SLUDGE IN THE GARDEN

Toxic PFAS in Home Fertilizers Made From Sewage Sludge

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ECOLOGY CENTER
Healthy People, Healthy Planet

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EXECUTIVE SUMMARY

Many home gardeners buy compost or commercial soil amendments to enhance soil nutrition. But new tests reveal concerning levels of toxic chemicals known as PFAS in fertilizer products which are commonly made from sewage sludge. These "forever chemicals" were found in all of the nine products tested by the Ecology Center of Michigan and Sierra Club and marketed as "eco" or "natural" and eight of the nine exceeded screening levels set by the state of Maine. PFAS in fertilizers could cause garden crops to be a source of exposure for home gardeners.

PFAS are per- and poly-fluoroalkyl substances, a class of widely used industrial chemicals, that persist for decades in the environment, many of which are toxic to people. In most places, industries are currently allowed to flush PFAS-containing waste into wastewater drains that flow to treatment plants. The chemicals are not removed during sewage treatment and instead settle in solid materials that are separated out from liquids in the treatment process.

Americans generate massive quantities of sewage waste each day. Nearly half of sewage sludges are treated to kill pathogens and then spread on farms, pastures, and wildlands for disposal, where nutrients like nitrogen improve soil productivity. The wastewater industry and EPA call these "biosolids." Unfortunately, biosolids carry a variety of persistent and toxic chemicals, in addition to PFAS, which can threaten our food supply and contaminate water sources.

The Sierra Club and the Ecology Center identified dozens of home fertilizers made from biosolids. We purchased nine fertilizers: Cured Bloom (Washington DC), TAGRO Mix (Tacoma, Washington), Milorganite 6-4-0 (Milwaukee, Wisconsin), Pro Care Natural Fertilizer (Madison, Georgia), EcoScraps Slow-Release Fertilizer (Las Vegas, Nevada), Menards Premium Natural Fertilizer (Eau Claire, Wisconsin), GreenEdge Slow Release Fertilizer (Jacksonville, Florida), Earthlife Natural Fertilizer (North Andover, Massachusetts), and Synagro Granulite Fertilizer Pellets (Sacramento area, California).

Our tests reveal that American gardeners can unwittingly bring PFAS contaminants home when they buy fertilizer that is made from sludge-biosolids. Eight of the nine products exceeded screening limits for two chemicals—PFOS or PFOA—set by Maine, the state with the most robust action on PFAS in biosolids. The chemicals were measured at levels that would not be acceptable for the state's agricultural soils. Of the 33 PFAS compounds analyzed in the products, 24 were detected in at least one product. Each product contained from 14 to 20 detectable PFAS compounds. Additional tests showed they also contained two to eight times greater mass of precursor compounds and hundreds to thousands of times more unidentifiable synthetic fluorine compounds.

Our testing provides a snapshot of PFAS levels in complex wastewater systems. The findings are in line with national surveys of PFAS in sludge-biosolids, and academic studies testing biosolids-based fertilizers and composts. Available evidence suggests that PFAS and related chemicals in sewage sludge could jeopardize the safety of the commercial food supply and home gardens. We recommend home gardeners do not purchase biosolids-derived fertilizers for use on fruit and vegetable beds. For the large-scale problem of disposing of sewage waste, however, simple solutions are elusive. The federal government and most states have done little to study the issue, let alone address it.

Our test results suggest that urgent changes are needed to halt the unnecessary uses of PFAS in commerce and minimize the amounts that are discharged into our wastewater system. EPA Administrator Michael Regan has pledged immediate action to reduce the threats posed by PFAS uses, but the agency's anemic responses to date, as well as structural barriers created by key environmental laws, make quick action unlikely and hinder even the most common-sense measures to contain the chemical crisis.

The EPA and states must take immediate action to keep PFAS and other persistent chemicals out of the wastewater system, biosolids, and the food supply. This means preventing industrial polluters from discharging PFAS in their wastewater drains. Agencies must survey the hazard of food production on highly contaminated soils and regulate land application of biosolids with high levels of PFAS and other chemicals. Industry must pay for the damages that PFAS production and use poses to people and the environment, including costly cleanups of contaminated places. The most efficient and effective way to protect people from the growing threat of PFAS exposure is to end the use of PFAS, with limited exemptions.

INTRODUCTION

Treated sewage sludge, or "biosolids," are commonly applied to farmlands and sold directly to home gardeners as compost, soil amendment, or fertilizer. We identified at least 30 different commercial fertilizers made from sewage sludge and sold at retailers like Lowe's, The Home Depot, Ace Hardware, and Menards, and direct from manufacturing or wholesale sites.

Many bear terms like "eco," "natural," or "organic" on the label. While biosolids are not allowed to be applied on farms growing certified organic fruits, vegetables, or dairy products (FDA 2013), one of the biosolids-based fertilizers we tested is used in school gardens in Washington, DC.

The EPA regulates pathogens and heavy metals like lead, cadmium, and mercury in biosolids, but does not set limits for other chemical contaminants that accumulate in sewage and wastewater, including PFAS (per- and poly-fluoroalkyl substances), which are a diverse group of synthetic (human-made) fluorochemicals used widely for their useful qualities of thermal and chemical resistance and persistence. PFAS are generally not well-regulated under national air, water, chemicals, or waste laws, but are widely understood to pose a serious health risk to people, wildlife, and the environment (Kwiatkowski 2020, Fenton 2020).

We tested a sample of nine products marketed to home gardeners for PFAS. Most products contained 100 percent sludge-biosolids. But none bear any warnings about the potential inclusion of PFAS or most other chemical contaminants. Just one had a warning about molybdenum for forage crops.

We sent the products to two commercial laboratories that used several methods to determine the concentrations of PFAS and related chemicals. Each product contained 14 to 20 of the 33 tested PFAS chemicals, with total concentrations ranging from 38 to 233 parts per billion (ppb). For reference, this is similar to concentrations found in fish collected in highly polluted areas and thousands of times higher than the amounts that are regulated in drinking water. PFAS from highly contaminated sludges from industrial sites have been determined to contaminate local water supplies and agricultural products. We are concerned that the concentrations of PFAS in fertilizers made from sludge-biosolids could lead to accumulation in food plants grown in fertilized beds in home gardens or agricultural fields.

The tests also revealed an additional two- to eight-fold higher concentration of "PFAS precursors," and roughly 150 to 6,000 times greater mass of unknown fluorochemicals in the fertilizer products. These mystery compounds, likely unmeasured PFAS chemicals or fluorine-based polymers, should be investigated to determine their sources as well as the hazards they pose to food crops and people.

We recommend people avoid unnecessary exposure to PFAS by not applying sewage sludge-based fertilizer or composts to home gardens. Shoppers can check the "Guaranteed Analysis" section of the product label that discloses the source of the fertilizer. If purchasing compost or topsoil, check product information for terms like "biosolids," "residuals," or "municipal waste," which could indicate it is made from sewage.

But quick fixes are more elusive for the threat sludge-based biosolids pose to the commercial food supply. The EPA requires biosolids be tested for phosphorus, pathogens, and nine heavy metals before land application in its Rule 503, but does not set any limits for any PFAS compounds (USEPA 1994). The EPA provides an annual update about the number of unregulated chemicals that have been detected in the materials (USEPA 2021a). In a 2018 report, the EPA's Inspector General raised concerns about gaps in its oversight of biosolids materials (USEPA 2018). It cautioned that the agency should consider the cumulative hazards posed by other persistent contaminants in biosolids and revise its public messages about biosolids safety (USEPA 2018).

The EPA and state governments can and should take urgent steps to prevent PFAS from being discharged from industrial drains into the wastewater system, using authorities under the Clean Water Act. Addressing the more dispersed uses of PFAS from consumer products is a harder task.

Despite being highly persistent, bioaccumulative, mobile, and toxic to people, PFAS chemicals are virtually unregulated. Three PFAS chemicals—PFOS, PFOA and PFHxS—are in the process of being phased out of commerce under the global United Nations Stockholm Convention, but thousands more are commonly used in a variety of consumer and industrial products.

The Trump-era EPA stalled listing these clearly harmful chemicals under the nation's clean air and water and

waste laws, and continued to approve new, poorly studied PFAS chemicals as alternatives to PFOS and PFOA. In the meantime, industries like metal plating, paper, and textile manufacturing continue to legally dump the chemicals into wastewater drains.

Current EPA Administrator Michael Regan has pledged to regulate some industries that discharge high levels of PFAS in wastewater, beginning with the companies who make PFAS chemicals. The EPA is gathering data on PFAS uses in a few key industries, with the intent of eventually requiring them to filter out PFAS or change their processes to avoid using PFAS. This is a slow process, however, and states should not wait years for the EPA to finalize national actions. The Clean Water Act also allows the EPA to set contaminant limits for biosolids, and the agency has pledged to do a safety screening for the hundreds of unregulated contaminants detected in biosolids in the next two years (USEPA 2019).

Several states, including **Maine**, **Massachusetts**, and **Michigan** are further along in addressing the issue.

After discovering high levels of PFAS in milk produced from dairy cattle feeding on contaminated fields, Maine is measuring the amount of PFAS in biosolids and ensuring that the materials do not contaminate agricultural lands (Maine 2021). When biosolids exceed screening levels, the state requires modeling or testing to ensure the repeat application has not pushed agricultural fields over the screening level of 2.5 ppb for PFOA and 5.2 ppb for PFOS. Maine's testing of one contaminated dairy found that the PFOS and PFOA levels in milk exceeded the concentrations it measured in the soils themselves. Unfortunately, Maine still allows contaminated biosolids to be spread on other agricultural lands.

Michigan is working to prevent contamination of biosolids by regulating the industries that dump PFAS chemicals down wastewater drains. It has identified a number of wastewater treatment plants with high levels of PFAS and requires some upstream industries to change practices or filter wastewater to remove PFAS (Michigan 2021). This is a slow and data-intensive process, yet it is highly effective in removing PFAS from wastewaters and therefore sludge. Interventions at seven highly contaminated wastewater systems reduced PFOS levels in biosolids by 90 to 99 percent. The state didn't study or report the impact these measures had on other PFAS chemicals. Unfortunately Michigan's newly proposed screening levels for PFOS and PFOA in sludge are much higher than Maine's limits, and will be less protective of agricultural fields in the state.

Michigan also has yet to institute testing protocols for agricultural products, like milk, that may be affected by biosolids use. Kay Fritz, a toxicologist at the Michigan Department of Agriculture and Rural Development,hinted at the thorny politics that slow this type of important monitoring and disclosure:

"If you test the milk and you find PFAS, then you have to tell the farmer. Then the farmer has to tell the co-op that takes the milk. Then, they say 'Oh no, we don't want any PFAS milk.' Then you put this farmer out of business immediately."

This approach does not, of course, eliminate the public health threat of PFAS, it just eliminates public access to information about the extent of the threat.

Colorado adopted new "narrative" standards for five categories of PFAS chemicals in 2020 and has surveyed PFAS levels in state surface waters. These standards will allow the state to require wastewater testing in key industries and will ultimately lead to permit restrictions on industrial sources (Colorado 2020).

Massachusetts, Vermont, and New Hampshire are testing PFAS levels in biosolids. Massachusetts has the long-term goal of "virtually eliminating" PFAS in biosolids but has not set a screening limit or management plan to achieve this goal (Massachusetts 2021). Vermont will require annual testing of soil, ground water, and plant tissue (Vermont 2020). New Hampshire instructs wastewater systems to test for PFAS using guidelines developed by the industry group the North East Biosolids & Residuals Association, and not apply sludge with high concentrations to land, but it doesn't clarify the numeric screening level online (New Hampshire 2021).

RECOMMENDATIONS

Ultimately, the only way to keep PFAS and other persistent chemicals out of biosolids is to limit their production and use. In the interim, federal and state governments as well as industry and wastewater treatment systems must take urgent action to clean up biosolids.

- **The federal government** must urgently act to end PFAS uses in commercial products and releases from industrial sites. To address PFAS in wastewater, it must set limits for PFAS and other persistent chemicals in biosolids products applied to farmlands or home gardens. The EPA must promptly list all PFAS in the Clean Water Act, which will allow state and federally granted wastewater permits to require testing and treatment to remove PFAS in wastewater. Immediate action is needed both for the industries producing PFAS and PFAS users—including metal plating, paper, textiles, and plastics—and industries using PFAS for fire suppression.
- **States** should regulate PFAS in their Clean Water Act rules. Forty-seven states have direct oversight over chemicals discharged into the wastewater system. They should investigate contamination of food and farmland, set up new management systems to keep contaminated biosolids from contaminating food and water supplies, and pursue remedial actions against industrial polluters. Most states also have the power to set rules related to biosolids disposal.
- The **chemical industry** must stop releasing PFAS into air, wastewater, surface water, and as solid wastes. It must immediately look for safe alternatives to PFAS in all products. PFAS chemicals should be phased out, with only limited exceptions for essential uses where safer alternatives are not presently available, such as certain materials used in medical devices.
- **Wastewater treatment plants** must investigate sources of PFAS discharged into their systems and intervene to capture PFAS before it enters their systems. This is important for all systems, not just those that sell biosolids-based fertilizers to home gardeners and landscaping services.
- **Agricultural producers** should not apply biosolids to their crop and pasture lands. Doing so risks permanently contaminating their soils with PFAS and other long-lasting chemical contaminants.
- **Home and community gardens** should check the "Guaranteed Analysis" label of fertilizers to ensure products are not made from biosolids. Ask landscapers or commercial providers if soil, topsoil, composts or other garden products are made from "biosolids"—which they may describe using vague terms like "residuals" or "municipal wastes"—and avoid purchasing them.
- **Companies making biosolids into home-use fertilizers** should more clearly disclose the presence of potentially harmful chemicals in their products and modify labels to direct these to be used only on lawns, ornamental plants, and other non-food uses.

BIOSOLIDS-BASED FERTILIZERS CONTAIN A VARIETY OF HARMFUL PFAS AND UNKNOWN FLUOROCHEMICALS

The Sierra Club and the Ecology Center identified and purchased fertilizer products made from biosolids and that are marketed directly for home use.

We identified more than 30 biosolids-based fertilizers, composts, and soil amendments for sale and purchased nine products from eight states and the District of Columbia. We sent the products to commercial laboratories that used three methods to measure synthetic fluorochemicals in the products.

Table 1: Fertilizers Tested

Product Name	Where Purchased	Biosolids Source	Percent Biosolids
Pro Care Natural Fertilizer	Lowes	Georgia—multiple locations	85.5 - 91.5
EcoScraps Slow Release Fertilizer	The Home Depot	Unknown—company based in Nevada	100
Milorganite 6-4-0 Fertilizer	The Home Depot	Milwaukee, WI—Metropolitan Sewerage District	100
Cured Bloom Soil Conditioner	W.S. Jenks & Sons Washington, DC	Washington, DC Water—Blue Plains Advanced Wastewater Treatment Plant	100
Menards Premium Natural Fertilizer	Menards	Unknown—company based in Wisconsin	100
GreenEdge Slow Release Fertilizer	The Home Depot	Jacksonville, FL—JEA sewer collection system	100
Earthlife Natural Fertilizer	York Woods Tree & Products, Eliot, ME	Quincy, MA—New England Fertilizer Company (NEFCO)	100
Synagro Granulite Fertilizer Pellets	Sacramento, CA Pelletizer	Elk Grove, CA— Sacramento Regional Wastewater Treatment Plant	100
TAGRO Mix	Ace Hardware	Tacoma, WA—Central Wastewater Treatment Plant	50

SPECIFIC PFAS

SGS AXYS Analytical Services measured levels of 33 specific PFAS chemicals in the soil products; 24 were detected. The sum of measured PFAS ranged from 38 to 223 ppb. The highest was in Cured Bloom soil conditioner, at 223 ppb. The laboratory analyzed two samples collected from a single bag of GreenEdge Slow Release fertilizer for validation.

Figure 1: PFAS in Home Fertilizers, parts per billion

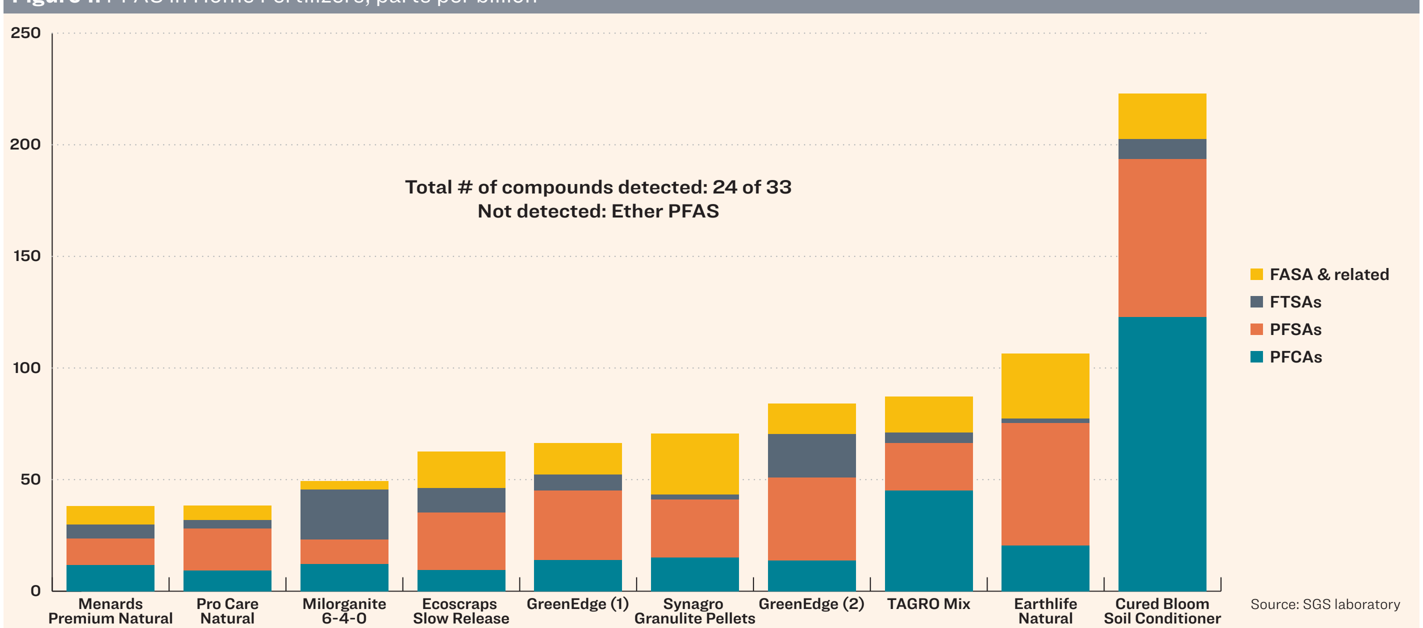


Table 2: PFAS Chemicals Analyzed in Home Fertilizers*

Perfluoroalkyl sulfonates (PFSAs)	Perfluoroalkyl carboxylates (PFCAs)	PFAS ethers	Perfluoroalkane sulfonamides (FASAs & Related)	Fluorotelomer sulfonic acids (FTSAs)
PFBS	PFBA	HPO-DA (GenX)	PFOSA	4:2 FTS
PFPeS	PFPeA	ADONA	N-MeFOSA	6:2 FTS
PFHxS	PFHxA	9Cl-PF3ONS*	N-EtFOSA	8:2 FTS
PFHpS	PFHpA	11Cl-PF3OUdS*	MeFOSAA	
PFOS	PFOA	*components of F-53B	EtFOSAA	
PFNS	PFNA		N-MeFOSE	
PFDS	PFDA		N-EtFOSE	
PFDoS	PFUnA		**Red** = detected in at least one product (multiple products in most cases).	
	PFDoA		**Black** = not detected	
	PFTrDA			
	PFTeDA			

**Analyzed by SGS Laboratory using Liquid Chromatography with tandem mass spectrometry (LC/MS/MS)*

PFOA AND PFOS

Maine is one of the only states that has guidelines to prevent biosolids from contaminating agricultural lands and groundwater. It requires all biosolids be tested for three PFAS chemicals prior to land application. When concentrations exceed a screening limit of 2.5 ppb for PFOA, 5.2 ppb for PFOS, and 1900 ppb for PFBS, the agricultural fields must also be tested to ensure repeated applications haven't led to soil concentrations over the screening limit. Eight of the nine products exceeded one or two of the state-based screening limits for biosolids and agricultural soils in Maine.

Table 3: 8 of 9 fertilizers exceeded screening limits for PFOS or PFOA in Maine

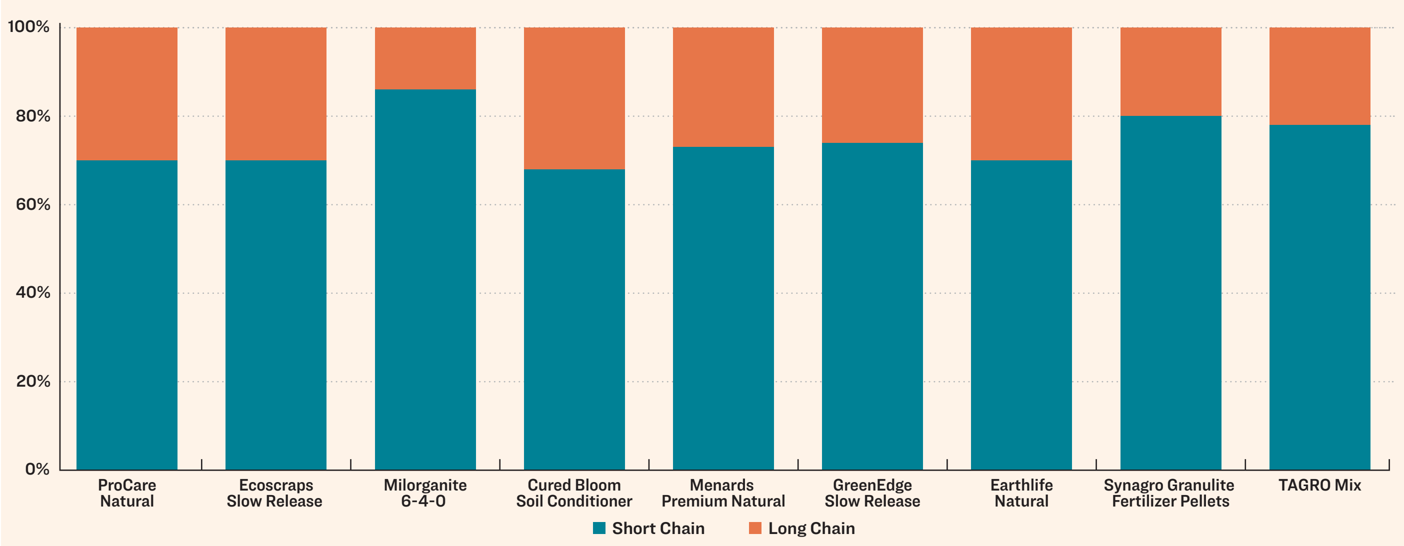
Product Name	PFOA	PFOS
Cured Bloom Soil Conditioner	23.8	22.1
Earthlife Natural Fertilizer	2.75	17.3
EcoScraps Slow Release Fertilizer	1.20	16.9
GreenEdge Slow Release Fertilizer (1)	1.39	13.5
GreenEdge Slow Release Fertilizer (2)	1.66	12.9
Menards Premium Natural Fertilizer	1.01	9.05
Milorganite 6-4-0 Fertilizer	0.67	8.66
Pro Care Natural Fertilizer	0.94	14.9
Synagro Granulite Fertilizer Pellets	0.95	3.71
TAGRO Mix	7.51	7.92

Biosolids limits: PFOA 2.5, PFOS 5.2 ***red*** *= exceeds screening level,* ***green*** *= below*

"SHORT-CHAIN" PFAS CHEMICALS

In addition to PFOA and PFOS and other related "long-chain" PFAS that have been largely removed from intentional production in the US, the products also contained "short-chain" PFAS which are generally unregulated, making up about 70 percent or more of the total amount of tested PFAS chemicals. Scientists and advocates have raised concerns that these "short-chain" PFAS chemicals are not safer than the PFOS and PFOA-type chemicals that they are replacing (Kwiatkowski 2020). They too persist in the environment, and are more mobile in water, harder to filter out with conventional water treatment methods, and appear to impact the same parts of the body as the first generation or "long-chain" PFAS.

Figure 2: Home Fertilizers Contain More Currently-Used "Short Chain" PFAS chemicals than "Long Chain" Chemicals

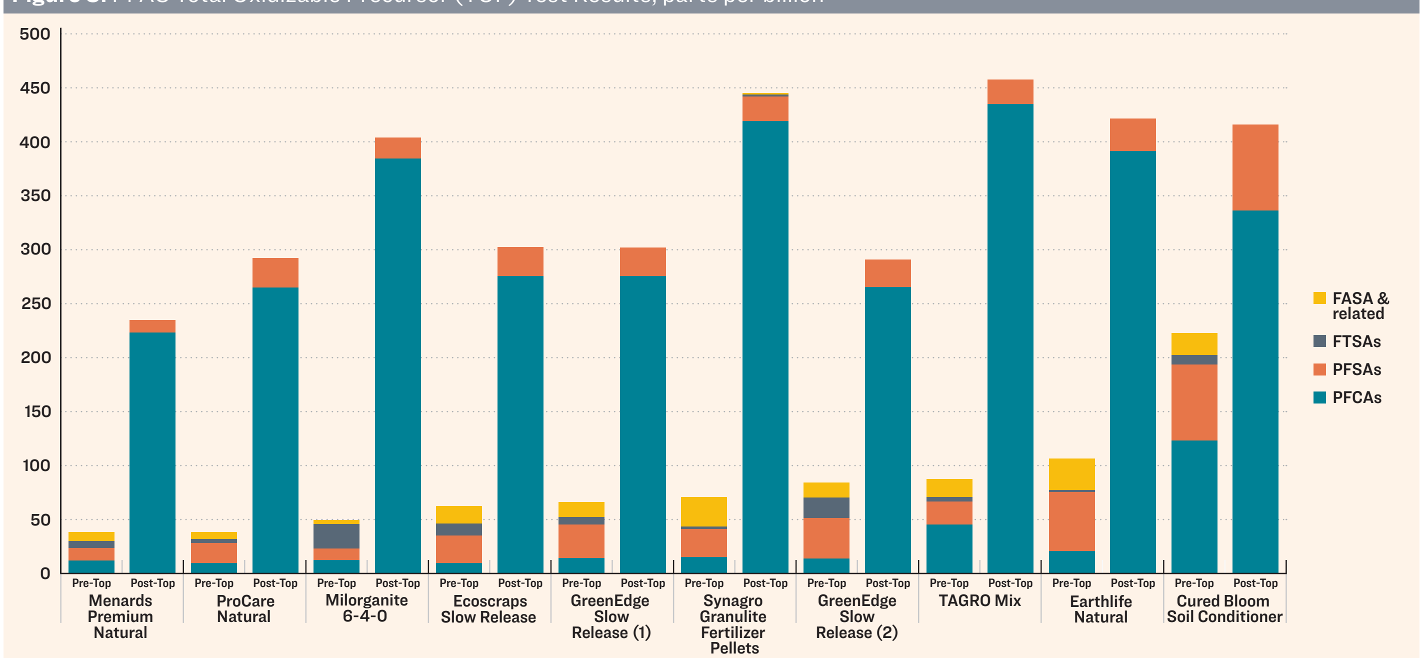


PFAS PRECURSORS

The EPA's preliminary field studies demonstrate that a class of complex but poorly studied PFAS chemicals act as "precursors" to the stable perfluoroalkyl acids and sulfonates measured in traditional studies. This means the precursors degrade over time to form the stable PFAS we measure using LC/MS/MS methods. To mimic this process in the laboratory, SGS subjected the fertilizer products to chemical oxidation in a test known as the "Total Oxidizable Precursor" or TOP Assay. TOP provides an indirect measurement of some PFAS chemicals by reducing complex PFAS chemicals to the perfluoroalkyl carboxylates and sulphonates measured by the LC/MS/MS assay. The TOP Assay done by SGS compared the sum of 33 specific PFAS measured before and after oxidation, which reflects the transformation of precursors into measurable PFAS. In the fertilizer products, oxidation resulted in measured PFAS levels 2 to 8 times higher than in the original samples. Total precursors ranged from 193 to 374 ppb in the products.

This finding highlights the fact that typical methods that measure just 20 to30 specific PFAS in water, biosolids, or other media underestimate the amount of these chemicals that will form over time in the environment. The TOP Assay is an important tool to quantify the unknown precursors that will be transformed to the PFAS of greatest health concern.

Figure 3: PFAS Total Oxidizable Precursor (TOP) Test Results, parts per billion



TOTAL FLUORINE

We also sent samples of the nine products to Galbraith Laboratories in Knoxville, TN, to test for other unknown fluorine-based chemicals in the fertilizer products. Instead of measuring specific PFAS compounds, Galbraith measured the fluorine content of samples using two tests to quantify both the "total fluorine" and "total inorganic fluorine" in each sample.

Inorganic fluorine, also called fluoride, comes nearly exclusively from natural sources. Inorganic fluorine made up only a tiny fraction of total fluorine measured in the biosolids. The rest comes from synthetic fluorochemicals. Although we don't know if these fluorochemicals are polymers like plastic, or mystery PFAS chemicals, they could pose a significant concern for people and the environment.

These are the first reported levels of total fluorine analyzed for biosolids in the US, and the results were remarkable. The 24 PFAS compounds measured in the biosolid products accounted for only a tiny portion of the total fluorochemicals present. In fact, the known PFAS made up less than 1 percent, and in most cases less than 0.1 percent, of the total mass of synthetic fluorinated chemicals in all the products.

Our findings are consistent with the only study we have been able to identify of fluorochemicals in sewage sludges, which was performed in Sweden (Eriksson 2015). Other chemists have found a similar dynamic when comparing known PFAS to total fluorine in products including treated papers and textiles (Robel 2017) and textile finishing agents (Mumtaz 2018).

Table 4: Total inorganic fluoride and total fluorine vs known PFAS in fertilizers

Product	Fluoride (ppb)	Total Fluorine (ppb)	Sum of known PFAS (ppb)
Cured Bloom Soil Conditioner	<500	131,000	223
Earthlife Natural	500	184,000	106
EcoScraps Slow Release	<500	179,000	62
GreenEdge Slow Release (1)	900	321,000	84
GreenEdge Slow Release (2)	1000	319,000	66
Menards Premium Natural	<500	215,000	38
Milorganite 6-4-0	<500	180,000	49
Pro Care Natural	<500	206,000	38
Synagro Granulite Fertilizer Pellets	600	61,000	71
TAGRO Mix	<1.0	13,000	83

At this point, we don't have an easy way to determine the type of unknown fluorochemicals in these and other products. Likely some of the mass comes from fluorine-based polymers (like PTFE, a fluorine-based plastic used for Teflon coatings and plastics like the Apple Watch Sport Band). The potential hazards posed by unknown PFAS in the products is not well studied. PFAS chemicals are used to make these fluorine-based plastics or polymers and are released during manufacturing, product use, or disposal, when wastes break down in the environment. Their presence indicates a potential long-term impact to the environment, including to water and food crops. More information is urgently needed, especially since the EPA's PFAS inventory contains more than 9,000 PFAS chemicals. The EPA and other government bodies do not track their production, use, or potential impacts to people and the environment.

TEST RESULTS ALIGN WITH PRIOR STUDIES

The samples we tested represent a snapshot of biosolids products. We would expect the concentrations of PFAS and other fluorochemicals to vary from product bag to product bag, depending on the contents that entered the wastewater treatment plant on different days and seasons.

While we studied a small number of commercial products, the measurements align with observations in academic studies of PFAS in biosolids, including one that used the TOP Assay to measure precursors (Lazcano 2020), and a study of total "extractable" organic fluorine (indicating fluorochemicals that are likely PFAS) in biosolids in Sweden (Eriksson 2015). The PFAS levels we measured in biosolid products are higher than levels measured in commercial composts not made of biosolids in two studies (Lazcano 2020, Choi 2019).

Table 5: PFAS and fluorine in biosolids, fertilizers, compost and other products, parts per billion

Material (Number of samples)	PFAS measured by LC/MS/MS	PFAS measured after oxidation with TOP Assay	Total fluorine	Reference
Biosolids-based home fertilizers (N=9)	38–233	234 to 445	13,000–321,000	This study
Swedish sewage sludges (N=4)	95–170	Not measured	600–2,700 ppb (extractable organic F)	Eriksson 2015
Biosolids-based home fertilizers (N=11)	9-199	50–320	Not measured	Lazcano 2020
Compost made from yard and food wastes (N=1)	~22	62	Not measured	Lazcano 2020
Non-biosolids commercial compost (N=6)	0.1–1.1	Not measured	Not measured	Lazcano 2020
Commercial compost not made from biosolids (N=7)	29–76	~30–110	Not measured	Choi 2019
Compost with no food containers and home compost (N=3)	2.4-7.6	<10	Not measured	Choi 2019

THE FATE OF PFAS IN WASTEWATER SYSTEMS, AGRICULTURAL SYSTEMS, AND THE FOOD SUPPLY

PFAS chemicals pose huge, complicated, and expensive management challenges at the end of their useful life. While this investigation highlights the challenges posed by wastewater disposal and biosolids reuse, it is important to note upfront that it is far simpler, less expensive, and more effective to stop using the chemicals in most consumer and industrial uses, rather than attempt to contain and manage wastes. The global lack of oversight means continued dispersed PFAS pollution as well as expensive and incomplete disposal and remediation. Government regulations on PFAS are being enacted decades too late; contamination of our bodies, our food supply, and our environment is already widespread.

PFAS ARE DISCHARGED FROM DOZENS OF INDUSTRIES AND HUNDREDS OF EVERYDAY PRODUCTS

Industries known to discharge PFAS into the wastewater system include metal plating, chemical manufacturing, plastics, paper and textile mills, printing, petroleum extraction, mining, paint manufacturing, and industrial laundries. Industries using PFAS-based fire-fighting foams include airfields, military bases, petroleum refining and storage, and firefighting, and fire-training sites.

PFAS are currently found in hundreds of consumer goods, meaning seemingly innocuous products used in homes and small businesses, like car washes, carpets, floor waxes, and non-stick pans, shed small amounts of PFAS into the wastewater system. While immediate actions can reduce the scope of the crisis and clean up pollution hot spots, we will be left with a burden of PFAS chemicals in homes and commercial buildings and a legacy of polluted soils, streams, and groundwaters that contribute PFAS to the wastewater system for decades to come.

BIOSOLIDS APPLICATIONS INTRODUCE PFAS IN THE FOOD SUPPLY

The EPA is investigating the threat posed by PFAS in biosolids, and data coming from these studies confirm that land application spreads PFAS through the food chain. The FDA has identified several other PFAS hot spots where water contaminated by biosolids application or industrial sources tainted dairy products or produce (FDA undated). Maine farmer Fred Stone's milk had similar levels of PFOS and PFOA, concentrations that exceeded the state's limit for milk , which is 210 parts per trillion (ppt). Tozier Dairy Farm in Fairfield, Maine, had similar problems, with concentrations of PFAS ranging from 12,000 to 32,000 ppt found in its milk. The remainder of milk sampled in Maine had undetectable levels of PFAS (less than 50 ppt).

> *"Data suggest that edible crops grown in soil conventionally amended with municipal biosolids may contain PFAS, and further studies are needed to characterize mechanisms of uptake from various soils and crops."* —(USEPA 2020)

In general, newer generation—or "shorter-chain"—PFAS are more mobile in water, less removed by water filtration systems, and more readily taken up by plants than longer-chain compounds. One study of vegetables that included celery, peas, radishes, and tomatoes grown in PFAS-tainted water found that different PFAS chemicals accumulated in different parts of the plant (Blaine 2014).

The FDA measured PFAS levels in the 20 to 200 ppt range for leafy greens grown near The Chemours Company's Fayetteville site in North Carolina. PFAS may have come from contaminated soils, water, or air deposition. A follow-up study in the area measured high levels of one chemical, PFDA, in tomatoes and potatoes (Li 2021).

While PFAS studies have focused on highly contaminated biosolids, there are reasons to be concerned about the concentrations detected in biosolids with lesser levels of contamination. While concentrations of PFAS measured in commercially sold vegetables and dairy products are generally much lower than those from polluted sites, even small amounts still pose a health concern, as they add to the overall burden of exposure to multiple sources.

In general, people are estimated to ingest far more PFAS from their diets than from their drinking water, unless their water has high levels of PFAS. We may also inhale the chemicals when they volatilize from water, absorb them through our skin, or ingest chemicals sticking to our hands or other items. Since the chemicals do not break down in the environment, levels in farm fields will slowly increase every time more biosolids are applied to a piece of land. The fertilizer products we tested are marketed for multiple applications per year to home gardens. The EPA reports that some farm fields have had biosolids continuously applied for up to 20 years.

CHALLENGES OF BIOSOLIDS DISPOSAL

When it comes to highly persistent chemicals in biosolids, such as PFAS, each of the current alternative disposal options will not fully destroy or contain chemical contaminants in sewage wastes.

The biosolids industry and governments have aggressively marketed land application of biosolids as a cost-effective disposal solution for the massive quantities of human waste, residential, and industrial wastewater produced daily. While the tenets of a circular economy and recycling would support the reuse of nutrients from human sewage, our homes and industries

OTHER CONTAMINANTS IN BIOSOLIDS

The Clean Water Act allows the EPA to set contaminant limits for biosolids, but the Investigator General has raised concerns about the agency's lack of action on more than 300 chemical pollutants detected in biosolids (EPA 2018). These include scores of other industrial chemicals and pharmaceutical drugs that persist during wastewater treatment and are measured in sewage sludge or remaining wastewater. However, not all of these chemicals are as mobile in the environment, or as potent in harming people and wildlife, as PFAS chemicals.

An EPA report from 2021 reviewed data from 2018 to 19 and found many different classes of contaminants in biosolids. These include dioxins and furans, PCBs, flame retardants, volatile hydrocarbons, pharmaceutical drugs, PAHs, pesticides, antimicrobial ingredients (parabens), heavy metals, and fragrance additives (EPA 2021a).

These chemicals persist through the wastewater treatment process and concentrate in the semi-solid materials and wastewater that are discharged back into rivers and lakes. Wastewater managers and regulators should account for the cumulative burden posed by the complex mixture of chemicals found in biosolids products, especially those applied to food crops.

are the source of hundreds of persistent chemicals that are spread back to land via this practice (USEPA 2021b).

There have been several high-profile cases where land application of biosolids spread harmful amounts of PFAS into the environment. In Alabama, 3M and Daikin released large amounts of PFAS wastes from Decatur facilities into the local wastewater system. The land application of highly contaminated biosolids over more than a decade contaminated 5,000 acres of farmland, community water supplies, and agricultural products. In Maine, two high-profile dairy producers have discovered that biosolids applications have permanently contaminated their pasture lands, rendered their dairy products unsellable, and impacted dozens of nearby wells.

The land application of sewage wastes is also a hidden but pervasive social equity issue. As with all issues of waste disposal, facilities tend to be located near marginalized communities that have less power to influence local land use decisions. People living near biosolids composting facilities experience odors, increased vehicle traffic, and the threat of local water contamination (Lowman 2013). Neighbors of farmers applying biosolids as fertilizer complain of illness, contaminated water supplies, and loss of property values and quality of life.

Unfortunately, other disposal options are also problematic. They can be expensive as well as energy- and space-intensive. When it comes to highly persistent and mobile chemicals like PFAS, each of these disposal options will not fully destroy or contain the chemical wastes. The EPA estimates that about 16 percent of the nation's biosolids are incinerated for energy recovery or waste reduction (USEPA undated). However, incineration is energy-intensive and may not destroy PFAS, which are highly heat-resistant. Instead, incineration can spew a range of harmful breakdown products into the air, ultimately contaminating land and water far from the incineration site (Stoiber 2020). Waste ash from incinerators still needs to be disposed of in landfills and managed in perpetuity.

According to the EPA, 22 percent of biosolids are disposed of in landfills (USEPA undated). Landfilling biosolids is space-intensive and expensive due to the volume produced. PFAS and other mobile chemicals leach out of the liquid wastes produced by landfills and need to be managed—either by reinjecting them back into the landfill or by filtering liquids to concentrate the chemicals onto a polymer or carbon filter material, which itself must be contained for centuries. Some landfills send liquid wastes to local wastewater treatment plants, which has the effect of sending PFAS and persistent chemicals back into circulation in land and waterways. Furthermore, even lined landfills will eventually leak, and PFAS and other persistent pollutants are commonly measured in the groundwater near landfills.

New technologies—supported by financial incentives from the government to invent them—are desperately needed to address PFAS and other persistent chemicals in wastewater. In the interim, we must act with haste to prevent controllable sources of PFAS discharge and ensure polluting industries—and not the public—pay for the cost of disposal.

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APPENDICES

A. DEFINITIONS

PFAS: Per- and polyfluoroalkyl substances are synthetic chemicals that contain a chain of carbon atoms where the hydrogens have been replaced with fluorine atoms. Technically, state laws define PFAS as a class of fluorinated organic chemicals containing at least one fully fluorinated carbon atom. The portion of the chemical containing the carbon-fluorine bond is highly stable and will not break down in the environment.

Biosolids: During wastewater treatment the liquids are separated from the solids. Those solids are then treated physically and chemically to produce a semi-solid product. The terms "biosolids" and "sewage sludge" are often used interchangeably. Technically, biosolids are the term for sewage sludges once they have undergone treatment and meet EPA criteria for land application. The fertilizer products we studied are identified as "biosolids derived" on product labels.

Sewage Sludge: The terms "biosolids" and "sewage sludge" are often used interchangeably. Technically, sludge is any solid, semisolid, or liquid residue removed during the treatment of municipal wastewater or domestic sewage. Sewage sludge includes solids removed during primary, secondary, or advanced wastewater treatment, scum, septage, portable toilet pumpings, type III marine sanitation device pumpings (33 CFR Part 159), and sewage sludge products. Sewage sludge does not include grit or screenings, or ash generated during the incineration of sewage sludge. [40 CFR 122.2].

Residuals: commonly referred to as "mill sludge," residuals are a product of the wastewater treatment process of waste paper recycling, wood pulping, and papermaking. The term is sometimes used as a euphemism for biosolids/sewage sludge in agricultural or garden products.

Total Fluorine: A measure of the human-made and naturally occurring chemicals containing fluorine. Our test results allow us to subtract the total inorganic fluorine content of biosolids and estimate the amount of carbon-based chemicals where fluorine replaces hydrogen atoms. PFAS are a family of organic fluorine chemicals.

Inorganic Fluoride: The inorganic anion of fluorine (F^-) is called fluoride. The fluoride ion can form various inorganic compounds such as calcium fluoride, sodium fluoride, aluminum fluoride, potassium fluoride, and magnesium fluoride. Fluoride ions occur on earth in several minerals, particularly fluorite. Sodium fluoride was the first chemical used for water fluoridation. Salts of fluoride are widely used as important chemical reagents and industrial chemicals, mainly used in the production of hydrogen fluoride for fluorocarbons.

PFOA: Perfluorooctanoate, an eight-chain PFAS chemical largely phased out of production globally. For decades, PFOA was a key processing aid used in fluoropolymer production.

PFOS: Perfluorooctane sulfonate, an eight-chain PFAS chemical largely phased out of production globally. PFOS is a surfactant that has been used in firefighting foams and other applications.

Short-chain PFAS: Short-chain refers to:

- Perfluoroalkyl carboxylic acids (PFCAs) with seven or fewer carbons (six or fewer carbons are perfluorinated)
- Perfluoroalkyl sulfonic acids (PFSAs) with five or fewer carbons (five or fewer carbons are perfluorinated) See ITRC for more details.

Long-chain PFAS: Long-chain refers to:

- PFCAs with eight or more carbons (seven or more carbons are perfluorinated)
- PFSAs with six or more carbons (six or more carbons are perfluorinated).
- See the Interstate Technology Regulatory Council (ITRC) for more details.

PFAAs: Perfluoroalkyl Acids, which include PFOA and PFOS, are essentially non-degradable under normal environmental conditions. Biotic and abiotic degradation of many polyfluoroalkyl substances, known as "PFAS precursors," may result in the formation of PFAAs. As a result, PFAAs are sometimes referred to as "terminal PFAS" or "terminal degradation products," meaning no further degradation products will form from them under environmental conditions. See ITRC for more details.

FASA: Perfluoroalkane Sulfonamides, such as perfluorooctane sulfonamide (FOSA), are used as raw material in the process to make perfluoroalkyl sulfonamide substances that are used for surfactants and surface treatments (known as ECF). FASAs can degrade to form PFAAs such as PFOS. Related to FASAs are FASAAs, perfluoroalkane sulfonamido acetic acids, and FASEs, perfluoroalkane sulfonamido ethanols.

FTSA: Fluorotelomer sulfonic acids. The n:2 fluorotelomer sulfonic acids have been detected in environmental matrices at sites where aqueous film-forming foam has been used, in wastewater treatment plant effluents, and landfill leachate. FTSAs are precursor compounds and can undergo aerobic biotransformation to form PFCAs.

PFSA: Perfluoroalkane sulfonic acids, or perfluoroalkyl sulfonates, are also used commercially and can be formed as terminal degradation products of select precursor polyfluoroalkyl substances, such as FASEs. PFSAs have been used in firefighting foam and in mist suppressants for metal plating. An example PFSA is PFOS.

PFCA: Perfluoroalkyl carboxylic acids, or perfluoroalkyl carboxylates, are used commercially and can be formed as terminal degradation products of select precursor polyfluoroalkyl substances, such as FTOHs. An example PFCA is PFOA.

PFAS ether or Ether-PFAS: Per- or Poly-fluoroalkylether compounds. Some PFAS ethers have been developed for use as replacements for other PFAS that are phased out of production and use. Chemours' replacement process aid for PFOA, called GenX, is a PFAS ether. Another such replacement is called ADONA. Others are more recently detected in the environment and near industrial sites, such as perfluoro-2-methoxyacetic acid (PFMOAA) and chlorinated perfluorinated polyether compounds (PFPECAs).

B. TEST RESULTS

Appendix Table 1: PFAS chemicals analyzed using SGS LC/MS/MS test methods for PFAS in biosolids

Perfluoroalkyl sulfonates (PFSAs)	Perfluoroalkyl carboxylates (PFCAs)	PFAS ethers	Perfluoroalkane sulfonamides (FASAs & Related)	Fluorotelomer sulfonic acids (FTSAs)
PFBS	PFBA	HPO-DA (GenX)	PFOSA	4:2 FTS
PFPeS	PFPeA	ADONA	N-MeFOSA	6:2 FTS
PFHxS	PFHxA	9Cl-PF3ONS*	N-EtFOSA	8:2 FTS
PFHpS	PFHpA	11Cl-PF3OUdS*	MeFOSAA	
PFOS	PFOA	*components of F-53B	EtFOSAA	
PFNS	PFNA		N-MeFOSE	
PFDS	PFDA		N-EtFOSE	
PFDoS	PFUnA		Red = detected in at least one product (multiple products in most cases). Black = not detected	
	PFDoA			
	PFTrDA			
	PFTeDA			

Appendix Table 2: PFAS measurements for 5 common classes of PFAS, parts per billion

		PFCAs	PFSAs	FTSAs	FASAs & related	Ether- PFAS	Total LC/MS/MS
Menards Premium Natural	Pre-TOP	12	12	6	8	ND	38
	Post-TOP	223	12	0	0	not tested	235
ProCare Natural	Pre-TOP	9	19	4	6	ND	38
	Post-TOP	265	28	0	0	not tested	292
Milorganite 6-4-0	Pre-TOP	12	11	22	4	ND	49
	Post-TOP	384	19	0	0	not tested	404
Ecoscraps Slow Release	Pre-TOP	9	26	11	16	ND	62
	Post-TOP	275	27	0	0	not tested	302
GreenEdge Slow Release (1)	Pre-TOP	14	31	7	14	ND	66
	Post-TOP	276	26	0	0	not tested	302
Synagro Granulite Fertilizer Pellets	Pre-TOP	15	26	2	27	ND	71
	Post-TOP	419	23	2	1	not tested	445
GreenEdge Slow Release (2)	Pre-TOP	14	37	19	14	ND	84
	Post-TOP	265	25	0	0	not tested	291
TAGRO Mix	Pre-TOP	45	21	5	16	ND	87
	Post-TOP	435	23	0	0	not tested	457
Earthlife Natural	Pre-TOP	21	55	2	29	ND	106
	Post-TOP	391	30	0	0	not tested	422
Cured Bloom Soil Conditioner	Pre-TOP	123	71	9	20	ND	223
	Post-TOP	336	80	0	0	not tested	416
Average Per Class Pre-TOP		27	31	9	16	ND	83
Average Per Class Post-TOP		327	29	0	0	not tested	357

C. LAB DETECTION LIMITS, HOW WE HANDLED NON-DETECTS, AND SEMI-QUANTITATIVE FINDING

Reporting limits for PFAS using LC/MS/MS ranged from as low as 0.208 ppb to 0.532 ppb for the individual PFAS compounds analyzed for the fertilizer products in our study. Limits for the two soil conditioners were a bit higher, due to the product composition. Cured Bloom soil conditioner reporting limits range from 0.264 ppb to 6.76 ppb and TAGRO Mix Conditioner ranged from 0.512 ppb to 12.8 ppb. Note that the two PFAS compounds with the highest reporting limit (12.8 ppb) were 5:3 FTCA and 7:3 FTCA. These chemicals were not detected in any of the products we sampled. Soil conditioners have the higher reporting limit because they are much less dense and lighter as a compost-like product (versus fertilizer pellets), and the sample weight was therefore much lighter.

We tally all PFAS detected above the reporting limit, including some detections flagged by the laboratory as having potential uncertainty about the exact value. These play a very minor role in the total amount of PFAS measured in fertilizer samples. We considered non-detects to be zero.

D. ADDITIONAL INFORMATION ABOUT NINE PRODUCTS TESTED

Wastewater treatment processes subject biosolids to treatment to reduce the pathogens, odors, and vector attraction characteristics. In the case of our project, all products are derived from Class A or “Exceptional Quality” biosolids.

We reviewed each product to determine the supplier and, where available, information on the wastewater treatment plant or biosolids source. In the case of three products— ProCare, Menards, and EcoScraps—we could not determine the origins of the biosolids or the wastewater treatment plant where they were processed. With regard to the Earthlife product, we know that it is pelletized by NEFCO in Quincy, Massachusetts, but we do not know the origins of the biosolids. The five remaining product processes were available and are described as fairly standard wastewater treatment systems with few listed physical variations.

E. APPLICATION RATES FOR HOME FERTILIZERS AND SOIL CONDITIONERS

Seven of the products in this study are soil fertilizers and two are soil conditioners. In general, fertilizers improve the supply of nutrients in the soil, directly affecting plant growth. Fertilizers improve a soil's physical condition (e.g., soil structure, water infiltration), indirectly affecting plant growth. In many cases, soil conditioners do both, improve nutrients and physical soil conditions. The soil conditioners tested in this project had a consistency similar to a compost or soil, while the fertilizers were in pellet form.

Soil conditioners and fertilizers have different recommended application rates and protocols for reapplication, which impacts the amount of PFAS and other persistent chemicals that end up in the soil. Recommended application rates are typically based on the nitrogen content.

Biosolid-based soil conditioners are normally blended with other materials and are often added to soil at much larger amounts than those marketed as fertilizers. Therefore, products with lower PFAS levels that also have low nitrogen levels could contribute relatively higher PFAS loads to a garden compared to fertilizer products with higher nitrogen levels.

The products we tested had a variety of application instructions. Some recommendations were based on soil current nitrogen content, and others simply suggested applying the product a certain number of times per year.

The impact to the environment from increased application rate and repeat applications of the product as it relates to the burden of PFAS is obvious: The more product that is applied, more frequently, the greater the load on the environment.

In terms of nitrogen percentages, Cured Bloomsoil conditioner was a lot more like the fertilizers in our study. The other soil conditioner, TAGRO Mix, was a blend of 50 percent biosolids, 25 percent sawdust, and 25 percent sand. It had a much lower nitrogen content and therefore more would need to be applied to achieve soil nutrient levels. However, the label application rate was only twice a year.

Appendix Table 3: Application Rates

Product	Total Nitrogen N_i (%)	Application Rate (Kg Product/ Kg Soil)	Application Frequency
ProCare All Natural	4	0.33%	"During growing season" 2 X per year." (Because of Phosphate)
Ecoscraps Slow Release	5	0.26%	4 X per yr
Milorganite 6-4-0*	6	0.22%	4 X per yr
Cured Bloom Soil Conditioner	3.9	0.34%	If C/N ratio > 15 additional N should be applied.
Menards Premium Natural	4	0.33%	Anytime during the growing season; may be reapplied every 8 weeks.
GreenEdge Slow Release	6	0.22%	4 X / yr warm climate 3 X / yr cold climate
Earthlife Natural	5.4	0.24%	3 X per yr
Synagro Granulite Fertilizer Pellets	5	0.26%	3 X per yr
TAGRO Mix	1.4	0.94%	2 X per year, spring and fall

F. DETAILED SAMPLING PROCEDURE AND PLANS

All samples were collected following the protocols outlined in the Michigan Department of Environment, Great Lakes and Energy *Biosolids and Sludge PFAS Sampling Guidance, Revised 10/17/2018*. Key components of the protocol include:

- The field technician did not wear clothing treated with fabric softener, clothing or boots treated with synthetic water- and/or stain-resistant materials, clothing made with or washed with water, dirt, and/or stain-resistant chemicals, or clothing chemically treated for insect- and/or UV protection. All clothing worn was cotton and laundered at least six times without use of fabric softener.
- The field technician wore powderless nitrile gloves for the entirety of the sampling event and changed gloves between each product, as well as any time the gloves came in contact with non-sampling materials.
- No sampling equipment was used in the process of collecting samples during this sample event in order to

reduce the potential of introducing contaminants. All samples were collected directly by using sterile bottles provided by the laboratories.

- All retain samples were collected in Ziploc baggies and were double-bagged.

Products were stored prior to the sample event in the field technician's garage in a room separate from parked vehicles until all chosen products were obtained. Product bags were kept closed, as packaged by the supplier, until the day of the sample event. Photos were taken of each product bag for documentation, and a video was taken of two complete sample processes to illustrate collection methodology, including the entire chain-of-custody and field documentation procedure.

The following is a summary of several elements of the project quality assurance plan.

- One duplicate sample was included to illustrate reproducibility at each laboratory.
- A temperature blank was sent by the lab in each cooler to assure the temperature of arrival of samples as received by each laboratory.*
- All samples collected were in lab-provided 250 milliliter amber glass bottles and stored separate from the other samples collected for each product, to minimize risk of cross-contamination inside Ziploc baggies and by black foam inserts with bubble wrap in between each layer.
- A retain sample was collected from each product and double bagged in clean Ziploc baggies, to be retained at the field technician's home in the potential event of future or for alternate method sampling event(s). The retain samples were properly labeled and maintained in a plastic storage container in a secure, dry location.
- All sample bottles, chain-of-custody, and return shipment packaging was provided by SGS AXYS Laboratory and lab-specific directions, where applicable, were followed.
- All samples collected were stored in a blue-ice-packed cooler, taped closed and shipped via FedEx overnight.* The samples were submitted to SGS AXYS Analytical Services Ltd., and Galbraith Laboratories, Inc. and analyzed for PFAS using the following methods.
    - SGS AXYS
        - PFAS by SGS AXYS Method MLA-110 LC-MSMS - Biosolids 430 PFAS MLA-110
        - TOP Assay SGS AXYS Method MLA-110 LC-MSMS (Conversion/Oxidation & Post)
    - Galbraith
        - TOTAL F - F (E9-3). http://galbraith.com/wp-content/uploads/2015/08/E9-3-Total-Fluorine-by-Oxygen-Flask-Combustion-ISE-GLI-Method-Summary.pdf
        - INORGANIC F - A09 (E9-1) http://galbraith.com/wp-content/uploads/2015/08/E9-1-Fluoride-Ion-by-ISE-GLI-Method-Summary.pdf

** It is important to note that both coolers experienced delays in their delivery to the respective labs for different reasons. Received temperatures were duly noted by each lab and requested analyses completed.*

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