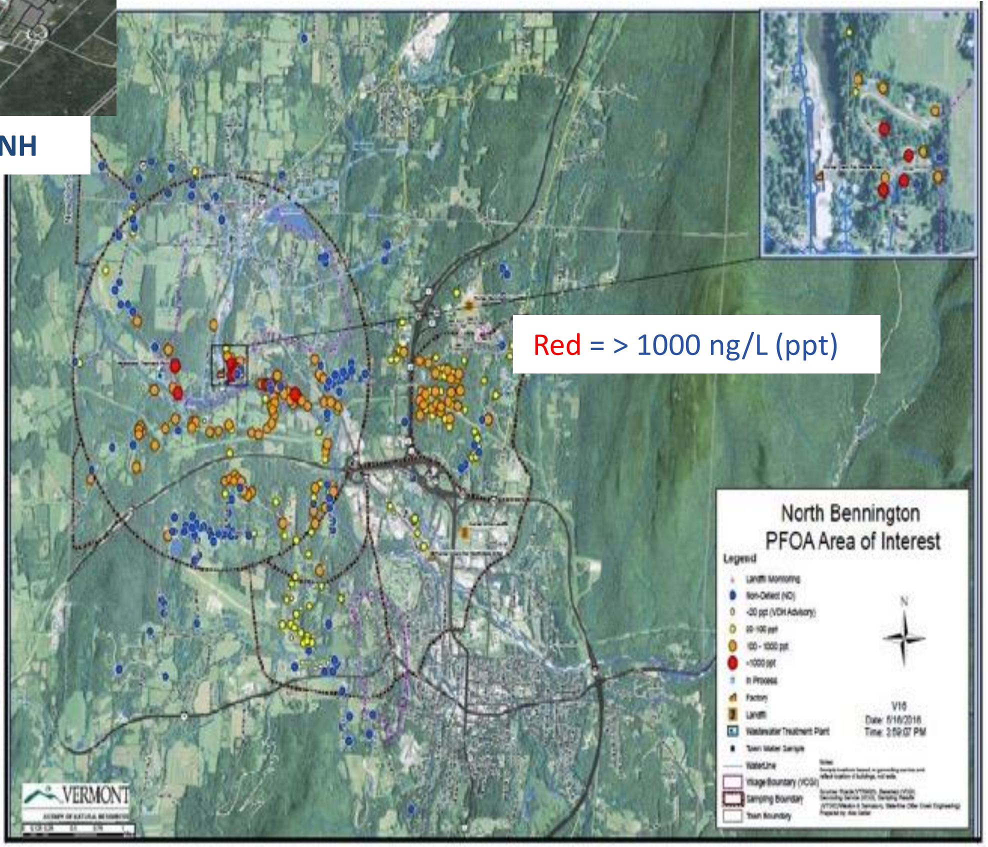
Coakley Landfill, NH

PFAS sources: Leaching from landfills or deposition from fabric coaters emitting PFAS to air → soils → groundwater.



Merrimack, NH
St. Gobain Performance Plastics, 2015 - 2016

Red = > 70 ng/L (ppt)



Red = > 1000 ng/L (ppt)

PFAS Chemistry/Fate

Buck et al. 2011. *Perfluoroalkyl and Polyfluoroalkyl Substances in the Environment: Terminology, Classification, and Origins*.

Large number of chemical groups and individual chemicals (>3000 used on the global market)

- Similar properties valuable in commerce
- Variable behavior in the environment

PFAS products may contain multiple isomers of the intended ingredients, residual intermediary compounds, byproducts, and – after release –degradation products.

Background (general info)

- Lowers surface tension and enhances spreading
- High chemical and thermal stability (C-F bonds)
- Very useful compounds
  - Stain-resistant carpets and fabrics
  - Food cartons, containers, wrappers
  - Surfactants and lubricants
  - Aqueous film-forming foams (AFFFs)
  - Flame retardants

PFAS Chemistry/Fate

- Two production methods that yield different products:
  - Electro-chemical fluorination (ECF)
    - Electrolysis of organic compound in HF
    - Breaking and branching of C-chain
    - ~70% linear/30% branched in PFOA/PFOS synthesis
  - Telomerization
    - Multiple step reaction
    - PFEI – PFAI – FTI – FTOH – variety of PFAS products
    - Linear reactants yield linear alkyl chain products (PFAI)
- Perfluoroalkyl acids (PFAAs) are the metabolites of PFAS precursors

PFAS Chemistry/Fate

- As acids and esters, PFAS compounds susceptible to ionization/dissociation and increased aqueous mobility
- Ionized forms likely to predominate in the environment and biota (including humans)
- Some PFAS compounds may degrade in the environment or biota, but will ultimately transform to very stable and persistent perfluoroalkyl acids (PFAAs)
- The yield rate of PFAAs from biotic and abiotic degradation depends on the precursors and degradation conditions
- Increasing C-chain length reduces leachability and increases bioaccumulation

PFAS Risk and Wastewater Residuals (PFAS in Wastewater)

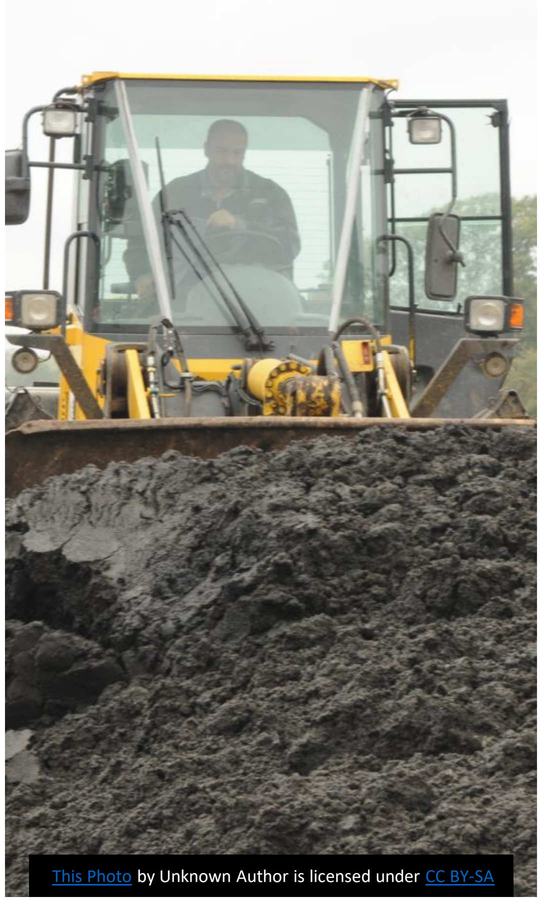
- Pervasiveness and persistence of PFAS in commerce and the environment ensure PFAS loading to WWTFs over the long-term
- Across facilities, influent PFAS loading can be variable both in composition and concentration.
- Historically PFOA and PFOS the most abundant, typically 5-50 ng/L (Margot et al. 2015)
- Total concentrations for common PFAS typically 30-150 ng/L (Margot et al. 2015)
- One study involving an industrial user found influent concentrations of 470 ng/L, 640 ng/L and 61,205 ng/L for PFOS, PFOA, and Perfluorooctanesulfonamide (PFOSA), respectively (Koch, 2015)

PFAS Risk and Wastewater Residuals (PFAS in Wastewater)

- Negligible treatment by conventional wastewater treatment, <5% removal
- Most removal via sorption to wastewater soils
- Organic matter partitioning process (Prevedouros et al. 2006)
- Longer carbon-chain (>6 C) tend to adsorb to solids and are removed in the sludge (Koch, 2015)
- Effluent PFAS concentrations slightly lower than influent

Study	PFOA (ng/L)	PFOS (ng/L)
Margot et al. 2015	13	12
Prevedouros et al. (2006)	24	13

PFAS Risk and Wastewater Residuals (PFAS in Residuals)



- PFAS is present in residuals
  - Variable compounds (results for 19 tabulated)
  - Variable concentrations
- Highest concentrations are found in residuals with direct industrial input (Lindstrom et al., 2011):

	4 WWTF	Decatur, AL
PFOA (ng/g):	<17	244
PFOS (ng/g):	58-159	3000
PFOSA (ng/g):	<44	244

- PFAS are also found in residuals without industrial input, but at lower concentrations.

PFAS Risk and Wastewater Residuals (PFAS in Residuals)

- In the 2000s, PFAS were found in typical biosolids in concentrations of tens of parts per billion (ppb), with a U. S. average of 34 ppb for PFOA and 403 ppb for PFOS (Venkatesan and Halden, 2013). Recent tests of land applied New England biosolids and residuals found average concentrations of 2.3 and 5.3.
- Recent studies including wastewater sludge

Study	PFOA (ug/Kg)	PFOS (ug/Kg)
Zareitalabad et al., 2013 (median)	37	69
Sepulvado et al., 2011 (range)	8 – 68	80 – 219

PFAS Risk and Wastewater Residuals (PFAS in Residuals)

2017 PFAS data compiled by NHDES and NEBRA,

22 facilities from NH and Northeast, 27 data points

Chemical	% detection	Conc. Range (ug/Kg)	Ave. Conc. (ug/Kg)
PFBA	20	0.54 – 140	34.6
PFPeA	8	18 – 27	22.5
PFHxA	84	0.21 – 75	11.0
PFHpA	26	0.077 – 2.8	1.1
PFOA	32	1.1 – 15	6.7
PFNA	30	1 – 3.6	2.6
PFBS	7	5.2 – 6.2	5.7
PFHxS	22	0.24 – 73	13.3
PFOS	62	0.59 - 390	34

PFOA/PFOS in biosolids/ residuals

vs.

PFOA/PFOS in other media

Biosolids & Residuals	**PFOA (ppb)**	**PFOS (ppb)**	
Regulatory standards	**none**	**none**	
Sampling of U. S. biosolids, 2001 (Venkatasen and Halden, 2013)	**34**	**403**	
A northern New England biosolids compost, 2017	**8.3**		
NH land applied solids, 2017, n=20, non-detects included at detection limit	**2.3**	**5.3**	Mean (includes 17 wastewater biosolids, 2 paper mill residuals, & 1 water treatment residual)
Northeast paper mill residuals	**1.6**	**25**	
Other media			
Household organic waste compost	**6** (median) **3.4 – 35** (range)		all PFAS combined
Dust in U.S. daycare centers, median values (Strynar and Lindstrom, 2008)	**142**	**201**	
Human blood, U. S. population 1999 average (CDC NHANES)	**5**	**30**	
Human blood, U. S. population 2012 average (CDC NHANES)	**2**	**6**	

PFAS Risk and Wastewater Residuals (PFAS in Soil)

- Land application of PFAS contaminated residuals results in detectable PFAS concentrations in the soil.
- Soil concentrations following land application reported in the literature:

Source	Type of loading	PFOS (ug/Kg)	PFOA (ug/Kg)
Washington et al., 2009	High PFAS	30 – 410	50 – 320
Sepulvado et al., 2011	Short-term Long-term	2 – 11 5.5 – 483	No data
Gottschall et al., 2017	One-time	0.2 – 0.4	0.1 – 0.8

PFAS in Soil – Land Application & Other Sites

Source	Type of loading	PFOA (ug/kg)	PFOS (ug/kg)
Washington et al., 2009, Decatur, AL biosolids	High PFAS in biosolids	50 – 320	30 – 410
Sepulvado et al., 2011 Chicago, IL biosolids	Short-term Long-term Control plots (cross contaminated?)	no data	2 – 11 5.5 – 483 22, 96
Gottschall et al., 2017, Ottawa, ON biosolids	One-time	0.1 – 0.8	0.2 – 0.4
Garden control soils, MN (=6) VT Dept. Health testing (n=100), for comparison NH DES soil testing 2016 (n=160)	No significant PFAS source Aerial deposition Aerial deposition	0.29 – 0.54 ND – 45 most < 10 ND – 33	0.93 – 2.1

For Comparison: PFOA/PFOS Soil Screening Levels

No significant risk likely from dermal, ingestion, etc. direct exposure from biosolids & soils.

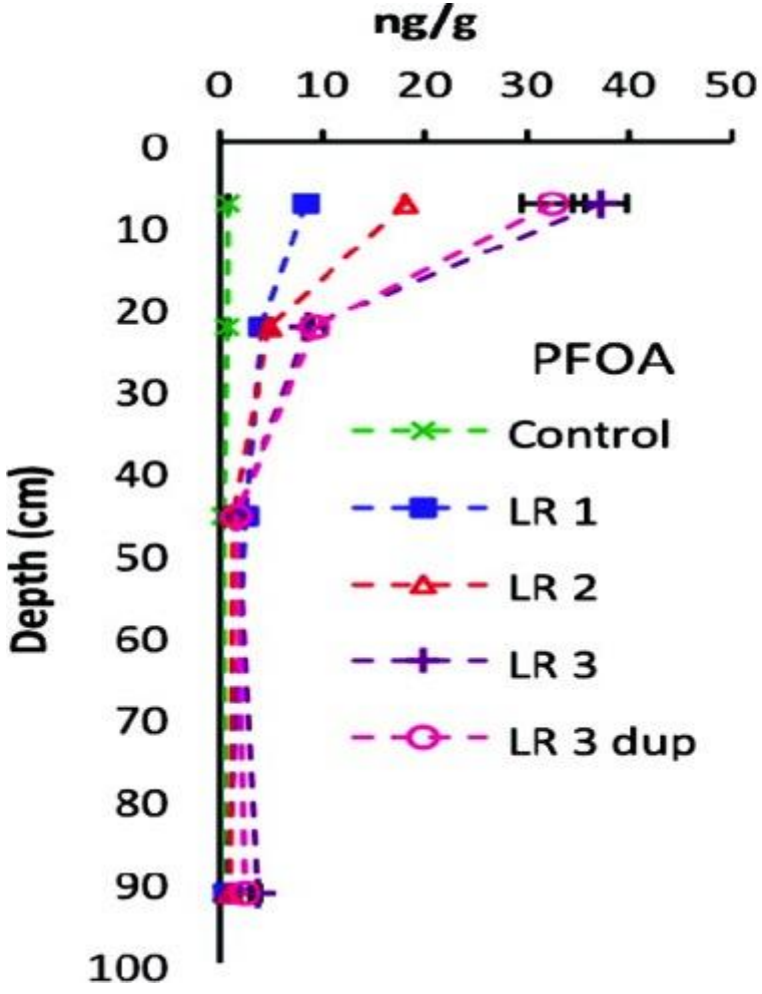
	PFOA (ppb)	**PFOS** (ppb)	**Notes**
Soil			
Australia, 2017 – soil screening level for 99% species protection	**650**	**6,600**	
Minnesota soil reference value, in effect in 2012	**2100**	**2100**	
NH DES Soil Screening Level	**500**	**none**	Based on risk from dermal exposure and ingestion
VT DEC Soil Screening Level	**300**		Based on risk from dermal exposure and ingestion

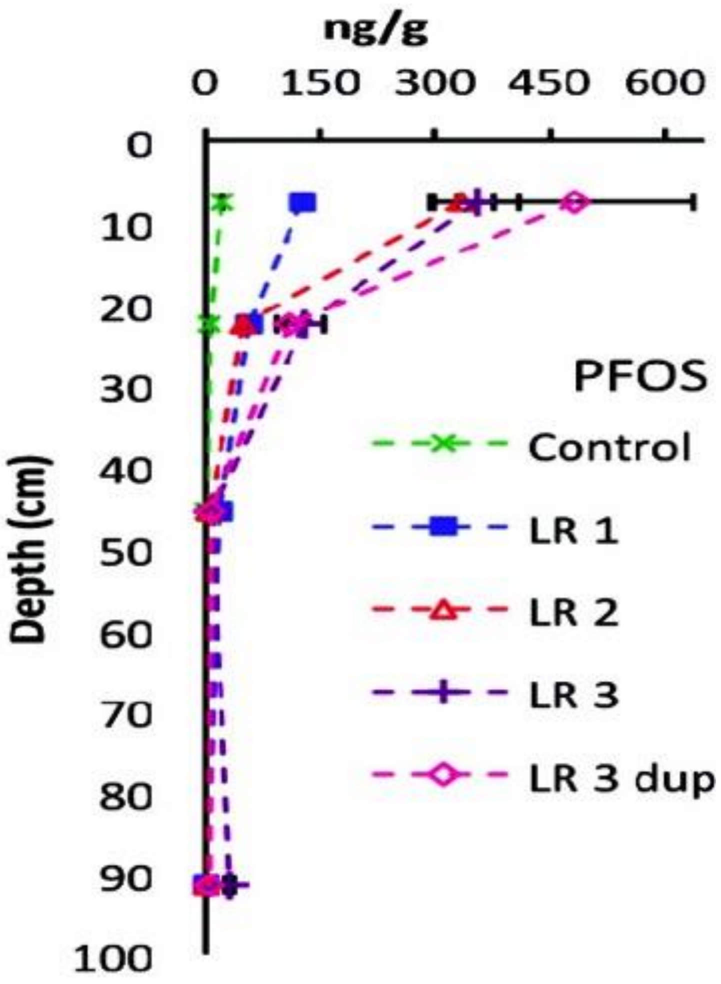
PFAS Risk and Wastewater Residuals (PFAS in Soil)

- PFAS soil concentrations can be correlated to residuals loading rate
- Correlation is especially strong for longer chain (>C8) PFCA.
- For short chain PFCA, soil concentration may correlate better with time from last application.
- PFAS concentrations in well water and surface water can be correlated to loading rate of short chain PFAS.
- Soil PFAS concentrations at depth may increase over time.
- Soil PFAS concentration can change as a result of precursor degradation.

PFCs move through soil

Sepulvado et al; *Environ. Sci. Technol.* 2011, 45, 8106-8112



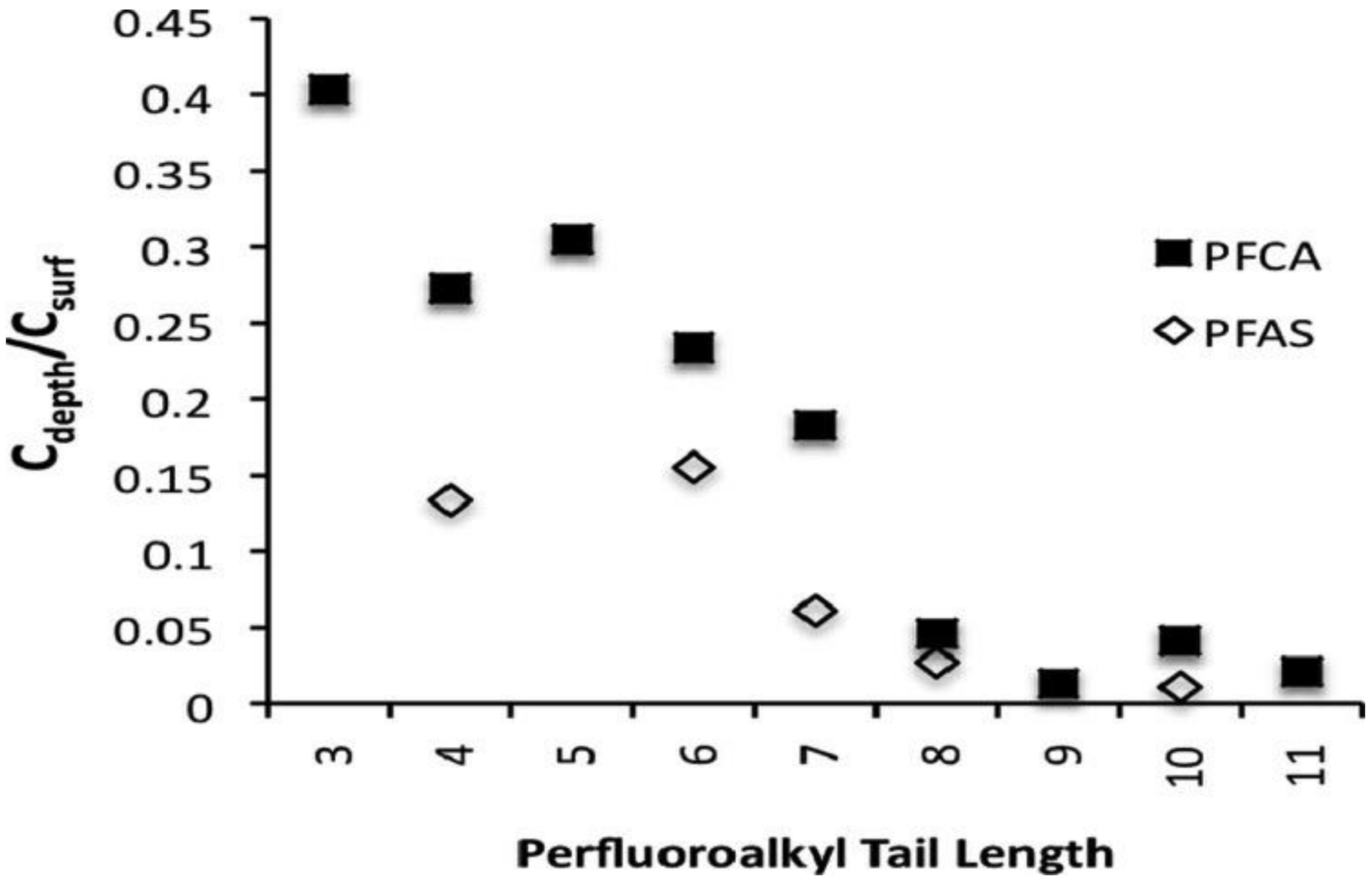


Concentrations of PFOA and PFOS with depth in the long-term plots at various loading rates.

Control = 0 Mg/ha
LR1 = 553 Mg/ha
LR 2 = 1109 Mg/ha
LR 3 and LR 3 dup = 2218 Mg/ha (dry weight basis).

Mobility varies with chemical structure

Sepulvado et al; *Environ. Sci. Technol.* 2011, 45, 8106-8112



Ratios of surface concentration (*C*surf) to concentration in the bottom soil core depth interval (60−120 cm, *C*depth).

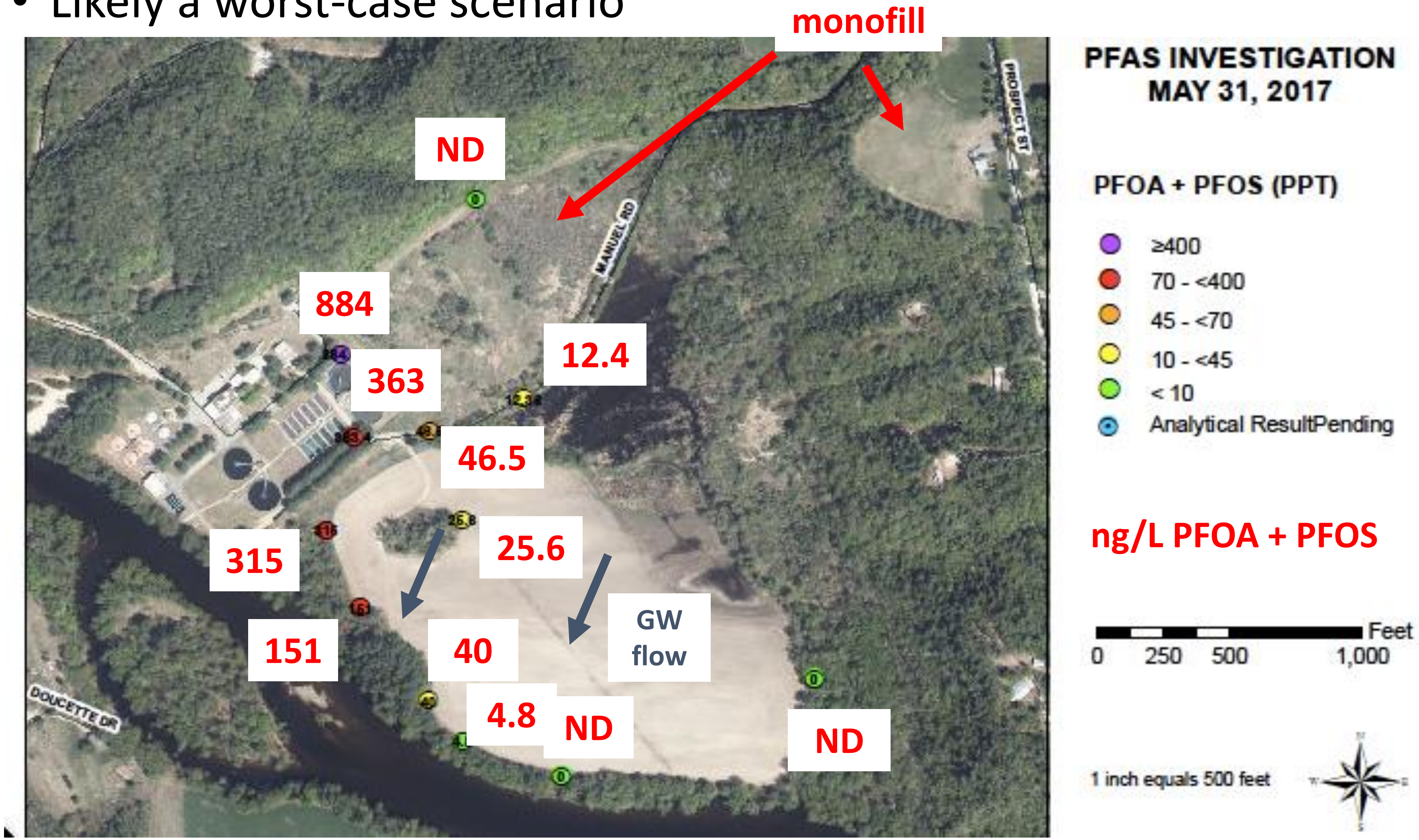
Ratios represent an average of the ratios calculated for the long-term plots for each biosolids loading rate.

PFAS Risk and Wastewater Residuals (Mobility/Leaching)

- Little direct evidence that residuals without obvious industrial PFAS contributions are a risk to public health via groundwater contamination following land application
- A determination of public health risk is influenced by several factors:
  - Type and quality of wastewater residuals,
  - PFAS compounds to be considered,
  - Field conditions (OM content, climate, soil type, depth to groundwater, etc.), and
  - Regulatory requirements (loading limits, land application restriction, drinking water standards, required setback, application rates).
- Differences in these factors from state to state can lead to different conclusions regarding public health risk

Monitoring well testing at biosolids monofill

- Monofill used in 1980s. Since ~1996, all biosolids from WWTP (11.5 MGD) have been land applied, some on farm field shown.
- Likely a worst-case scenario



Residuals management is being negatively impacted right now.

Regulatory response in March 2017 drives recycle paper mill residuals to landfill and composting business to laying off workers.

Movement of PFAS to tile drains & shallow groundwater

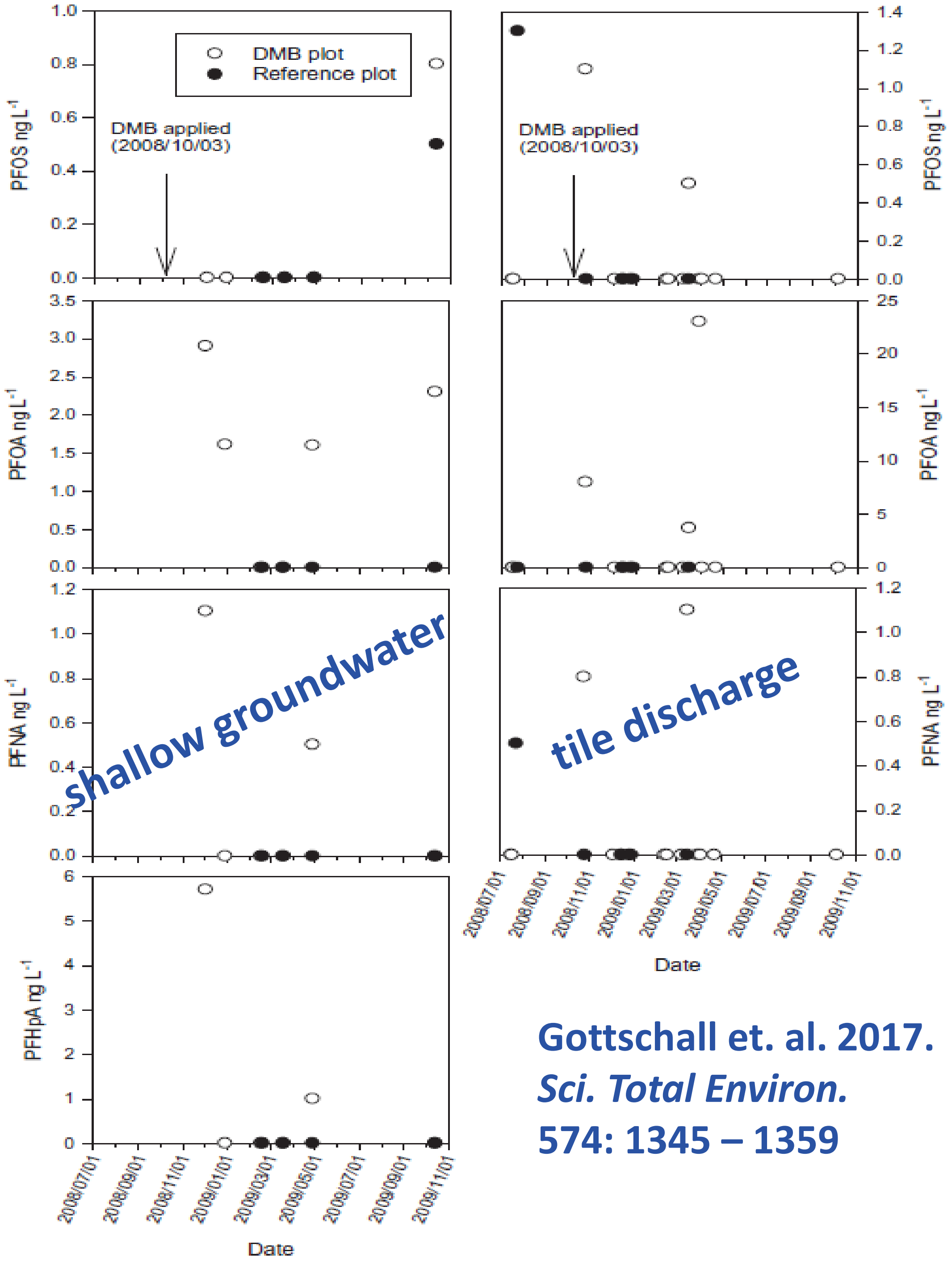
Study site in Ontario:

- Humid continental climate
- Corn, wheat, soy rotations
- Very light tillage
- Systematic tiling, 15m spacing, about 1m depth
- Ottawa biosolids (mixed residential, industrial, commercial):
- 1.6 *ug*/kg PFOA, 7.2 *ug*/kg PFOS
- Treated by AD, centrifugation
- 22 Mg dw/ha (9.8 tons dw/ac)
- Moldboard plow to ~ 20cm
- Planted to winter wheat

Gottschall et. al. 2017. *Sci. Total Environ.* 574: 1345 – 1359

Conclusions (Gottschall et al. 2017)

- Perfluorinated chemicals detected in both groundwater and tile discharge after a single large biosolids application.
- Chemicals detected for months after the application.
- Relative contributions of leaching through soil matrix, and preferential flow through macropores are unknown.
- No groundwater standards or guidelines exceeded



Gottschall et. al. 2017. *Sci. Total Environ.* 574: 1345 – 1359

PFAS Risk and Wastewater Residuals (Mobility/Leaching)

What does the scientific literature tell us about leachability of PFAS:

- PFAS can and does move through the vadose zone to groundwater
- Correlations between biosolids/PFAS loading and observed groundwater and surface water concentrations have been observed
- One potential set of conservative soil screening levels for protection of groundwater were calculated for PFOS (3 ug/kg) and PFOA (3 ug/kg) (Xiao et al. 2015)
- Observation in groundwater can follow release to surface soils by years if not decades, especially for longer chain PFAS (C8 and higher)

PFAS Risk and Wastewater Residuals (Mobility/Leaching)

- Sorption in the soil does occur and is best described as a sorption equilibrium reaction
- PFAS sorption equilibria (log K_d)are influenced by:
  - **PFAS carbon chain length**
  - **Organic carbon content**
  - pH
  - Cation concentrations
  - Specific surface area/clay content
  - Types of soil minerals

PFAS Risk and Wastewater Residuals (Mobility/Leaching)

Conclusions on PFAS risk:

- The ubiquitous presence of PFAS in plant, animal, and human tissue as well as air, soil, and water resources confirms the obvious mobility of these chemicals
- However, there is little information to answer our original question
- A little perspective on PFAS risk from wastewater residuals:
    - PFAS are in residuals because they have been widely used for decades and persistent in the environment
    - Presence in residuals is not evidence of risk or even significant exposure in excess of current everyday exposure
    - Uncertainty on extent of public health risk

Reactions of State Regulatory Agencies to PFAS Contamination of Groundwater

- State environmental agencies are under public and legislative pressure to adopt regulations to protect groundwater
- Adopting widely variable groundwater/drinking water PFAS standards based on still evolving understanding of PFAS toxicology.
  - Confusion over precursors and degradation pathways
  - Some are summing PFAS – may not be appropriate
- Proposals to adopt stringent PFAS regulations on other media
  - Hazardous waste determinations
  - Soil screening standards to protection groundwater
  - Screening standards in biosolids/residual to protect groundwater
- Proposed standards based on questionable modeling
  - Overly conservative assumptions (loading rates, aquifer size, dilution/attenuation, etc.)
  - Modeling based on lab studies/testing, limited field verification
  - Poor understanding of PFAS-soil equilibria and soil organic matter partitioning

PFOA/S drinking water standards / screening levels: diverse values

Jurisdiction		PFOA (ppt)	PFOS (ppt)	Notes
Advisory or Regulatory Standard				
U. S. EPA, 2016	**Advisory**	**70**		for combined
New Hampshire, 2016, AGWQ	**Standard**	**70**		for combined
Vermont, 2016	**Standard**	**20**	**20**	
Australia, January 2017 interim drinking water guidance	**Advisory**	**5,000**	**500** (includng PFHxS)	
Australia, April 2017 final drinking water guidance	**Advisory**	**70**	**560** (including PFHxS)	
Canada, proposed June 2016; screening values November 2017	**Advisory**	**200**	**600**	
Michigan, non-cancer values, 2014		**420**	**11**	
Minnesota drinking water (as of 2016) (as of 2017)	**Standard** **Advisory**	**300** **35**	**300** **27**	PFBA & PFBS = 7000 Adopted 5/2017
New Jersey health-based guidance Proposed	**Advisory**	**40** **14**		
West Virginia (as of 2016)	**Standard**	**400** or **500**		
Maine CDC, 2014, health-based MEG		**100**		
Maine residential groundwater RAG	**Advisory**	**560**	**130**	
California – Office of Environmental Health Hazard Assessment, Nov. 2017	**Prop. 65 Listing**			Listed because of reproductive toxicity concern

VT Adds PFAS to Hazardous Waste Regulations

VT 21 & 22: Liquid wastes containing PFOA and/or PFOS > 20 parts per trillion (ppt) = hazardous waste

(e.g. if the PFOA concentration is 15 ppt and the PFOS concentration is 6 ppt then there is an exceedance of the standard)

Some exemptions for wastewater residuals and sewage going to WRRF, but is septage exempted, or is that a hazardous waste now?

Attempts to Define Safe PFAS Levels in Soils/Residual for Protection of GW

- **Alaska, 2018**
  - Proposed migration-to-groundwater soil cleanup level: PFOA: 0.29 *u*g/kg (ppb)
    PFOS: 0.53 *u*g/kg
- **New York, 2017**
  - NYDEC PFOA + PFOS: 72 *u*g/kg
- **Maine, 2017-18**
  - Initial (rote modeling - SEVIEW (SESOIL & AT123D)): PFOA: 0.438 *u*g/kg
    PFOS: 0.908 *u*g/kg
  - Current provisional (adapted from EPA RSLs 2017): PFOA: 2.5 *u*g/kg
    PFOS: 5.2 *u*g/kg
  - NEBRA encouraging ME DEP to remove any residuals screening levels at this time and wait for the science to catch up.
- **U. S. EPA, 2017 RSLs** – anticipated? temporary? PFOA: 0.00017 *u*g/kg (!)
  Someone reported this at a recent conference. Real? PFOS: 0.00038 *u*g/kg (!)

Perspective: Wastewater & biosolids mirror modern life.

- Wastewater solids management is not optional.
- Wastewater solids can be landfilled; incinerated; or treated, tested, & applied to soil as biosolids. The latter usually is best environmentally, overall.

Perspective

- PFAS are clearly mobile – found throughout the world
- PFAS are in wastewater & residuals because they have been widely used for decades and are persistent in the environment
- Presence in wastewater & residuals is not evidence of risk or even significant exposure in excess of current everyday exposure
- Uncertainty on extent of public health risk; health studies vary.
- PFOA & PFOS are phased out in No. America. Human blood serum levels down 50%+ over ~15 years.
- Is this is a legacy issue, at least for PFOA & PFOS? Modern biosolids/residuals are less of a concern than historic ones.

Perspective

- There remain scientific & regulatory uncertainty & debate over appropriate limits in drinking water.
- The core concern for biosolids & residuals management is potential leaching of PFAS impacting drinking water.
- Initial leaching modeling has instigated concern, but most of that modeling includes:
  - worst-case-scenario assumptions
  - output of concentrations in soil pore water/top of groundwater table
  - no dispersion or dilution factors
- Regulatory agencies that adopt low (<70 ppt) PFAS standards for drinking water or groundwater are finding it hard to enforce and mitigate all locations, because there are many.
  - EPA stresses that the 70 ppt is a public health advisory level for *lifetime drinking* water; some call it overly conservative; some call for a drinking water level as low as 1 ppt (impractical).
  - With PFOA & PFOS levels already declining dramatically in humans, states need to assess what public health benefit is gained for considerable cost in chasing groundwater protection at lower levels.
- Biosolids managers should apply the same best management practices as for all biosolids and their CECs/ microconstituents, especially source control (e.g. landfill leachate).

Perspective: Bioassays get at whole system impacts.

- 1980s & ‘90s: Sopper (Penn State Univ.): testing of plant and rabbit health on sites reclaimed with biosolids (with focus on heavy metals)
- 2000s: Brown (Univ. of WA), USDA, and others: testing of plant and rabbit health on sites reclaimed with biosolids
- 2010: University of Guelph – fate of endocrine disruption during biosolids treatment processes
- 2010: College of William and Mary: bioavailability of PDBEs using earthworms and crickets in laboratory
- 2013: Park, et al. (Tom Young team, UC Davis): Triclosan has “little relative impact on overall community composition…” and “TCS slightly increased biomarkers of microbial stress, but stress biomarkers were lower in all biosolid treated soils, presumably due to increased availability of nutrients mitigating potential TCS toxicity.”
- 2013: Puddephat thesis (Lynda McCarthy team, Ryerson Univ.): lab bioassays in Ontario using earthworms, springtails, *brassica rapa*, beans, corn, & aquatic organisms

Puddephat / McCarthy research (Puddephat, 2013)

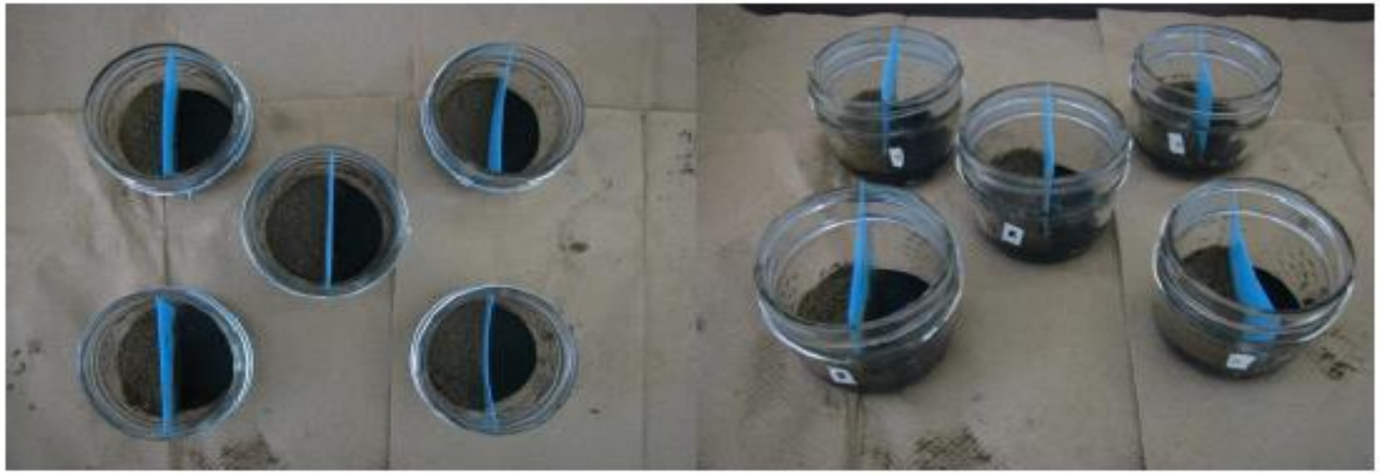


Figure 17: Avoidance chamber setup for *Folsomia candida*

Brassica rapa

Zea mays

Figure 30: Feeding of Earthworms in Ryerson Long-Term Bioassay Chambers. Image shows the mating chambers atop the Evan's Boxes

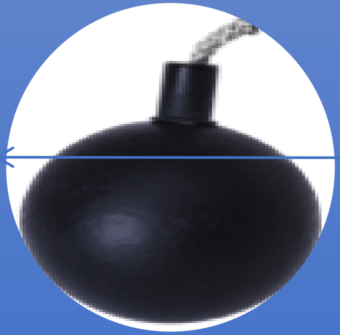
Conclusions of Puddephat / McCarthy:

Puddephat, 2013:

"…biosolids had little negative impact on the terrestrial biota examined and as a general rule, there was no impact observed. Where effects were observed, the majority of instances were positive. In the few instances where there was negative impact observed, for example in the initial growth stages of the plant bioassays, with further development of the organism, there was no longer a significant difference between the reference and treatment plants."

And PFAS were most likely in those biosolids at levels higher than today's biosolids.

Q: Where do normal, modern biosolids applications lie on the continuum of PFAS impacts to groundwater?

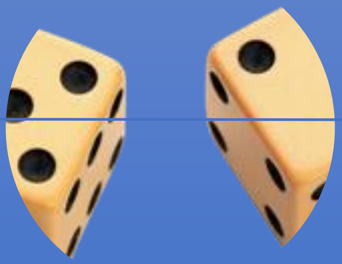


Historic residuals impacted by PFAS manufacturer

(e.g. 3M, Decatur, AL; NE farm with high PFOS likely from 1980s papermill residuals use)

Historic / modern residuals heavily applied repeatedly

(Sepulvado et al. 2011



Modern residuals applied semi-annually with setbacks, etc.

EQ biosolids used for several years - home settings

(e.g. 3 sites in NH)

Higher concern

Where drinking water & ground water standards are set will determine our level of concern.

Minimal to no concern

Reactions can impact residuals/biosolids recycling

It seems premature…

- …to set lower drinking water numbers (MCLs, etc.); EPA PHA is being applied and provides high level of protection.
- …to set soil or wastewater or residuals concentration screening or enforcement levels. The science is not there yet.

Meanwhile…

NH Legislation – a dozen bills in 2017 & 2018

Pushing lower drinking, groundwater, and surface water standards

NJ proposed: 14 ppt for PFOA in drinking water

PA proposed: 6 ppt for PFOA in drinking water

NEBRA Response to PFAS Concerns

- NEBRA pursuing answers via facilitation of relevant research and guidance:
  - PFAS Advisory Group
  - Fact Sheets & Perspective
  - PFAS & Residuals Sampling & Analysis Guidance
  - Literature Review
  - PFAS Research with UNH & NH DES
  - Webinars on PFAS issues
- Working with state agencies and legislatures to deal with PFAS risk in a measured and thoughtful manner (need to avoid regulatory over-reaction)
- Some resources (e.g. recording of analysis webinar) are here: https://www.nebiosolids.org/nebra-publications
- More resources for members & upon request.

Core research question:

"Does land application of wastewater residuals (paper mill solids, municipal biosolids, etc.) at fertilizer rates with current common regulatory requirements and proper industrial source controls represent a risk to public health from PFAS contamination of groundwater via leaching and/or surface water via runoff?"

Research Needs

- **Field research** – Evaluate extent of issue re biosolids/residuals with thoughtful, planned testing of current & historic biosolids use sites, including groundwater, surface water, soils at various depths, plant tissues, & considering other potential sources of contamination, age of biosolids used, number of applications, etc.
- **Field research looking forward** - Leaching column studies and full-scale field work at actual land application sites with no legacy biosolids or other PFAS concerns.
- **Basic data on key PFAS parameters**: determine appropriate data inputs to models (e.g. Koc).
- **What about other PFAS besides PFOA & PFOS?** – much data still to be developed.
- **Modeling**: Adapt models for PFAS and *field verify* them to provide screening and guidance.
- **Analysis** – approved methods needed
  - EPA Method 537 Rev.1.1 – the only current validated method, just for drinking water. Modified methods are being applied variably, and data are suspect
  - Methods for solids and waters other than drinking water may be approved under Solid Waste program this year. Years still before they are approved under Clean Water Act.

<u>**Ultimate goal of states**</u>: What is an acceptable concentration of PFAS in biosolids/residuals that is protective of groundwater when biosolids/residuals land applied at fertilizer rates on an annual basis?

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QUESTIONS?

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