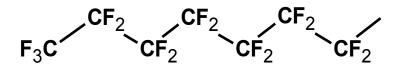


NEBRA Literature Review: Sources of PFAS in Agricultural **Production** v.1.0

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I. Introduction

Over the last several years, significant concern about potential public health impacts resulting from exposure to poly- and perfluoroalkyl substances (PFAS) has arisen within the general public and among state regulatory agencies. These concerns have generally followed discovery of PFAS contamination of groundwater and/or drinking water associated with industrial and fire-fighting uses or production of PFAS. While investigating other potential sources of PFAS emissions to the environment beyond industry, state environmental agencies have tended to focus on waste management sites and facilities such as landfills, wastewater lagoons, and residuals (biosolids, septage, and short paper fiber (SPF)) land application sites. Concurrent to these investigations, states, particularly in the Northeast, have proceeded to adopt very stringent PFAS groundwater and drinking water standards. Initially, these regulations focused on perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS) emulating the federal health advisory set by the U.S. Environmental Protection Agency (USEPA) in May 2016 (70 ng/L or ppt for the sum of PFOA and PFOS). More recently, states have expanded their lists of regulated PFAS and further tightened the standards for PFAS in groundwater and drinking water (e.g. MA has adopted a 20 ng/L limit for the sum of 6 PFAS in drinking water and groundwater). Detection of PFAS in groundwater associated with residuals management sites has prompted some state agencies in the Northeast to impose PFAS testing requirements and use restrictions on residuals and to imply that additional restrictions may be necessary and forthcoming. Aside from known industrial and fire-fighting point-sources, environmental agencies seem to implicate residuals land application as the only significant source of PFAS contamination to the process of agricultural production. This paper will attempt to answer the following question through a review of the relevant literature:

Are there historical and current sources of PFAS contamination in agricultural production that represent a significant potential risk to public health and the environment when assessed according to environmental standards currently being developed and adopted in the Northeast?

II. Current Environmental Standards and Risk Thresholds

To answer the above question, it is necessary to identify the health-based standards being applied to environmental media and agricultural products. State environmental agencies, particularly in the Northeast, have been under pressure from the public and sometimes their state legislatures to adopt stringent regulations for PFAS in groundwater and drinking water. In addition, these state agencies have begun to adopt and impose standards for other environmental media. As more toxicological research is performed, PFAS risk-based standards and guidelines for food and ecological risk are being developed. These standards for various media are being rapidly adopted and even updated to more stringent standards.

In February of 2019, the USEPA published *EPA's Per- and Polyfluoroalkyl Substances (PFAS) Action Plan.* This document outlines USEPA's plan for managing risk associated with PFAS (USEPA 2019). The document describes numerous proposed projects, both short-term and long-term. Included among these activities are developing additional toxicological data pertaining to PFAS, guidance on site cleanup, ecological risk standards, and drinking water standards (USEPA 2019). The adoption of stringent federal standards in any of these areas may result in greater consistency in regulatory standards between states but will likely have a significant influence on the answer to the central question of this paper. Table 1 is an overview of some of the current environmental standards and guidelines employed by various states. Included in the table are PFAS thresholds for drinking water, groundwater, surface water, soil, and recycled residuals/wastes. Comparable standards imposed by other countries are included as a reference.

Jurisdiction	Standard ¹	PFOA (ppt)	PFOS	5 PFAS ² (ppt)	Notes	
	Stanuaru	(ppt)	(ppt)	(ppt)		
Drinking Water Limits U.S. EPA (2016) Advisory		7	70		ppt (ng/L) is customary measure for PFAS in water Public Health Advisory (PHA) – combined total concentration	
U.S. CDC – ATSDR (Oct.	Advisory	/	0			
2018)	Advisory	78/21	52/14		For Adult / For Child. Also PFHxS: 517/140, PFNA: 78/21	
CA Prop 65 Listing (2017)						
CA OEHHA notification					Reproductive toxicity concern; requires labeling	
levels	Regulatory	Detection	Detection		Adopted August 2019; level at which public water supply must notify local government.	
CA OEHHA response	Developing	5.1	6.5		Planned for October 2019: response level at which drinking water source is taken offline	
levels	Developing	10	40		https://oehha.ca.gov/water/notification-levels-chemicals-drinking-water	
MA – DEP (2020)	Guidance			20	For the sum of 6 PFAS, the 5 plus PFDA	
MN – Health Dept. (2019)	Regulatory	35	15		Health Risk Levels (HRLs). PFHxS: 47 ppt	
NH – DES (2020)	Regulatory	12	15		PFNA: 11 ppt. PFHxS: 18 ppt. All are also groundwater standards.	
NY – Health Dep. (2020)	Regulatory	10	10			
Australia Health (2017)		560	70			
					e ratios of the measured levels to the limits for PFOA + PFOS shall not	
Canada Health (Dec.			600		e.g. 400 ppt is combined limit. Canada also set 20 ppt limit on PFNA &	
2018)	Regulatory	200	600		ppt for other PFAS. BC PFOS limit = 300 ppt.	
European Union (2018)	Developing	100	100	500 (see note)	Proposed advisory; sum of all PFAS limit: 500 ppt	
United Kingdom (2009)	Guidance	300	300	(000.000)	Admin. Level 1 (lowest drinking water screening values)	
Surface Water Limits						
				1	Applied to evaluation of wastewater effluent discharges. 11 ppt if	
MI (2015)	Regulatory	420	11 or 32		drinking water source	
Groundwater Limits		•	•			
U.S. EPA	Draft interim	70 (40 for	each alone)		Proposed interim groundwater screening values	
					PFBS = 40,000 ppt. These must be met for ending work on site	
					cleanups. If more than 1	
U.S. Dept. of Defense					kind of PFAS is present, limits are 40 ppt each PFOA & PFOS, 40 ppb	
(DoD)	Guidance	400	400		PFBS.	
MA – DEP	Regulatory			20	For the sum of 6 PFAS, the 5 plus PFDA	
MI – DEQ (now EGLE)	Regulatory	7	0		For groundwater used for drinking water	
NH – DES (2020)	Regulatory	12	15		PFNA: 11 ppt. PFHxS: 18 ppt. All are also drinking water standards.	
Most other states		No standards		-		
Soil & Materials		PFOA (pph)	PFOS (ppb)		ppb (ug/Kg) is customary units for PFAS in soils, sludges, biosolids, etc.	
Screening	Guidance	(ppb) 0.172	(ppb) 0.378			
U. S. EPA (2018) AK – DEC (2018)	Proposed/on hold	0.172	0.378		Regional Screening Levels (RSLs) modeled to protect groundwater	
AK - DEC (2018)	Proposed/offitiold	0.29	0.55		Proposed – but on hold - Soil Cleanup, groundwater risk Remedial Action Guidelines (RAGs) for soil cleanup based on	
ME – DEP (Oct. 2018)	Regulatory	9.5	21		migration to groundwater risk modeling	
WE - DEF (OCC. 2018)	Regulatory	5.5	21		For screening solid waste for beneficial use; applied to biosolids by	
					Maine DEP when moratorium on biosolids use imposed in March	
					2019. ME is the only state to screen biosolids for PFAS. NEBRA does	
ME – DEP (2017)	Regulatory	2.5	5.2		not believe these are appropriate for use with biosolids.	
MI – DEQ (2016)	Criteria	350	0.22		Groundwater Surface Water Protection Criteria	
			0.22		PFHxS: 100 ppb, PFNA: 100 ppb. S-1 direct contact res. soil clean-up	
			100		values; lowest in U. S.	
NH – DES (2019)	Regulatory	200	100			
NH – DES (2019) TX – CEQ (2017)	Regulatory Protective level	200	25 / 50			
, ,	v ,				Soil screening level based on dermal contact & ingestion (not	

Table 1. PFAS Regulatory & Guidance Limits for Drinking Water & Other Media (Beecher and Barol	ldi
2020) Complete, updated version available at <u>https://www.nebiosolids.org/pfas-biosolids</u>	

1-The standards and guidance limits included here are the most stringent (lowest values) of which we are aware; some additional jurisdictions have established more lenient (higher value) limits.

2-Sum of 5 of the 6 UCMR 2013 PFAS chemicals: PFNA, PFOA, PFOS, PFHpA, PFHxS (the 6th UCMR PFAS chemical is PFBS)

A review of the data shows a broad range of regulatory standards/guidelines applied to any of the several environmental media listed. This appears to reflect a divergence of opinion regarding the toxicological risk associated with PFAS and the resulting risk assessment methodology used to develop risk-based standards. Also, a substantial difference in PFAS soil/solids standards exists because the public health risk from direct exposure (incidental ingestion and dermal absorption) to PFAS-contaminated soils is deemed to be significantly less than the risk from consumption of drinking water contaminated by PFAS leached from soil or recycled wastes.

A. Drinking Water and Groundwater

Variation in drinking water and groundwater standards included in Table 1 is evident in both the concentrations proposed as standards as well as the contaminants that would be regulated. In 2019, the Michigan Department of Health and Human Services proposed guidance values for PFOA and PFOS of 9 ng/L and 8 ng/L respectively, while USEPA and most states have retained the federal advisory level of 70 ng/L for the total concentration of both. At this time, USEPA has included only PFOA and PFOS in its advisory standards, while states like New Hampshire, Vermont, and Massachusetts propose to regulate 4, 5, or 6 different PFAS, respectively. States proposing to regulate PFAS in drinking water or groundwater are at different stages in the development and adoption of standards. The regulatory climate is changing rapidly. It is likely that the scope and stringency of PFAS groundwater and drinking water standards will only increase.

B. Soil/Solids

At this time, most states have not adopted PFAS standards for soil clean-up, and only Maine has set a standard for recycling of waste solids (biosolids, wastewater, or solid waste). Prior to rising concerns about PFAS contamination of groundwater, some states may have had direct contact standards that were protective of exposure (incidental ingestion and dermal absorption) to PFAS contaminated soils. Generally, these standards have only been adopted for PFOA and PFOS and are in the 300-500 ug/Kg range. For many years, Vermont's 300 ug/Kg standard was the lowest for direct contact with PFOA-contaminated soil. In December 2019, The N.H. Department of Environmental Services (Larson 2019) developed risk-based soil contact standards for four PFAS in the 100-200 ug/Kg range.

However, the overriding public health concern of state environmental agencies is protection of drinking water/groundwater from leaching of PFAS from contaminated soils. Based on various chemical leaching/transport models, some states have proposed soil PFAS standards, primarily for PFOA and PFOS, to protect drinking water sources. For example, Table 1 shows that EPA has suggested soil screening guidance values of 0.172 ug/Kg and 0.378 ug/Kg for PFOA and PFOS, respectively. Maine has imposed screening standards for beneficial use of soil wastes, including biosolids, of 2.5 ug/Kg PFOA and 5.2 ug/Kg PFOS. In Germany, land application of biosolids is regulated, and a combined concentration of 100 ug/Kg has been adopted for land applied sewage sludge (Navarro et al. 2017). Clearly, some of the standards to protect drinking water/groundwater are orders of magnitude more stringent than standards for protection from direct contact with PFAS contaminated soil.

C. Ecological Risk

As mentioned above, USEPA and some states have begun to consider ecological risk to wildlife, both terrestrial and aquatic, from PFAS in the environment. Research has been ongoing, looking at the effects of PFAS on natural ecosystems. Of particular interest are areas where aqueous film-forming foams (AFFF) have been used for fire suppression leaving relatively high concentrations of PFAS in the soil. These sites are frequently severely disturbed and have limited ecological value. However, the mobility of PFAS makes offsite ecological damage a concern (Bridges et al. 2020). Bridges et al. (2020) suggest that, using current wildlife toxicity reference values, appropriate screening values would range from 1 to 10 ug/L for surface water and from 10 to 30 ug/Kg for aquatic sediments. The surface water screening levels are much higher than human drinking water standards, making it unlikely that ecological risk would be a concern for drinking water and groundwater (Bridges et al. 2020). However, aquatic sediments may be a concern.

D. Food and Agricultural Products

Currently, most states and the federal government have not established any kind of standard or guidance for PFAS in food or agricultural products. One exception, Maine, has established an action threshold for milk of 210 ng/L (Maine Dept. of Agriculture, Conservation and Forestry 2020). The U.S. Food and Drug Administration (FDA) has performed limited sampling and testing of the American food supply for PFAS contamination (FDA 2019). FDA has proposed using USEPA's reference doses (RfD) for perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) of 0.02 μ g/Kg body weight (bw)/day, as a measure of food safety (USFDA 2020). In 2008, the European Food Safety Authority (EFSA) established a maximal total daily intake (TDI) of 1,500 ng/Kg bw/day for PFOA (1.5 ug/Kg) and for PFOS, 150 ng/Kg bw/day (0.15 ug/Kg) (Herzke et al. 2013).

III. PFAS Input to Agricultural Production

PFAS are a large chemical family commonly used in commerce and consumer products since the 1940s. These chemicals are persistent, mobile, and widely distributed in the environment throughout the world (Stemmler and Lammel 2010). Ubiquitous use and presence of PFAS in myriad consumer products ensure their release into the environment (soil, water, and air) and waste management systems (landfills, wastewater treatment facilities, compost facilities, etc.) either by direct use of products containing PFAS or by indirect means resulting from unintended contamination of products or migration from point sources (industrial use or production, airports, fire-training areas). Similarly, farms will be the recipients of PFAS contamination from the same direct and indirect sources. The literature suggests that the primary sources of PFAS resulting in contamination of agricultural soils are migration from point sources or areas of high PFAS contamination, use of biosolids or other residuals, and use of contaminated irrigation water (Perez et al. 2017, Ghisi et al. 2018). Most research has focused on activities or situations that unintentionally add PFAS to agricultural soils and ultimately impact the products (meat, milk, feed, produce, etc.) derived from the soil. Less information is available on potential contamination from other products used on farms that are not directly involved in the raising of crops, animals, or other farm products.

A. Indirect Sources of PFAS

The persistence and mobility of PFAS make it ubiquitous in our soil, water, and air. Several studies have attempted to determine background concentrations of PFAS in areas not affected by known point sources and, in some cases, observed PFAS in areas absent of direct human activity (Rankin et al. 2016). Research involving PFAS contamination of agricultural soils following land application of biosolids has frequently detected PFAS (primarily PFOA and PFOS) in control plots and samples (Sepulvado et al. 2011, Gottschall et al. 2017). In a study by the Minnesota Dept. of Health, control sites all had detectable levels of PFAS in soil and produce (Sher et al. 2014). Therefore, it is logical to assume that many farms would be subject to PFAS contamination (background concentrations) as these chemicals cycle through the environment. Table 2 is a compilation of research attempting to quantify background concentrations for various environmental media.

Study	Media	PFOA Conc.	PFOS Conc.	Total PFAS	Comments
Vedagiri et al. 2018	Soil	Range	Range		N=38 to 40, Collected
veuagin et al. 2018		0.059 – 1.84 ug/Kg	0.018 – 2.55 ug/Kg		from U.S. and Canada
				Range ∑PFCA	N=62, Collected from all
Rankin et al. 2016	Soil	Top of range	Top of range	0.029 - 14.3 ug/Kg	continents
	5011	3.44 ug/Kg	3.13 ug/Kg	Range ∑PFSA	32 PFAS considered
				<loq -="" 3.27="" kg<="" td="" ug=""><td></td></loq>	
Zareitalabad et al.	Soil	Median	Median		Based on a review of
2013		0.124 ug/Kg	0.472 ug/Kg		various other studies
Vedagiri et al. 2018	Freshwater	Range	Range		Review of limited data
Veddgirlet di. 2010	sediments	<0.05 - 0.2 ug/Kg	<0.1 - 2.2 ug/Kg		from 4 studies
Zareitalabad et al.	Freshwater	Median	Median		Review of limited data in
2013	sediments	0.27 ug/Kg	0.54 ug/Kg		the literature
		Range	Range		Based on a review of 14
Vedagiri et al. 2018	Surface water	0.45 - 287 ng/L	0.8 – 138 ng/L		studies, means were
Vedugin et ul. 2010		Range of Means	Range of Means		calculated both as
		0.26 – 46 ng/L	0.26 – 46 ng/L		arithmetic and geometric
Zareitalabad et al.	Surface water	Median 3.1 ng/L	Median 3.2 ng/L		Based on a review of
2013					world-wide surface water
					data
	Drinking water				Measured 17 PFAS from
		Median	Median	Median ∑PFAS	25 drinking water
Boone et al. 2018		6.32 ng/L	2.28 ng/L	19.5 ng/L	treatment facilities
					including 24 states in the
					U.S.
	Drinking water	Range	Range		Includes data from all 50
		<20 to 349 ng/L	<40 to 1,800 ng/L		states
Vedagiri et al. 2018		Range of	Range of Geometric		
		Geometric Means	Means		
		<20 to 22 ng/L	<40 to 43 ng/L		

Table 2. Background PFAS Concentrations in Various Environmental Media

It should be noted that, in many of the studies listed above in Table 2, some PFAS was detected in most samples collected to establish background concentrations, with PFOA and PFOS frequently the most abundant compounds. For example, Rankin et al. (2016) determined that all their soil samples

collected from around the world had quantifiable concentrations of perfluorocarboxcylic acids (PFCA) and that perfluoroalkyl sulfonic acids (PFSA) were quantifiable in all but one sample. PFOA and PFOS were the two most commonly detected compounds (Rankin et al. 2016). Boone et al. (2018) tested source water and finished drinking water from 25 public drinking water systems in 24 different states. Source water included both groundwater and surface water. All 50 samples collected (source and treated) had quantifiable levels of PFAS (Boone et al. 2018).

Another significant indirect source of PFAS contamination in agriculture is migration, especially by aerial transmission and deposition, from PFAS point sources such as manufacturing/use facilities. In a joint fact sheet, the N.H. Department of Environmental Services (NHDES) and the N.H. Department of Agriculture, Markets, & Food (NHDAMF) reported on results of soil testing for 160 samples collected from 10 farms in Litchfield and Merrimack, NH within 5 miles downwind of the Saint-Gobain Performance Plastics facility in Merrimack, NH. PFOA results for these samples ranged from below detectable levels to 33 ug/Kg (NHDES and NHDAMF 2016). In a field 10 Km away from a large fluorochemical industrial park in China, the soil concentration for the sum of 12 PFAS compounds (Σ PFAS) ranged from 2.09 ug/Kg to 3.75 ug/Kg (Liu et al. 2019). For the same field, the range of the ∑PFAS in crops was 1.36 ug/Kg to 63.4 ug/Kg (Liu et al. 2019). Brandsma et al. (2018) studied the dispersal of PFOA and GenX (the commercial name for the ammonium salt of 2,3,3,3-tetrafluoro-2-(heptafluoropropoxy) propanoic acid (HFPO-DA)) around a fluoropolymer manufacturing plant in the Netherlands. The study determined the concentration of GenX, PFOA, and the summed concentration of 13 PFAS within plant tissue as well as on the surface of the plant for both leaves and grass within 3 km of the fluoropolymer facility (Brandsma et al. 2018). GenX was detected in all leaf and grass samples, with levels ranging from 1.0 to 27 ug/Kg on a wet weight basis (ww) in/on the grass and from 4.3 to 87 ug/Kg ww in/on leaves (Brandsma et al. 2018). PFOA levels in/on grass and leaves ranged from 9.7 to 11 ug/Kg ww and 0.9 to 28 ug/Kg ww, respectively (Brandsma et al. 2018). Declining concentration gradients of GenX and PFOA with increasing distance suggest that the facility is a point source, with contaminants transported by aerial dispersal (Brandsma et al. 2018). Aerial transport of volatile or particle-bound PFAS can be deposited on plants and surrounding soil. Plants can then absorb the PFAS directly through leaves or uptake from the soil (Ghisi et al. 2018).

Waste management facilities, such as landfills and wastewater treatment facilities (WWTF), have also been implicated as point sources of PFAS via aerial transport and deposition onto agricultural fields (Ghisi et al. 2018). With the exception of land application of biosolids, most of the evidence suggesting that WWTF are PFAS sources, particularly as it relates to agriculture, are anecdotal and not the subject of rigorous research regarding the potential scope and magnitude of PFAS release. Hamid and Li (2016) have documented impacts to surface water and aquatic sediments from treatment plant effluent. Multiple studies have researched the effects on agricultural soils from land application of biosolids (Sepulvado et al. 2011, Gottschall et al. 2017, Washington et al. 2009, Clarke and Smith 2011). But the least studied method of PFAS release from WWTFs is via air emissions. Hamid and Li (2016) report that the magnitude and composition of PFAS air emissions is dependent on the treatment process (activated sludge, extended aeration, facultative lagoons), but provide no data on actual dispersal, deposition, and resulting soil concentrations. Through dispersion modeling, it is estimated that 110 g/yr. to 320 g/yr. of total PFAS are emitted from aeration basins of activated sludge facilities and dispersed to the surrounding environment (Hamid and Li 2016). Theoretically, if distributed evenly over an area of 1000 acres (1.6 sq. mi.), 100 g of total PFAS would result in a soil concentration in the acre-plow-layer of 0.11 ug/Kg total PFAS (see Appendix I, A). In reality, PFAS would likely be distributed over a wider area and at diminishing concentrations with increasing distance from a WWTF, but the potential impact to adjacent farmland is evident. Likewise, Ahrens et al. (2011) observed PFAS emission from two solid waste landfills and estimated annual total PFAS emissions from these landfills at 99 g/yr. and 1000 g/yr., respectively. Once again assuming 100 g/yr. total PFAS emissions, landfills could be expected to have similar impact on agricultural soils as a WWTF in terms of total PFAS, but with a significant shift in the type and abundance of compounds that contribute to total PFAS (Ahrens et al. 2011).

Other indirect sources of PFAS contamination include airports, fire training areas, or other areas of increased PFAS product use or disposal (Ghisi et al. 2018, Xiao et al. 2013). The result of each of these situations can be a "hotspot", an area of very high soil PFAS concentrations. Within a hotspot, PFAS can absorb to soil particles and be moved by soil erosion and deposited downwind (Xiao et al. 2013). According to Xiao et al. (2013), surface soil concentrations for PFOA and PFOS as high as 13 ug/Kg and 18 ug/Kg respectively were detected at a sampling location on the windward side of a hill 4 miles away from a known hotspot (Xiao et al. 2013).

B. Direct Sources of PFAS

Direct sources of PFAS contamination in agriculture include all the chemicals and products intentionally used to aid or enhance agricultural production (fertilizers, feed, pesticides, cleaners, lubricants, etc.) that contribute to PFAS contamination of agricultural sites and products. The use of biosolids and PFAS-contaminated irrigation water have been identified in the literature as the primary direct sources of PFAS contamination in agriculture (Perez et al. 2017, Ghisi et al. 2018). These direct sources of PFAS contamination have been studied and their qualitative and quantitative impacts on agriculture explored in the literature. Other direct sources such as pesticides and fertilizers have received far less attention, and data quantifying their contribution to PFAS contamination of agricultural sites and products, and, ultimately, human exposure, is almost non-existent.

Organic residuals such as biosolids, papermill sludge, septage, and municipal solid waste compost, including food waste composts, have been shown to contain PFAS with various constituents and concentrations (Choi et al. 2019, NEBRA 2019, Schaider et al. 2016, Venkatesan and Halden 2013). All of these residuals are solids or semi-solids discharged from a treatment or waste management system. As waste-derived products, it is more likely than not the residuals will contain PFAS. Several studies between 2001 and 2017 have surveyed and estimated PFAS in sewage sludge (NEBRA 2019). Other studies have documented elevated PFAS concentrations in surface soils and the movement of PFAS down through the vadose zone following land application (Sepulvado et al. 2011, Gottschall et al. 2017). Additional studies have explored uptake of PFAS from biosolids-amended soil (Blaine et al. 2013, Blaine et al. 2014, Wen et al. 2016, Navarro et al. 2017). Venkatesan and Halden (2013) performed a survey of archived biosolids collected in 2001 from 94 WWTFs in 32 states. In this study, the mean 57 Spr for the 13 compounds tested was 539 ±224 ug/Kg dry weight (dw) (Venkatesan and Halden 2013). The two most abundant PFAS were PFOS and PFOA with mean concentrations of 403 ±127 ug/Kg dw and 34 ±22 ug/Kg dw respectively (Venkatesan and Halden 2013). For each dry ton of biosolids land applied to an acre of soil at these PFAS levels, theoretically, soil PFAS concentrations would increase by about 0.5 ug/Kg ∑PFAS, 0.4 ug/Kg PFOS, and 0.03 ug/Kg PFOA in the acre plow layer (see Appendix I, B). Although a strong case can be made that PFAS levels in sludge have abated since 2001, it is likely that current PFAS soil concentrations are the result of more heavily contaminated residuals recycled in the past (NEBRA 2019). While PFAS are considered leachable and tend to move toward groundwater, longchain compounds (>C8) tend to be retained in the vadose zone for years and possibly decades

(Lindstrom et al. 2011, Xiao et al. 2014). Other organic residuals having similar PFAS loads can be expected to have a similar impact on soil concentrations. For example, Choi et al. (2019) assayed municipal solid waste (MSW) composts for 17 perfluoroalkyl acids (PFAA). Composts that included food packaging showed Σ PFAS concentrations ranging from 28.7 to 75.9 µg/Kg (Choi et al. 2019) and PFOA + PFOS concentrations as high as 11.5 µg/Kg. For each dry ton/acre of MSW compost applied to the soil, PFAS soil concentrations could increase by 0.08 ug/Kg Σ PFAS and 0.011 ug/Kg PFOA + PFOS. Even manure may have detectable levels of PFAS. To the extent that animal feed or water contains PFAS contamination, it can be excreted with urine and feces and add to PFAS soil loads when manure is recycled (Kowalczyk et al. 2012, Kowalczyk et al. 2013). In 2017, Maine Department of Environmental Protection (MEDEP) measured PFOA (3.2 ug/Kg) and PFOS (20.3 ug/kg) in dairy cow manure at the Stoneridge Farm, where the original source of PFAS soil contamination is unknown, but likely occurred in the late 1980s (MEDEP, 2017). High levels of PFOS, at least, appear to cycle within an agricultural system.

Ghisi et al. (2018) identify the use of PFAS contaminated groundwater or surface water for irrigation as a potentially significant source of PFAS in agriculture. In a limited study by the Minnesota Department of Health conducted in 2010, PFAS contaminated well water resulted in more frequent detections and higher PFAS concentrations in soil and produce from home gardens than observed in control samples from home gardens where wells were not contaminated (Sher et al. 2018). For PFOA, Zhang et al. (2016) observed a significant correlation between use of irrigation and PFOA in agricultural products, suggesting that the source of PFOA was irrigation water. Blaine et al. (2014A) conducted a study looking at uptake of nine PFAA by lettuce and strawberries when using reclaimed water with Σ PFAA concentrations ranging from 0.2–40 µg/L. The authors concluded that using "reclaimed water" is a pathway for human PFAS exposure (Blaine et al. 2014A). Also, plant uptake and bioaccumulation may have been accentuated by the fact that irrigation does not add organic matter that favors retention of PFAA in the soil (Blaine et al. 2014A). If one assumes a Σ PFAS concentration of 20 ng/L, comparable to the median concentration observed in the Boone et al. (2018) drinking water study, for each acre-inch of irrigation, soil concentrations would theoretically increase by about 0.002 ug/Kg Σ PFAS (see Appendix I, C).

The use of fertilizers and pesticides is a common practice in agricultural production. Direct application to farmland of fertilizers or pesticides containing PFAS could potentially have a significant impact on PFAS loading to the soil and subsequently on human exposure. However, little attention has been focused on these potential PFAS sources in the scientific literature. Most references to PFAS in fertilizers refer to the land application of biosolids. In Kowalczyk et al. (2013), reference is made to "naturally contaminated" feed which was grown on fields where a highly PFAS-contaminated fertilizer was used. Choi et al. (2019) suggest that "contaminated water and applied fertilizers" may have been the source of PFAS in compost produced from yard waste. For pesticides, there is more information available, but little quantitative data on impacts to agriculture. In pesticide formulations, PFAS are frequently PFOS derivatives used as surfactants and wetting agents (United Nations 2011). Except for sulfluramid, an insecticide used in insect baits whose federal registration was withdrawn, most PFAS were considered inert ingredients and were not listed on pesticide labels (United Nations 2011). For example, potassium N-ethyl-N-[(heptadecafluorooctyl) sulfonyl] glycinate (CAS no. 2991-51-7) and 3 [[(heptadecafluorooctyl)sulfonyl]amino]-N,N,N-trimethyl 1-propanaminium iodide (CAS no. 1652-63-7) are two PFOS-related compounds approved for use as surfactants in pesticide formulations (United Nations 2011). Ghisi et al. (2018) has suggested that PFAS used as an emulsifier in phytosanitary

products could contribute to contamination of the edible parts of vegetables. Lassen et al. (2013) have identified use of mono- and di-substituted perfluorinated phosphonic acids (mono-PFPAs and di-PFPAs) as a new class of fluorinated acids in a variety of applications, including defoaming agents in pesticides. PFAS surfactants, although costly, are also highly effective wetting agents and have been used when hydrocarbon surfactants are not effective (Kissa 2001). PFAS surfactants can be efficacious in formulations at concentrations as low as 50-150 ppm (Kissa 2001). At a surfactant concentration of 50 ppm in an herbicide formulation applied at a rate of 2 liters per acre, the soil concentration of the PFAS surfactant used would theoretically increase by 0.11 ug/Kg (see Appendix I, D).

Most of the products or materials mentioned above are applied directly to farmland, and, if PFAS-laden, will add directly to the PFAS burden in the soil. Once in the soil, human exposure to PFAS can occur through uptake into the human food supply (produce, meat, milk, etc.) or through fugitive release from farms and subsequent environmental exposures (drinking water, surface water, dust, etc.).

Some agricultural products are not used in the fields but in other parts of agricultural operations. For example, cleaning products used in dairy operations or animal feed bought from offsite. Again, these potential sources of PFAS have been largely ignored and are hard to quantify. Kowalczyk et al. (2013) did explore the metabolism of PFAS ingested by dairy cows in contaminated feed, but the focus was on metabolic processes and not the prevalence or quantification of PFAS contamination in animal feed in general. The efficacy and versatility of PFAS as surfactants and wetting agents makes their use in cleaning products desirable (Kissa 2001). The two PFOS-related compounds mentioned in the paragraph above are not only used in pesticide formulations, but have also been used in cleaning products (United Nations, 2011). PFAS are such unique and useful chemicals that they are used in a multitude of products that might make their way into farming operations. Frequently, these products do not list PFAS on their labels because they are not "active ingredients" and, until 10-15 years ago, were considered "inert and non-toxic" (United Nations 2011). Consequently, it is likely that PFAS has been inadvertently introduced into agricultural systems from multiple unrecognized sources.

IV. PFAS Contamination of the Environment and Agricultural Products and Compliance with Rapidly Developing Regulations and Standards

The forgoing discussion demonstrates that agricultural systems receive PFAS, both directly and indirectly, and that the result is contamination of agricultural soils and products. Depending on the concentrations, such PFAS contamination may represent a risk of human exposure and potentially adverse health effects. State and federal agencies have been rapidly developing and/or updating regulations and guidelines to prevent health impacts from PFAS in agricultural settings. Does contamination from past or current agricultural practice constitute a risk when measured against evolving regulations?

A. Soils

Regarding PFAS contamination of the environment, the primary concern is release of PFAS to agricultural soils directly from farming or indirectly from off-farm sources. Once in the soil, human PFAS exposure from multiple pathways, including drinking water and food (meat, milk, and produce), can result. Do current PFAS soil concentrations, especially agricultural soils, meet current risk-based standards for protection of human health? Table 3 is a comparison of PFOA and PFOS concentrations

for selected soil standards, soil background, soils following biosolids application, and theoretical increase in soil concentrations from certain potential PFAS sources.

Table 3. A Comparison of PFOA and PFOS Soil Standards to Agricultural Soil Concentrations and
Contributions from Potential Sources

Source	PFOA (ug/Kg)	PFOS (ug/Kg)	Comments				
	Standards						
NH-DES (2019)	200	100	Soil screening level based on dermal contact & ingestion				
ME – DEP (Oct. 2018)	9.5	21	Remedial Action Guidelines (RAGs) for soil cleanup based on migration to groundwater risk modeling				
U. S. EPA (2018)	0.172	0.378	Regional Screening Levels (RSLs) modeled to protect groundwater				
	Back	ground Soil Concentra	tions				
Vedagiri et al. 2018	0.059 - 1.84	0.018 – 2.55	Conc. range				
Rankin et al. 2016	3.44	3.13	Max conc. observed				
Zareitalabad et al. 2013	0.124	0.472	Median values				
	Soil Conc. Following Biosolids Land application						
Washington et al., 2009	50-320	30-410	Conc. range following multiple application of biosolids impacted by industrial production or use				
Sepulvado et al., 2011		2-11	Conc. range following 3 years of biosolids applications				
Gottschall et al. 2016	0.12-0.83	Below detection - 0.4	Conc. range after one biosolids application				
Theo	oretical Annual Incr	ease in Soil Concentra	tion from PFAS Sources				
WWTF Air Emissions (Ahrens et al. 2011)	0.0076	0.049	Assumes even deposition over an area of 1000 acres of 7g PFOS and 45g PFOS				
Landfill Air Emissions (Ahrens et al. 2011)	0.0022	0.0005	Assumes even deposition over an area of 1000 acres of 2g PFOA and 0.5g PFOS				
Biosolids application (Venkatesan and Halden 2013)	0.03	0.40	Assumes application of 1 dry ton/acre containing 34 ug/Kg PFOA and 403 ug/Kg PFOS				
Irrigation water (Boone et al. 2018)	0.0003	0.0007	Assumes 1 acre-inch of irrigation containing 2.3 ng/L PFOA and 6.3 ng/L PFOS				
ISW Compost (Choi et al., 2019) 0.011 (PFOA + PFOS)		Assumes application of 1 dry ton/acre containing 11 ug/Kg PFOA + PFOS					

Except for soils where biosolids impacted by industrial users is applied, agricultural practices and indirect sources of PFAS don't appear to raise soil concentrations above levels of concern for human dermal contact and ingestion as represented by the lowest-in-the-nation N.H. Department of Environmental Services soil screening standards adopted in 2019. However, for standards adopted to protect groundwater and drinking water, represented by the USEPA soil screening level and the Maine Department of Environmental Protection (ME DEP) remedial action guidelines (RAGs), it is clear that observed "background" levels from around the world could occasionally exceed soil concentrations intended to protect such waters. Indirect PFAS sources such as WWTFs, landfills, PFAS manufacturing facilities, and PFAS hotspots can add to the PFAS burden of agricultural soils independent of any farm practices and at levels that might cause soil screening levels for drinking water protection to be exceeded (Liu et al. 2019, Brandsma et al. 2018, Ahrens et al. 2011, Xiao et al. 2013, NHDES and NHDAMF 2016). Regarding farming practices, the literature points to land application of residuals as the

PFAS source mostly likely to raise soil concentrations to levels of concern (Sepulvado et al., 2011, Gottschall et al. 2016).

B. Agricultural Products

For the purposes of this paper, agricultural products refer primarily to meat, milk, and produce. With the exception of milk in Maine, where the Maine Department of Agriculture, Conservation and Forestry (MDACF) has established a screening value of 210 ng/L for PFOS, as yet, there have been no food-specific standards adopted by states or the federal government. Using USEPA's reference doses (RfD) for perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS), of 0.02 µg/Kg bw/day, U.S. FDA has set benchmarks for human PFAS exposure from the food supply (USFDA 2020). The European Food Safety Authority (EFSA) has established a maximal total daily intake (TDI) of 1,500 ng/Kg bw/day for PFOA (1.5 ug/Kg) and for PFOS, 150 ng/Kg bw/day (0.15 ug/Kg) (Herzke et al. 2013). The literature has established the ability of plants to translocate and even bioaccumulate PFAS from the soil into various plant organs and elucidated some of the factors that affect plant uptake: PFAS soil concentration, chemical carbon-chain length and functional groups, plant species and organ, etc. (Ghisi et al. 2018). Likewise, the ability of livestock to absorb PFAS following ingestion and the partitioning of these chemicals in various organs and tissues and elimination via feces, urine, or milk has been explored in the literature (Lupton et al. 2011; Kowalczyk et al. 2012, Kowalczyk et al. 2013). That the public is exposed to PFAS via the food supply is evident. The question is whether that exposure rises to the level of significant risk.

In 2012, FDA tested 12 raw milk samples and 49 retail milk samples from across the country (USFDA 2020). Only one raw milk sample from a dairy farm receiving biosolids had detectable levels of PFAS (USFDA 2029). In 2018-2019, FDA tested the milk and cheese from two farms in New Mexico known to have contaminated groundwater from AFFF use at a neighboring military base (USFDA 2020). The milk from one farm had PFOS concentrations that ranged from 1,620 ng/L to 5,680 ng/L (USFDA 2020). From this limited testing, FDA deemed the milk from this farm a significant human risk and it was discarded (USFDA 2020). In Maine, milk testing was conducted in 2016 on a farm known to have high soil concentrations of PFOA and PFOS, and the milk was determined to contain unacceptable levels of PFOS (MDACF 2019). To ensure the safety of milk sold in Maine, MDACF established its action threshold and performed a state-wide screening of milk sold in the state (MDACF 2019). In all twenty-six samples collected, PFAS was below detectable levels (MDACF 2019). MDACF also tested milk from three commercial dairy farms, two of which had applied biosolids in the past, and none of the dairies had detectable PFAS concentrations in their milk (MDACF 2019). In summer 2020, another dairy farm in Maine was found to have PFOS and other PFAS contamination, with one test showing PFOS in milk at 32,200 ng/L (MDACF 2019). The source of contamination on this farm is being investigated by MDACF, MEDEP, and CDC (MDACF 2019). Based on this admittedly limited evidence, it appears that PFAS concentrations, and particularly PFOS, are unlikely to exceed acceptable levels in milk in the absence of a significant, local source of PFAS contamination.

In 2018 and 2019, FDA conducted two PFAS (16 PFAS) surveys of the general food supply including commodities such as meat, fish, milk, cheese, and produce (USFDA 2020). The vast majority of samples showed no detection of PFAS (USFDA 2020). In both surveys, PFOS was detected in tilapia at concentrations of 87 ng/Kg and 83 ng/Kg, and, in one of the studies, PFOS was detected in ground

turkey at 85.7 ng/Kg. FDA concluded that these limited surveys provided no evidence that PFAS was a human health concern in the general food supply. In 2010 and 2011, Herzke et al. (2013) conducted a survey assessing the levels of 14 PFAS in 20 different vegetables collected in each of four countries (Belgium, Czech Republic, Italy and Norway). Overall, the authors conclude that PFAS concentrations in vegetables was generally low and that perfluorosulfonic acids (PFSA) were usually not detected except for PFOS in spinach (Herzke et al. 2013). The most commonly detected compound was PFOA ranging from 8 to 121 ng/Kg fresh weight (fw) (Herzke et al. 2013). Ultimately, the authors concluded that, based on this limited study, human PFAS exposure from vegetables is low and the intake of PFOA from vegetables in Europe is about four orders of magnitude lower than the recommended EFSA TDI (Herzke et al. 2013). In 2019, Schecter et al. (2010) collected composite samples of 31 different foods from 5 grocery stores in Dallas, Texas and tested for 11 PFAS as well as other synthetic organic chemicals. Of the 11 compounds for which testing was performed, three PFAS were detected during the survey, PFOA, perfluorobutanesulfonic acid (PFBS), and perfluorohexanesulfonic acid (PFHxS). PFOA, the most frequently detected PFAS, was found in 17 of 31 foods at concentrations ranging from 0.07 ug/Kg wet weight in potatoes to 1.80 ug/Kg in olive oil (Schecter et al. 2010). Based on their study results, Schecter et al. (2010) estimated the daily dietary intake for PFOA was 0.060 ug/day. For an 80-kilogram adult, this amounts to a daily intake rate of 0.00075 ug/Kg bw/day. This is well below the FDA and EFSA food safety standards for PFOA quoted above.

For assessing the influence of agricultural practices on PFAS levels in foods, the food surveys reviewed above share a common flaw that limits their value in assessing PFAS in agricultural operations. The samples include some products that might not strictly be considered "farm products" and/or have received significant processing and packaging that could have added PFAS after the raw ingredients left the farm (Schecter et al. 2010). But these surveys of the food supply do provide some confidence that general agricultural output does not expose the public to significant risk from PFAS exposure. However, anomalous situations do exist. For example, after researching the effects of PFAS emissions from a fluorochemical facility in China on soil and crops within 10 kilometers of the facility, Liu et al. (2019) concluded that consumption of crops from the study area could represent a risk to children and adults with a high vegetable diet. Although the soil and crop contamination were not the result of farming practices, Liu et al. (2019) show crops grown in areas of high PFAS contamination, regardless of the source, can lead to excessive PFAS exposure.

As has been previously discussed, farmland can be contaminated with high levels of PFAS both directly and indirectly. An example of direct contamination would be land application of biosolids from a WWTF receiving discharge from PFAS industrial users. An indirect source would include aerial release and deposition from a facility using or manufacturing PFAS. Regardless of the PFAS source, produce, milk, meat, or animal feed produced on a farm having high PFAS soil concentrations would be suspect. In terms of any of these products, what constitutes a "high" concentration is unclear – probably in the high hundreds of ug/Kg for individual PFAS. But in the absence of another obvious PFAS source, farms, especially those that use organic residuals, can be subject to increased regulatory scrutiny.

Groundwater and drinking water contamination are an immediate concern, but ecological risks are also beginning to be investigated (USEPA 2019). A survey of sediments in China's third largest freshwater lake found PFOS concentrations ranging from 4.8 ug/Kg (dw) to 21.7 ug/Kg (dw) (Pan et al. 2010). New Jersey performed a survey of sediments from 11 waterbodies and detected PFOS in 11 out of 14 samples ranging in concentration from 0.289 ug/Kg up to 27.1 ug/Kg (Goodrow et al. 2018). Using

current wildlife toxicity reference values for PFOS, traditional ecological models derive aquatic sediment screening levels of 10-30 ug/Kg (Bridges et al. 2020). By that standard, both Pan et al. (2010) and Goodrow et al. (2018) found PFOS concentrations in sediments, not necessarily associated with a specific point source, that could represent an ecological concern. If a farm was identified as a source of PFOS contamination in aquatic sediments of a local waterbody, it might to be subject to regulatory action to minimize or eliminate ecological risk factors.

V. Conclusions

Are there historical and current sources of PFAS contamination in agricultural production that represent a significant potential risk to public health and the environment when assessed according to environmental standards currently being developed and adopted in the Northeast?

In answer to our central question, the foregoing review of the literature has certainly identified situations on specific farms and farming practices that contribute to PFAS contamination of agricultural soils and products. These levels of PFAS contamination are only considered a public health risk when measured against current and evolving standards (MDACF 2019, USFDA 2020, Liu et al. 2019). Proposed soil standards for the protection of drinking water (see Table 3) are so stringent that so-called "background" PFAS soil concentrations could constitute a threat to drinking water from leaching of PFAS (Rankin et al. 2016). Release of PFAS from local point sources, such as PFAS production or use facilities, can raise PFAS concentrations in nearby farm fields to levels that make underlying groundwater/drinking water susceptible to unacceptable contamination (NHDES and NHDAMF 2016, Liu et al. 2019, Brandsma et al. 2018). These are indirect sources of PFAS to agricultural production, which have nothing to do with farming practices and are most often unrecognized and impossible to eliminate. PFAS are used so ubiquitously in commerce that their release to the environment is inevitable. The chemical persistence and mobility of PFAS ensure that, once released, these chemicals will cycle through the environment, including farmland.

Evidence of risk from direct sources of PFAS and actual farming operations is less compelling. Recycling of wastes onto farmland, such as biosolids land application and irrigation with reclaimed water, have been shown to raise PFAS concentrations in the soil, which increases the risk of groundwater contamination, and with potentially concurrent increases in PFAS in farm products (meat, milk, and produce) (Sepulvado et al., 2011, Lupton et al. 2011, Kowalczyk et al. 2013, Blaine et al. 2014). However, with the exception for milk in Maine, there are no risk-based standards for specific farm products. The three cases of unacceptable PFAS contamination of milk referenced above resulted from unusually high levels of on-farm PFAS contamination, involving the use of wastewater residuals use in one instance and contaminated groundwater in another (MDACF 2019, USFDA 2020). The contamination source in the third instance is being investigated. These cases are considered anomalies, because additional general surveys of retail milk supplies and even sampling of milk from dairy farms using biosolids, have revealed no problems (MDACF 2019, USFDA 2020). Several surveys of the general food supply showed no significant risk to human health from PFAS (Herzke et al. 2013, Schecter et al. 2010, USFDA 2020). From these studies, it can also be inferred that agricultural processes do not pose a significant risk to public health from PFAS in the human diet. In the one study that purported to show a potential human health risk, the indirect source of PFAS was a fluorochemical production facility (Liu et al. 2019).

One deficiency in the literature is the lack of information on PFAS in certain chemicals and products that might be used on farms. Pesticides and cleaning products used for sanitation in milking operations are two examples. Brief references in the literature indicate that PFAS can and has been used in these types of products, but there is very limited published data on PFAS concentrations in product formulations and certainly no data on potential residual PFAS concentrations remaining after use (Kissa 2001, United Nations, 2011). For pesticides and disinfectants, PFAS are generally considered inert ingredients and USEPA does require them to be listed on the label. PFAS are extremely useful and efficacious chemicals, and any product that purports to be a surfactant, wetting agent, dispersant, or anti-foaming agent could contain PFAS (Kissa 2001). Consequently, there are many products containing PFAS that can make their way onto farms and inadvertently add to the PFAS burden in soil and/or directly contaminate farm animals and/or products. When regulatory standards are measured in the ppb and ppt range, accounting for all potential PFAS sources is imperative.

Currently, the scientific and technical literature do not support the proposition that past or current agricultural practices constitute a significant risk to human health from PFAS exposure. Those rare situations where farm products were considered unacceptable for human consumption involved farms with unusually high PFAS concentrations. In some instances, indirect sources of PFAS contamination on farms, such PFAS manufacturing and use facilities, WWTFs, and landfills, may be as problematic as actual farm practices. PFAS contamination will continue to be a problem for the farming community for the following reasons:

- 1) PFAS are persistent and mobile,
- 2) PFAS are extremely useful chemicals, and, as some of these chemicals are banned or heavily regulated, the fluorochemical industry finds alternative PFAS, and
- 3) The regulatory community continues to broaden the list of regulated PFAS and the trend is toward ever lower and more stringent standards.

Literature Cited

- Ahrens, L., Shoeib, M., Harner, T., Lee, S.C., Guo, R., and Reiner, E.J., 2011. Wastewater Treatment Plant and Landfills as Sources of Polyfluoroalkyl Compounds to the Atmosphere. Environmental Science & Technology, 2011, 45, 8098–8105.
- Beecher, N. and Baroldi, L., 2020. PFAS Regulatory & Guidance Limits for Drinking Water & Other Media. Northeast Biosolids and Residuals Association (NEBRA), August. 2020, v.6.
- Blaine, A., Rich, C., Hundal, L., Lau, C., Mills, M., Harris, K., and Higgins, C., 2013. Uptake of Perfluoroalkyl Acids into Edible Crops via Land Applied Biosolids: Field and Greenhouse Studies. Environmental Science & Technology (2013) 47, 14062–14069.

- Blaine, A., Rich, C., Sedlacko, E., Hundal, L., Kumar, K., Lau, C., Mills, M., Harris, K., and Higgins, P., 2014. Perfluoroalkyl acid distribution in various plant compartments of edible crops grown in biosolids-amended soils. Environmental Science & Technology, (2014) 48, 7858–7865.
- Blaine, A., Rich, C., Sedlacko, E., Hyland, K., Stushnoff, C., Dickenson, E., and Higgins, C., 2014 (cited as 2014A). Perfluoroalkyl Acid Uptake in Lettuce (Lactuca sativa) and Strawberry (Fragaria ananassa) Irrigated with Reclaimed Water. Environmental Science & Technology, (2014) 48, 14361–14368.
- Boone, J.S., Vigo, C., Boone, T., Byrne, C. Ferrario, J., Benson, R., Donohue, J., Jane Ellen Simmons, J.E., Kolpin, D.W., Furlong, E.T., and Glassmeyer, S.T., 2018. Per- and polyfluoroalkyl substances in source and treated drinking waters of the United States. Science of the Total Environment, (2019) 653, 359–369.
- Brandsma, S.H., Koekkoek, J.C., van Velzen, M.J.M., and de Boer, J., 2018. The PFOA substitute GenX detected in the environment near a fluoropolymer manufacturing plant in the Netherlands. Chemosphere, (2019) 220, 493-500.
- Bridges, K., Conder, J., and Arblaster, J., 2020. PFAS Toxicology and Risk Assessment: State of the Science. Geosyntec's PFAS Webinar Series, Webinar #2, April 14, 2020.
- Choi, Y.J., Lazcano, R.K., Yousefi, P., Trim, H., and Lee, L.S., 2019. Perfluoroalkyl Acid Characterization in U.S. Municipal Organic Solid Waste Composts. Environmental Science & Technology Letters, 2019, 6,6, 372-377.
- Clarke, B. and Smith, S., 2011. Review of 'emerging' organic contaminants in biosolids and assessment of international research priorities for the agricultural use of biosolids. Environment International, Vol. 37 (2011) 226-247.
- Ghisi, R., Vamerali, T., and Manzetti, S., 2018. Accumulation of perfluorinated alkyl substances (PFAS) in agricultural plants: A review. Environmental Research, (2019) 169, 326–341.
- Goodrow, S.M., Ruppel, B., Lippincott, L., and Post, G.B., 2018. Investigation of Levels of Perfluorinated Compounds in New Jersey Fish, Surface Water, and Sediment. New Jersey Department of Environmental Protection, Div. of Science, Research, and Environmental Health, (2018), SR15-010.
- Gottschall, N., Topp, E., Edwards, M., Payne, M., Kleywegt, S., Lapena, D.R., 2017. Brominated flame retardants and perfluoroalkyl acids in groundwater, tile drainage, soil, and crop grain following a high application of municipal biosolids to a field. Science of the Total Environment, 574, 1345–1359.
- Hamid, H. and Li, L.Y., 2016. Role of wastewater treatment plant in environmental cycling of poly- and perfluoroalkyl substances. Ecocycles: Scientific Journal of European Ecocycles Society, (2016), 2(2) 43-53.

- Herzke, D., Huber, S., Bervoets, L., D'Hollander, W., Hajslova, J., Pulkrabova, J., Brambilla, G., De Filippis, S.P., Klenow, S., Heinemeyer, G., and de Voogt, P., 2013. Perfluorinated alkylated substances in vegetables collected in four European countries; occurrence and human exposure estimates. Environmental Science Pollution Research, (2013), 20, 7930–7939.
- Kissa, E., 2001. Fluorinated Surfactants and Repellents, 2nd ed. Surfactant Science Series, Volume 97, Marcel Dekker, Inc., New York, 2001.
- Kowalczyk, J., Ehlers, S., Furst, P., Schafft, H., and Lahrssen-Wiederholt, M., 2012. Transfer of Perfluorooctanoic Acid (PFOA) and Perfluorooctane Sulfonate From Contaminated Feed Into Milk and Meat of Sheep: Pilot Study. Archives of Environmental Contamination and Toxicology, (2012) 63,288–298.
- Kowalczyk, J., Ehlers, S., Oberhausen, A., Tischer, M., Furst, P., Schafft, H., and Monika Lahrssen-Wiederholt, M., 2013. Absorption, Distribution, and Milk Secretion of the Perfluoroalkyl Acids PFBS, PFHxS, PFOS and PFOA by Dairy Cows Fed Naturally Contaminated Feed. Journal of Agricultural and Food Chemistry, (2013) 61, 2903–2912.
- Larson, D., 2019. Direct Contact Risk-Based Soil Concentrations. Memorandum, N.H. Dept. of Environmental Services, Dec. 11, 2019.
- Lassen, C., Jensen, A.A., Potrykus, A., Christensen, F., Kjølholt, J., Jeppesen, C.N., Mikkelsen, S.H., and Innanen, S., 2013. Survey of PFOS, PFOA and other perfluoroalkyl and polyfluoroalkyl substances. The Danish Environmental Protection Agency, Copenhagen, Denmark, Environmental Project No. 1475, 2013.
- Lindstrom, A., Strynar, M., Delinsky, A., Nakayama, S., McMillan, L., Libelo, L., Neill, M., Thomas, L., 2011. Application of WWTP Biosolids and Resulting Perfluorinated Compound Contamination of Surface and Well Water in Decatur, Alabama, USA. Environmental Science & Technology, 45 (19), 8015–8021.
- Liu, Z., Lua, Y., Song, X., Jones, K., Sweetman, A.J., Johnson, A.C., Zhang, M., Lua, X., Su, C., 2019. Multiple crop bioaccumulation and human exposure of perfluoroalkyl substances around a mega fluorochemical industrial park, China: Implication for planting optimization and food safety. Environment International, (2019), 127, 671–684.
- Maine Dept. of Agriculture, Conservation and Forestry 2020. Per- and Polyfluoroalkyl Substances (PFAS): Is Maine milk Safe? MDACF website, 2020 https://www.maine.gov/dacf/ag/pfas/index.shtml.
- Maine Dept. of Environmental Protection, 2017. Stone Farm, Arundel, Sample Collection Data Report Summary. June 15, 2017
- North East Biosolids & Residuals Association (NEBRA), 2019. NEBRA Literature Review: PFAS & Wastewater Residuals. NEBRA, v.2.0, November 2019.

- N.H. Dept. of Environment Service (NHDES) and N.H. Dept. of Agriculture, Markets, and Food (NHAMF), 2016. Results from agricultural soil screening testing for perfluorooctanoic acid (PFOA) in Southern New Hampshire. Joint department fact sheet, Aug. 10, 2016.
- Pan, G., Zhou, Q., Luan, X., and Fu, Q.S., 2010. Distribution of Perfluorinated Compounds in Lake Taihu (China): Impact to Human Health and Water Standards. Research Center for Eco Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China, 2010.
- Perez, A., Poteete, C., Louie, F., Garner, L., and Scott, P., 2017. State of the science and meta-analysis of crop uptake of per- and polyfluoroalkyl substances (PFAS). Presented at the DIOXIN 2017 Symposium, Vancouver Canada.
- Rankin, K., Mabury, S.A., Jenkins, T.M., and Washington, J.W., 2016. A North American and global survey of perfluoroalkyl substances in surface soils: Distribution patterns and mode of occurrence. Chemosphere, (2016), 161, 333-341.
- Sepulvado, J., Blaine, A., Hundal, L., Higgins, C., 2011. Occurrence and Fate of Perfluorochemicals in Soil Following the Land Application of Municipal Biosolids. Environmental Science and Technology, 45 (19), 8106–8112.
- Schaider, L., Ackerman, J., and Rudel, R., 2016. Septic systems as sources of organic wastewater compounds in domestic drinking water wells in a shallow sand and gravel aquifer. Science of the Total Environment, 547 (2016) 470–481.
- Schecter, A., Colacino, J., Haffner, D., Patel, K., Opel, M., Päpke, O., and Birnbaum, L., 2010.
 Perfluorinated Compounds, Polychlorinated Biphenyls, and Organochlorine Pesticide
 Contamination in Composite Food Samples from Dallas, Texas, USA. Environmental Health
 Perspectives, volume 118, number 6, June 2010.
- Scher, D., Kelly, J., Huset, C., Hoffbeck, R., Souther, K., Hansen, E., Korinek, J., Messing, R., and Yingling,
 V., 2014. The Perfluorochemicals in Homes and Gardens Study (PIHGS): Implications for human exposure assessment. Minnesota Department of Health, St. Paul, MN, 2014.
- Stemmler, I. and Lammel, G., 2010. Pathways of PFOA to the Arctic: variabilities and contributions of oceanic currents and atmospheric transport and chemistry sources. Atmospheric Chemistry and Physics, (2010), 10, 9965–9980.
- Navarro, I., de la Torre, A., Sanz, P., Porcel, M.A., Pro, J., Carbonell, G. and Martínez, M., 2017. Uptake of perfluoroalkyl substances and halogenated flame retardants by crops grown in biosolids amended soils. Environmental Research, (2017), 152, 199–206.
- North East Biosolids & Residuals Association (NEBRA), 2019. NEBRA Literature Review: PFAS & Wastewater Residuals. NEBRA, v.2.0, November 2019.
- United Nations Stockholm Convention on Persistent Organic Pollutants, 2011. Guidance on alternatives to perfluorooctane sulfonic acid and its derivatives. UNEP/POPS/POPRC.6/13/Add.3/Rev.1

- U.S. Environmental Protection Agency, 2019. EPA's Per- and Polyfluoroalkyl Substances (PFAS) Action Plan. EPA 823R18004, February 2019, 72 pp.
- U.S. Food & Drug Administrative, 2020. Per and Polyfluoroalkyl Substances (PFAS). U.S. FDA Webpage, https://www.fda.gov/food/chemicals/and-polyfluoroalkyl-substances-pfas, July 31, 2020.
- Venkatesan, K, and Halden, R., 2013. National inventory of perfluoroalkyl substances in archived U.S. biosolids from the 2001 EPA National Sewage Sludge Survey. Journal of Hazardous Materials, 252–253, (2013), 413–418.
- Washington, J., Ellington, J., Hoon, Y., and Jenkins, T., 2009. Results of the Analyses of Surface Soil Samples from Near Decatur, Alabama for Fluorinated Organic Compounds. U.S. EPA, EPA Office of Research and Development.
- Wen, B., Wu, Y., Zhang, H., Liu, Y., Hu, X., Huang, H., and Zhang, S., 2016. The roles of protein and lipid in the accumulation and distribution of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) in plants grown in biosolids-amended soils. Environmental Pollution, 216 (2016) 682-688.
- Xiao, F., Gulliver, J., Simcik, M., 2013. Transport of Perfluorochemicals to Surface and Subsurface Soils. Center for Transportation Studies University of Minnesota, Report No. CTS 13-17.
- Xiao, F., Simcik, M., Halbach, T., Gulliver, J., 2014. Perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) in soils and groundwater of a U.S. metropolitan area: Migration and implications for human exposure. Water Research, 72 (2015), 64-74.
- Zhang, Y., Tan, D., Geng, Y., Wang, L., Peng, Y., He, Z., Xu, Y., and Liu, X., 2016. Perfluorinated
 Compounds in Greenhouse and Open Agricultural Producing Areas of Three Provinces of China:
 Levels, Sources and Risk Assessment. International Journal of Environmental Research & Public
 Health, 2016, 13, 1224.

Appendix I

PFAS Loading Calculations

Note: A PFAS loading calculations spreadsheet is available at https://www.nebiosolids.org/research-pfas-biosolidsresiduals/ (contact NEBRA for access)

- A. PFAS soil loading resulting from atmospheric transport and deposition of PFAS from wastewater treatment facilities (WWTF)
 - Assume 100 grams of total PFAS (Hamid and Li 2016) are distributed evenly over 10 acres adjacent to a WWTF,
 - 100 grams total PFAS ÷ 10 acres = 10 g total PFAS/A,
 - 10 g/A ÷ 1000 g/kg = 0.01 kg/A,
 - 0.01 kg/A X 2.2 lbs./kg = 0.022 lbs./A,
 - Assume an acre plow layer weighs 2 million pounds/A,
 - 0.022 lbs/A ÷ 2 million lbs. = 0.011 lbs./million lbs./A = 0.011 ppm/A = 0.011 mg/kg/A,
 - 0.011 mg/kg/A X 1000 ug/kg = 11 ug/kg/A, and
 - Total PFAS soil concentrations would increase by 11 ug/kg over a 10-acre area adjacent to a WWTF emitting 100 g total PFAS per year or 0.11 ug/kg over a 1000-ac area.
- B. SPFAS, PFOS, and PFOA soil loading from each dry ton of "average" biosolids land applied
 - Assume biosolids containing the following mean concentrations are land applied; ∑PFAS = 539 ug/kg dry weight (dw), PFOS = 403 ug/kg dw, and PFOA = 34 ug/kg dw (Venkatesan and Halden 2013),
 - For ∑PFAS, 539 ug/kg ÷ 1000 ug/mg = 0.539 mg/kg = 0.539 ppm ∑PFAS dw,
 - 0.539 ppm = 0.539 lbs. Σ PFAS per million lbs. biosolids,
 - 1 million lbs. ÷ 2000 lbs./ton = 500 tons, 0.539 lbs. ÷ 500 T = 0.0011 lbs./T,
 - 0.0011 lbs./T applied to an acre of soil, 0.0011 lbs./T X 1 T/A = 0.0011 lbs./A,
 - Assume an acre plow layer weighs 2 million pounds/A,
 - 0.0011 lbs./A ÷ 2 million lbs./A = 0.00054 lbs. per million lbs./A = 0.00054 ppm = 0.00054 mg/kg,
 - 0.00054 mg/kg X 1000 ug/mg = 0.54 ug/kg,
 - ∑PFAS soil concentrations would increase by 0.54 ug/kg for each dry ton of biosolids land applied, and
 - Using the same calculations for PFOS and PFOA would yield theoretical increases in soil concentrations of 0.40 ug/kg and 0.03 ug/kg respectively for each dry ton of biosolids land applied.

- C. **SPFAS** soil loading from each acre-inch of irrigation using a PFAS contaminated water source
 - Assume the source of water for irrigation is contaminated with ∑PFAS at a concentration of 20 ng/L (Boone et al. 2013),
 - One acre-inch of irrigation = 102,790 liters/A,
 - 20 ng/L Σ PFAS x 102,790 L/A = 2,055,800 ng Σ PFAS applied over one acre,
 - 2,055,800 ng/A ÷ 1000 ug/ng ÷ 1000 ug/mg ÷ 1000 mg/g ÷ 1000 g/kg = 0.0000021 kg ∑PFAS/A,
 - 0.0000021 kg/A X 2.2 lbs./kg = 0.0000045 lbs. ΣPFAS/A,
 - Assume an acre plow layer weighs 2 million pounds/A,
 - 0.0000045 lbs./A ÷ 2 million pounds/A = 0.0000022 lbs./million lbs. = 0.0000022 ppm ∑PFAS,
 - 0.0000022 ppm = 0.0000022 mg/kg X 1000 ug/mg = 0.0022 ug/kg, and
 - Σ PFAS soil concentrations would increase by 0.002 ug/kg for each acre-inch of irrigation applied.
- D. PFOS-based surfactant soil loading from the application of an herbicide using PFAS as an inert ingredient
 - Assume 2 liters of a common herbicide is applied to an acre of crop land and a PFOS-based surfactant is part of the herbicide formulation at a concentration of 50 ppm (Kissa 2001),
 - 50 ppm = 50 mg/L X 2 liters/A = 100 mg/A,
 - 100 mg/A ÷ 1000 g/mg ÷ 1000 kg/g = 0.0001 kg/A,
 - 0.0001 kg/A X 2.2 lbs./kg = 0.00022 lbs./A,
 - Assume an acre plow layer weighs 2 million pounds/A,
 - 0.00022 lbs./A ÷ 2 million lbs./A = 0.00011 lbs./million lbs. = 0.00011 ppm = 0.00011 mg/kg,
 - 0.00011 mg/kg X 1000 ug/mg = 0.11 ug/kg, and
 - The concentration of PFOS-based surfactants in the soil would increase by 0.11 ug/kg after each application.

Appendix II

Compiled Abstracts and Notes

See separate document available at <u>https://www.nebiosolids.org/research-pfas-biosolidsresiduals/</u> (contact NEBRA for access)