Evidence of compost contamination with per- and polyfluoroalkyl substances (PFAS) from "compostable" food serviceware

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LETTER

Evidence of compost contamination with per- and polyfluoroalkyl substances (PFAS) from "compostable" food serviceware

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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) have been used to waterproof and greaseproof food serviceware for decades. Health concerns about these compounds have drawn attention to the potential for contamination of the food system. Finished compost (n = 3) made from manure and food serviceware labeled "compostable" generated at a large fair was found to contain 12 or 13 of the 28 PFAS compounds sampled for, in concentrations ranging from 1.1 to $183 \,\mu$ g/kg (Σ_{28} PFAS range = $209-455 \,\mu$ g/kg). Of note, perfluorooctanoic acid, a known carcinogen, was found at concentrations between 47.2 and $55.5 \,\mu$ g/kg. In contrast, fresh manure contained only perfluoroctanesulfonic acid at $3.7 \,\mu$ g/kg, and separated food waste from the fair composted with grass clippings and livestock bedding had no detectable PFAS in 2022, and Σ_{28} PFAS = $9.6 \,\mu$ g/kg in 2019. Including compostable serviceware in compost likely contaminates the finished compost and threatens surrounding groundwater and surface waters, in addition to increasing potential crop uptake.

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I. INTRODUCTION

A. Background

Compostable food serviceware options commonly include biodegradable plastics, paper, cardboard-based, or molded fiber materials. As consumer concern over the environmental impacts of plastic has grown, many single-use paper, cardboard-based, and molded fiber serviceware options have increasingly entered the market with the goal of improving sustainability. These single-use disposable food serviceware materials lack the impermeability of plastics and have been treated with per- and polyfluoroalkyl substances (PFAS) for decades because of their water- and oil-resistant properties. This includes coatings on serviceware designed to be compostable. Specific formulations of serviceware coatings have changed over time, with increased recognition of human health impacts from older chemistries, exemplified by industry shifts away from perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA). However, there is growing concern that currently-used chemistries likely also present human health risks.¹ PFAS compounds are themselves not readily decomposed during composting due to the strength of carbon-fluorine bonds.² Thus, their persistence in finished compost is of concern.³

The changing landscape of product development and regulation and guidance around the presence of specific PFAS or organofluorine compounds, more generally, has generated confusion among consumers and producers alike. Two commonly used methods of measuring these compounds that produce very different results are the liquid chromatography tandem mass spectrometry (LC-MS/MS) methods and the total or extractable organofluorine (TOF) tests. The former can detect the presence of specific compounds at very low levels, and the latter, while less sensitive, is less expensive, nonspecific, and can detect many more fluorinated organic compounds that would be missed using LC-MS/MS.⁴ Both approaches have been used in the development of regulations and guidelines.

Composts are a possible source of PFAS contamination into the environment and agricultural food pathways.^{3,5} Although no PFAS thresholds have, to our knowledge, been set for composts, the northeastern U.S. state of Maine restricted land application of biosolids for "beneficial use" to materials with less than 1900, 5.2, and 2.5 µg/kg (ppb) of perfluorobutanesulfonic acid (PFBS), PFOS, and PFOA, respectively. This restriction was put into effect prior to a complete ban on land application of biosolids passed by the State Legislature in 2022 due to PFAS contamination concerns.⁶ Maine has also released agronomic guidance for soil PFOS, suggesting that levels as low as $6.4 \mu g/kg$ may warrant investigation due to the potential for forage uptake and concomitant contamination of meat or dairy.⁷ Additionally, Maine has crafted remedial action guidelines for contaminated sites with soil levels of 7100, 3.6, and 1.7 µg/kg (ppb) of PFBS, PFOS, and PFOA, respectively, as these levels may allow leaching to contaminate groundwater above residential groundwater guidelines.⁸ Though Maine has implemented some of the first such thresholds in the country, updated federal human intake guidance suggests that levels are likely to be instituted at even lower concentrations in the future.^{9,}

At the federal level, Denmark is the only country that we are aware of to prohibit intentional use of PFAS (including short- and long-chain compounds) in compostable food serviceware. The Danish law, which took effect in 2020, allows for continued PFAS use in food serviceware products if a functional barrier would prevent migration of PFAS into food, however.¹¹ The Danish ban sets an intentionally-added PFAS indicator value of $20\,000\,\mu g$ TOF per kg of material (20 ppm). This is $5 \times$ lower than the $100\,000\,\mu$ g/kg TOF (100 ppm) certification maximums set by the Biodegradable Products Institute and the Compost Manufacturing Alliance (BPI, and CMA, respectively). The BPI and CMA maximums took effect in 2020^{12,13} and are the most common guidance referenced in North America for PFAS in compostable food serviceware. While the TOF method used in these guidelines is expected to produce higher results due to the wider breadth of chemicals detected, it is notable that the levels are up to three orders of magnitude higher than Maine's soil remedial action guidelines for PFOS and PFOA.

B. PFAS in compostable serviceware and finished composts

While concern for PFAS migration from serviceware to food has been a priority of recent research, published reports detailing the impact of compostable serviceware on finished compost are sparse. However, several studies are suggestive of the role which serviceware may have in increasing PFAS contamination levels in finished compost.

Yuan *et al.*¹⁴ reported a compostable serviceware product with a 6:2 fluorotelomer alcohol (6:2 FTOH) concentration of $499 \,\mu g/\text{kg}$. Timshina *et al.*¹⁵ reported total PFAS concentrations in paper- and plant-based straws as high as 29 ng/straw, with perfluorobutanoic acid (PFBA) and PFOA being the most frequently detected. Strakova *et al.*¹⁶ found compostable serviceware which exhibited oil beading, ranging from 560 to 1200 mg TOF/kg, with identified PFAS primarily dominated by 6:2 FTOH, ranging in concentration from 92 to $4766 \,\mu g/kg$, with a median value of $580 \,\mu g/kg$. Note that the concentration of the 6:2 FTOH, the highest individual PFAS compound reported, is 1000 times less than the TOF value, signaling that there are many additional fluorinated organics that are not being quantified by the LC-MS/MS method. This conclusion is reinforced by the recent study of 42 examples of Canadian fast food serviceware and packaging by Schwartz-Narbonne et al.,¹⁷ which utilized a suite of analytical techniques, finding the highest level of total F $(1-1.3 \text{ g F/m}^2)$ in four samples of molded fiber bowls that were marketed as compostable. Follow-up targeted analysis of 55 PFAS compounds was only able to identify 0.02%-0.06% of the total F in compostable bowls. Further analysis after hydrolysis improved comparison of the F mass balance of the bowls to 2.4%-5.6% of the total F, by freeing 6:2 FTOH, 6:2 fluorotelomer methacrylate (6:2 FTMAc), and 6:2 fluorotelomer acrylate (6:2 FTAc) from larger compounds, likely side-chain fluorinated polymers. Though targeted analysis had found the greatest PFAS concentrations in compostable bowls to be either 6:2 FTOH (294-885 µg/kg) or 6:2 FTMAc (430- $681 \mu g/kg$), non-targeted analysis identified 6:2 fluorotelomer unsaturated carboxylic acid (6:2 FTUCA) as the most abundant PFAS, with two n:3 fluorotelomer carboxylic acids also identified in two of the samples.

Choi *et al.*¹⁸ found perfluoroalkyl acid (PFAA) concentrations in composts that included food packaging to be approximately an order of magnitude greater than composts that did not contain food packaging (ranging from 28.7–75.9 vs 2.38–7.6 μ g/kg, respectively). Though research in this area is limited, this initial evidence points to a strong likelihood that compostable serviceware used as compost feedstock will ultimately contaminate the finished compost product.

C. Opportunistic research opportunity

The Maine Organic Farmers and Gardeners Association (MOFGA) has held an annual agricultural fair emphasizing sustainable rural living since 1977, which attracts tens of thousands of attendees over three days. Striving for zero waste, the fair organizers have composted food waste starting in the beginning years of the event, eventually including "compostable" single-use food serviceware-the provision of which is a requirement for participating food vendors. Dissatisfactory compostability of food serviceware led event organizers to begin a policy of postdisposal separation of serviceware and food wastes for separated composting. As PFAS contamination of agricultural land became a topic of great public concern in Maine in 2022, MOFGA decided to test its most recent compost (from 2019). Surprised by the results, follow-up testing was performed to confirm initial findings as well as to further elucidate the environmental and agricultural contamination pathways present in this example. Here, we describe the results, with the hope that they will serve as a starting point for future studies. Though not designed for robust statistical analysis, the results provide an opportunity to better understand the PFAS loading potential to compost from serviceware.

II. METHODS

A. History and preparation of the compost

Event food wastes have been composted following this agricultural fair, since its establishment in 1977. Compostable serviceware had been a requirement of food vendors prior to 2008; however, that requirement was reinforced that year with a list provided to vendors of acceptable companies that produced compostable serviceware. In 2014, food vendors received a prescriptive list of allowable compostable serviceware with listings of specific approved items instead of the previous list of approved companies. This was to prevent issues of "greenwashing" or other confusion among food vendors and to make compost volunteers' jobs easier, streamlining the sorting process by reducing the number of unique serviceware products.

Prior to 2015, food wastes were co-composted with serviceware (Table I; Fig. S1 in the supplementary material).³⁴ From 2015 on, compost volunteers sorted serviceware and food wastes for separated composting, with food wastes being co-composted with grass clippings from fairgrounds and bedding and manures from a diversity of display and demonstration livestock present at the event. Since 2017, food waste compost has been maintained in a windrow with a tractor-powered windrow turner.

Serviceware has been collected every year since 2015 in a 30 yard³ (22.9 m³) roll-off open top dumpster, and compacted by tractor weight. In 2015, food serviceware was composted with dairy bedding sand, removed from a manure lagoon after separation from manure slurry and rain-washed. The "dirty sand" had been added to the serviceware at an approximately 1:1 ratio by volume. After 2015, food serviceware was composted with dairy manure from a local farm, at a ratio of approximately 20 yard³ (15.3 m³) of dairy manure to 30 yard³ (22.9 m³) of food serviceware. Serviceware composts have at all times been maintained as a turned pile by a front-end loader.

All composts in all years were entirely rain-fed, with no additional water applied. Though 2018 serviceware compost had been land-applied prior to this investigation, it was not on a food producing area. Remaining serviceware compost materials from any year will not be land-applied, serviceware from the 2022 event was separated from food waste and, subsequently, disposed of as typical solid waste.

B. Sample collection and analysis

In August 2022, Northern Tilth, LLC (Belfast, Maine, 04915, USA) was contracted by MOFGA to collect compost samples for

TABLE I.	Compost	components.
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Compost	Years	Components
Combined food waste and serviceware	2014 and prior	Food waste, serviceware, grass clippings, display animal bedding, and manure
Food waste	2015– 2019, 2022	Food waste, grass clippings, display animal bedding, and manure Serviceware and sand from a
Serviceware Serviceware	2015 2016–2019	manure lagoon (1:1) Serviceware and dairy manure (3:2)

PFAS analysis. Northern Tilth has emerged as the primary private PFAS sampling entity in the state of Maine, with many years of experience collecting samples using carefully designed protocols. Initial sampling was performed on food waste-based and serviceware-based composts created from the 2019 fair event. Each compost sample was composited from ten subsamples to limit impacts of potential heterogeneity of the material.

Follow-up testing was performed in the same manner in December 2022 on the 2019 serviceware compost, a combined pile of serviceware composts created following the 2014–2017 events, manure from a local dairy that had supplied manure for cocomposting with serviceware in 2018 and 2019, food waste compost recently made from the 2022 event, and a locally sourced commercial compost used in a demonstration at the 2022 event (hereafter referred to as "demonstration compost"). Sampling choices were made to confirm initial findings and to further elucidate the environmental and agricultural contamination pathways present in this example.

All samples were sent to Alpha Analytical, Inc. (Mansfield, Massachusetts 02048, USA) for analysis of 28 PFAS compounds, listed in Table II, using the LC-MS/MS method modified from the EPA method 537.¹⁹

PFAS analyte	Abbreviation	
Perfluorobutanoic acid	PFBA	_
Perfluoropentanoic acid	PFPeA	7 July 2023 19:47:24
Perfluorobutanesulfonic acid	PFBS	ıly 2
1H,1H,2H,2H-Perfluorohexanesulfonic acid	4:2FTS	023
Perfluorohexanoic acid	PFHxA	19:4
Perfluoropentanesulfonic acid	PFPeS	17:2
Perfluoroheptanoic acid	PFHpA	4
Perfluorohexanesulfonic acid	PFHxS	
Perfluorooctanoic acid	PFOA	
1H,1H,2H,2H-Perfluorooctanesulfonic acid	6:2FTS	
Perfluoroheptanesulfonic acid	PFHpS	
Perfluorononanoic acid	PFNA	
Perfluorooctanesulfonic acid	PFOS	
Perfluorodecanoic acid	PFDA	
1H,1H,2H,2H-Perfluorodecanesulfonic acid	8:2FTS	
Perfluorononanesulfonic acid	PFNS	
N-Methyl perfluorooctanesulfonamidoacetic acid	NMeFOSAA	
Perfluoroundecanoic acid	PFUnA	
Perfluorodecanesulfonic acid	PFDS	
Perfluorooctanesulfonamide	FOSA	
N-Ethyl perfluorooctanesulfonamidoacetic acid	NEtFOSAA	
Perfluorododecanoic acid	PFDoA	
Perfluorotridecanoic acid	PFTrDA	
Perfluorotetradecanoic acid	PFTA	
2,3,3,3-Tetrafluoro-2-[1,1,2,2,3,3,3-ND	HFPO-DA	
heptafluoropropoxy]-propanoic acid		
4,8-Dioxa-3h-perfluorononanoic acid	ADONA	
Perfluorohexadecanoic acid	PFHxDA	
Perfluorooctadecanoic acid	PFODA	

III. RESULTS AND DISCUSSION

A. Results

PFAS analysis results (Fig. 1; Table SI in the supplementary material)³⁴ show that composts made with largely uncontaminated dairy manure and serviceware can contain 20–45 times the total PFAS concentration compared to compost made primarily with food waste. The most noticeable differences between serviceware composts from 2019 and older were the markedly higher levels of the shorter-chained PFBA, PFPeA, and PFHxA in the 2019 compost, and greater concentrations of longer-chained PFNA, per-fluorodecanoic acid (PFDA), and PFDoA, and also, possibly PFUnA, PFTrDA, and PFTA, in the more weathered composts from 2014–2017.

The most recent food waste compost (2022) and the commercially available compost purchased for a demonstration (demonstration compost) were entirely below the reporting limit for all PFAS analytes measured. The 2019 food waste compost did contain relatively small amounts of some of the same analytes found in the serviceware composts from that year.

B. Discussion

Choi *et al.*¹⁸ found composts including food packaging to contain approximately an order of magnitude greater concentrations of PFAAs than composts which did not include food packaging. Despite a significant dilution impact from co-composting with relatively uncontaminated manure, we report total PFAA

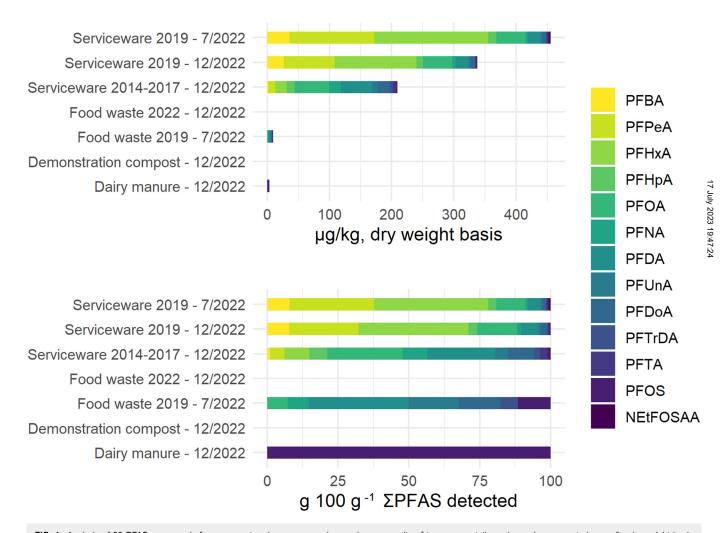


FIG. 1. Analysis of 28 PFAS compounds from compost and manure samples, each a composite of ten representative subsamples, presented on $\mu g/kg$ dry weight basis and as a proportion of Σ PFAS detected. PFPeA and PFOA values in 07/22 sampling of Serviceware 2019 compost should be considered estimated, as they were re-extracted on dilution, with the method required holding time exceeding in order to quantitate the results within the calibration range. Detailed results are available in Table SI in the supplementary material.³⁴

concentrations in serviceware inclusive composts $(209-453 \,\mu g/\text{kg})$ ranging from 2.5× to 5.8× the maximum concentration of total PFAAs (75 $\mu g/\text{kg}$) reported by Choi *et al.*,¹⁸ even when limiting the comparison to the 12 PFAAs detected across both reports. In both cases, PFOA levels found in serviceware inclusive composts were at least as great as the 2.5 $\mu g/\text{kg}$ regulatory threshold previously set for biosolids by the state of Maine, with our report showing that compost made with serviceware can exceed that threshold 18 times over. These levels are markedly higher than soil levels at sites with no known sources of contamination^{20,21} and may contribute PFAS loading to groundwater contamination via leaching⁶ and to surface waterways via sediment loading.²²

While the Σ_{28} PFAS results from 2019 serviceware inclusive compost are concerning, this likely represents a near-to-worst-case scenario. Most serviceware composting instances will likely not involve the separation of serviceware from the overall compostable waste stream. However, should serviceware compostability concerns have been addressed elsewhere in a similar manner by segregated composting, it is likely that the C:N ratio of nitrogenous material added to the recipe will play a large role in dilution, and subsequent PFAS concentration of the finished compost. Had these serviceware composts utilized a more nitrogen-rich co-composting material to produce the appropriate C:N ratio to begin the composting process, less co-composting material would have been needed, and the PFAS concentrations in the final composts could very well have been even greater.

Unlike biosolids, which are typically diluted through spreading and incorporation into soil, compost is also occasionally used as a standalone growing medium in raised beds or via the implementation of no-till deep compost mulch vegetable and fruit growing approaches.^{23–25} Using compost in this manner could exacerbate the potential for PFAS-contaminated material to contribute to crop plant uptake, localized surface runoff, or leaching to groundwater.

The similarity in the distribution of PFAAs found in greatest concentrations in serviceware composts reported here, with those reported in Choi *et al.*,¹⁸ (PFBA, PFPA, PFPA, PFHxA, PFOA, PFDA) further the evidence that serviceware is likely responsible for this source of PFAS contamination of composts. As 6:2 FTOH has been the PFAS compound most frequently reported in compostable serviceware, ^{14,16} its aerobic biotransformation may be responsible for the presence of PFBA, PFPeA, and PFHxA in serviceware compost reported here, as those perfluorocarboxylic acids (PFCAs) are known aerobic biostransformation products of 6:2 FTOH.² The related compound 8:2 FTOH is, likewise, known to biotransform to PFBA, PFPeA, and also PFOA, among other compounds.²

Other potential biotransformations are also of concern. First, though polyfluoroalkyl phosphate esters such as 6:2 diPAP and 8:2 diPAP were not sampled for here, these are precursors to PFPeA, PFHxA, and PFOA,²⁶ the three PFAS compounds found in the greatest concentrations in the serviceware composts (Fig. 1; Table SI in the supplementary material)³⁴. Research investigating PFAS in toilet paper²⁷ found 6:2 diPAP concentrations ranging from 1.25 to $9.15 \mu g/kg$, expected to be a remnant from the paper-making process. This is suggestive that all paper products, including those meant to be absorbent (e.g., paper napkins), or otherwise unexpected to have received a PFAS treatment for water- or grease-resistance, may be contributors of precursor compounds

which may be transformed into PFCAs in finished compost. Second, our results show the presence of NEtFOSA, a precursor which can degrade to release PFOS,² the only other sulfonate detected in the serviceware compost among PFAAs. That said, PFOS was also present at similar levels in the manure sample and was detected in one food waste sample indicating that manure and food could also be sources of this contaminant.

Another finding from this case study is that the 2019 serviceware compost samples contain less of the longer chain and more of the shorter chain compounds than the 2014-2017 serviceware composts. This could result in a greater tendency of PFAS in the compost to leach into water systems, as the shorter chain compounds are more mobile and less likely to sorb to soil and compost particles.^{28,29} The difference could be due to a longer maturation time resulting in higher concentrations of larger, immobile compounds and leaching of the smaller molecules as compost continues to mineralize. Alternatively, this result could reflect a shift in industrial/manufacturing PFAS usage from larger to smaller molecules over time. The impetus behind the switch to shorter chain compounds was their lower tendency to bioaccumulate and biomagnify in fauna;³⁰ thus, they were not expected to reach toxic levels in animal tissue. However, PFAS that remain in the water phase are more likely to be taken up by plants, and also to be translocated to other tissues from the roots³¹ resulting in a higher potential for exposure through food and water.

While these serviceware-inclusive compost results are of great concern, there is solace in the low- to entirely nondetectable levels of PFAS compounds found in food and manure-based composts presented here. The 2019 food compost pile did contain relatively small amounts of some long-chain PFCAs. This contamination food waste and serviceware materials, loading food waste compost of for transport using the same equipment used to turn serviceware composts, or their presence at low levels in food.³

In European and Canadian surveys of PFAS in disposable food packaging and serviceware, less than 1% of TOF present and less than 6% of total F present, respectively, could be assigned to specific PFAS compounds.^{16,17} This suggests a large quantity of PFAS compounds in use, but not commonly targeted for analysis, reinforcing the importance of performing TOF analysis in tandem with specific PFAS analysis.

Another aspect regarding PFAS-contaminated soil or compost, infrequently discussed—but of potential concern—are possible health risks to farmers and composting facility workers stemming from inhalation and/or ingestion of contaminated organic matter, which is easily made windborne, and can easily leave a considerable residue on hands.^{32,33}

IV. CONCLUSIONS

We found that compost made from manure and compostable serviceware was contaminated with 20–45 times more PFAS chemicals than separated food waste composted with grass clippings and manure. A sample of manure from the source farm was shown to only contain one PFAS (PFOS) at a relatively low concentration (3.7 ppb), whereas the serviceware compost contained 12–13 PFAS ranging from short-chain (C-4) to long-chain (C-14) PFCAs, a

small amount of PFOS, and a sulfonate precursor compound. Based on this case study, compostable serviceware is a potential source of PFAS contamination of composted food waste, and the current guidelines limiting the amounts of fluorinated organics in compostable serviceware are insufficient to prevent contamination of compost with PFAS at levels above Maine's soil PFOA remedial action guideline to prevent leaching to groundwater, and Maine's former screening threshold for biosolids for "beneficial use."

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Ethics Approval

Ethics approval is not required.

Author Contributions

Caleb P. Goossen: Conceptualization (lead); Data curation (lead); Investigation (lead); Writing – original draft (lead). Rachel E. Schattman: Writing – review & editing (equal). Jean D. MacRae: Conceptualization (supporting); Investigation (supporting); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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34See supplementary material online for supplemental Fig. 1 and Table I as described in the text.

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