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PFAS occurrence and distribution in yard waste compost indicate potential volatile loss, downward migration, and transformation†

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We discovered high concentrations of PFAS ($18.53 \pm 1.5 \mu\text{g kg}^{-1}$) in yard waste compost, a compost type widely acceptable to the public. Seventeen out of forty targeted PFAS, belonging to six PFAS classes were detected in yard waste compost, with PFCAs ($13.51 \pm 0.99 \mu\text{g kg}^{-1}$) and PFSAs ($4.13 \pm 0.19 \mu\text{g kg}^{-1}$) being the dominant classes, comprising approximately 72.5% and 22.1% of the total measured PFAS. Both short-chain PFAS, such as PFBA, PFHxA, and PFBS, and long-chain PFAS, such as PFOA and PFOS, were prevalent in all the tested yard waste compost samples. We also discovered the co-occurrence of PFAS with low-density polyethylene (LDPE) and polyethylene terephthalate (PET) plastics. Total PFAS concentrations in LDPE and PET separated from incoming yard waste were $7.41 \pm 0.41 \mu\text{g kg}^{-1}$ and $1.35 \pm 0.1 \mu\text{g kg}^{-1}$, which increased to $8.66 \pm 0.81 \mu\text{g kg}^{-1}$ in LDPE and $5.44 \pm 0.56 \mu\text{g kg}^{-1}$ in PET separated from compost. An idle mature compost pile revealed a clear vertical distribution of PFAS, with the total PFAS concentrations at the surface level approximately

Environmental significance

The presence of PFAS in various ecosystems is a critical issue, with growing evidence of their harmful effects. We discovered significant PFAS contamination in yard waste compost, a concern given its extensive use in agriculture. PFAS, often co-existing with plastics in compost, underscores the urgent need for creative strategies to minimize risks such as water pollution and exposure to humans and wildlife through land application. We also uncovered PFAS's vertical movement and potential transformation in compost, pointing to possible volatile loss and downward migration. This insight is vital for comprehending PFAS fate in compost and fostering innovative removal techniques. The outcome will transform compost into a tool for enhancing soil health, rather than a conduit for persistent organic pollutants.

58.9–63.2% lower than the 2 ft level. This difference might be attributed to the volatile loss of short-chain PFCAs, PFAS's downward movement with moisture, and aerobic transformations of precursor PFAS at the surface.

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† Electronic supplementary information (ESI) available: Supporting texts on compost carbon analysis, dissolved organic matter analysis, FTIR analysis of plastics, advanced image analysis of plastics surface, quality assurance and quality control (QA/QC), UPLC-MS/MS analysis; supporting tables on PFAS concentrations in different compost types, moisture content and maturity indices, carbon content, mass-labeled PFAS internal standards, FTIR operating conditions, UPLC-MS/MS operating conditions, extended forms of PFAS, one-way ANOVA and multivariate two-way ANOVA analysis, linear regression analysis; supporting figures of an idle compost pile, FTIR spectra of plastics and indices, cracks on plastic surface, total PFAS versus DOM at different depths, total PFAS versus total carbon at different depths, concentrations of the investigated PFAS at different depths, and runoff from the compost site. See DOI: <https://doi.org/10.1039/d3em00538k>

1. Introduction

The pervasive presence of per- and polyfluoroalkyl substances (PFAS) in the environment is concerning due to their potential long-lasting and harmful effects on humans, animals, and other organisms.^{1–5} Because of the widespread and unrestricted historical use of PFAS in daily life products and injudicious disposal, PFAS have become pervasive in the environment.^{6,7} A recent study identified 160 different PFAS in biosolids and compost with a total PFAS concentration of 580–1300 $\mu\text{g kg}^{-1}$.⁸ One pathway of PFAS transport to agricultural lands is *via* biosolids and compost application.^{9,10} For instance, approximately 320–920 $\mu\text{g kg}^{-1}$ of perfluoroalkyl acids (PFAAs) were identified in agricultural land that received municipal biosolids for more than ten years.¹¹ Approximately 50% of sewage sludge in Europe and North America is processed for agricultural use,¹² and 35% of US households utilize compost for gardening.^{13,14} Besides the negative impact on the soil microbial



community,^{15–18} plant uptake of PFAS has been investigated, which can create significant health consequences for humans and livestock.^{19–24} Over time, PFAS can leach from compost into surface runoff, contaminating natural water bodies, and can also percolate through soil, polluting groundwater.^{8,9} This led many US states, such as Maine and Vermont, to ban PFAS-containing compostable materials (*e.g.*, sewage sludge) for agricultural use.^{25,26}

We critically summarized the PFAS concentration in compost from different studies (Table S1†) and identified no single studies on pure yard waste compost, possibly because of the minimal chance of PFAS contamination of yard waste. Studies reported 0.03–133.30 $\mu\text{g kg}^{-1}$ PFAS in biosolid and manure compost,^{8,27} 31.5–70.8 $\mu\text{g kg}^{-1}$ in municipal organic compost (*i.e.*, food waste, leaves, grass, and horse manure),²⁸ 8.6–16.3 $\mu\text{g kg}^{-1}$ in green waste compost (*i.e.*, kitchen waste, yard trimmings),²⁹ and 6.8–11.84 $\mu\text{g kg}^{-1}$ in commercially available compost (*i.e.*, organic feed unknown).³⁰ Though these studies investigated PFAS in different types of compost, exploring PFAS in pure yard waste compost remains limited. Typically perceived as the purest type of compost, yard waste compost is widely used in many households for gardening. Thus, we investigated forty PFAS in mature yard waste compost, investigated incoming and composted plastics to understand plastics-PFAS co-occurrence, and investigated an idle mature compost pile to elucidate the vertical distribution profile of PFAS. The findings of this study will elucidate the extent of PFAS contamination in yard waste compost and inform the development of global remediation strategies for PFAS in solid materials.

2. Materials and methods

2.1. Yard waste compost

The City of Fargo (North Dakota, USA) residents can drop off yard waste at various locations year-round for free collection by the city authority. The Fargo yard waste compost site accepts tree leaves and grass from the residents. Unfortunately, there has been a tendency to dump unwanted substances on the site (*e.g.*, plastics, paper, packaging) alongside yard waste, mainly due to the lack of awareness and supervision. Yard waste composting typically takes 3–4 months.³¹ The mature compost from the 13-acre Fargo compost site is then piled together for subsequent screening. We collected mature compost in October 2020 just after final screening with a Komptech Trommel Screen, which ensured proper homogenization. When the compost site has a surplus of mature compost, they store it inside the locked fence (Fig. S1†). We also collected yard waste compost from the surface and 2 ft depth of the compost storage pile from three locations at least six feet apart (composts A, B, and C) in July 2021. The compost storage pile, approximately 1500 cubic feet in volume, has been idle inside a locked fence (100 ft \times 100 ft) for over six months.

We also separated and collected bulk plastics from incoming fresh yard waste and composted yard waste. All the compost and plastic samples were collected in LC-MS grade methanol-cleaned plastic zip-lock bags and stored in

a refrigerator at 4 °C before analysis and extraction. After proper homogenization, a certain mass of the composts was extracted using the ASTM D3987-12 protocol,³² and the ammonium and nitrate concentrations in the extracts were quantified to calculate the maturity index ($\text{NH}_4^+/\text{NO}_3^-$) of the compost samples.³³ The extracts were filtered through 0.45 μm Whatman filter papers (Sigma Aldrich, MO, USA) and the organic carbon (as dissolved organic matter) of the extracts was measured using a Shimadzu TOC Analyzer (Text S1 and Table S2†). All the collected compost samples were fully mature (maturity index <0.5) (Table S3†). The total carbon, organic carbon, and total inorganic carbon concentrations of all the compost samples were quantified using a Shimadzu TOC Solid Sample Analyzer (Text S2 and Table S2†). Monthly total precipitation and average temperature data at the compost site from Jan 2020 to Dec 2021 are provided in Table S4.†

2.2. PFAS extraction and analysis

Homogenized compost samples were sieved twice through a 1.5 mm pore-sized LC-MS grade methanol-cleaned aluminum net. Ten grams of compost from each compost were taken in triplicates in pre-cleaned 50 mL polypropylene tubes. Then, 100 μL mass-labeled PFAS internal standard (MPFAC-HIF-ES, Wellington Laboratories, Guelph, ON, Canada) was added to the sample and adequately mixed (Table S5†). The targeted analytes, extracted internal standards, and non-extracted internal standards were similar to the EPA draft method 1633 (ref. 34) (Table S6†). 10 mL of LC-MS grade methanol (Fisher Scientific, USA) was added to the sample tubes for extraction on a shaker (Thermo Scientific, MA, USA) at 200 rpm for 24 hours, followed by room temperature centrifugation at 10 000 rpm for 2 hours. After centrifugation, methanol was poured into pre-cleaned 10 mL polypropylene tubes and concentrated to 1 mL with a nitrogen blowdown evaporator. For PFAS extraction from plastics, plastics were first dried at room temperature and dusted off loose organics using compressed air. Plastics were cut down into small pieces (\sim 10 mm), and a certain mass was extracted by following the same protocol for compost. A detailed characterization procedure for these plastics, which includes crack analysis on the surface, can be found in the ESI (Text S3, S4, and Table S7†). We also prepared procedural blank samples to assess background contamination during extraction (Text S5†). The extraction recovery for the mass-labeled PFAS internal standards was 58–126%, similar to previous studies that used a low concentration of ammonium salts with methanol for PFAS extraction from compost (Table S1†).

We targeted forty PFAS (demonstrated in the EPA draft method 1633 (ref. 34)) in compost and plastics from seven PFAS classes. These include eleven perfluorocarboxylic acids (PFCAs), eight perfluorosulfonic acids (PFSAs), three fluorotelomer carboxylic acids (FTCAs), three fluorotelomer sulfonic acids (FTSSs), seven perfluoroalkane sulfonyl fluorides (PASFs) or sulfanomido compounds, five perfluoroether carboxylic acids (PFECAs), and three perfluoroether sulfonic acids (PFESAs). The PFAS were quantified using an ultra-performance liquid chromatograph (UPLC) coupled with a Triple Quadrupole mass



spectrometer (Vanqish UPLC-Altis MS, Thermo Scientific, USA) in the negative ionization mode. UPLC separation was carried out using a Phenomenex Luna Omega C18 column (2.1×100 mm) following an already published protocol (Text S6 and Table S8†).^{35,36} The measured PFAS mass was normalized by the dry mass of compost. Statistical analysis and plotting were performed using OriginPro (version 2023b, 10.05). Two-way multivariate analysis (MANOVA) was performed to understand the impact of sampling depth and moisture on the distribution of PFAS. We also conducted a one-way analysis of variance (ANOVA, $\alpha \leq 0.05$) for the total carbon and dissolved organic matter (DOM) at the surface and at 2 ft depth in composts A–C to find the correlation with PFAS distribution. Additionally, linear regression analysis (significant at $\alpha \leq 0.05$) of DOM *versus* total PFAS at varying depths was conducted to understand the correlation between DOM and total PFAS distribution. Pearson correlation was performed to test for the co-occurrence of different PFAS at different depths. Furthermore, we calculated the ratio of PFAS concentrations at 2 ft to the PFAS concentrations at the surface level. Linear regression analysis was performed between these values and the water solubility and vapor pressure values of PFAS to elucidate the correlation (significant at $\alpha \leq 0.05$).

3. Results and discussion

3.1. PFAS in yard waste compost

Out of the forty targeted PFAS, seventeen compounds spanning six PFAS classes (excluding FTCAs) were found in yard waste compost (Table S9† and Fig. 1). Dominantly, 72.5% were PFCAs and 22.1% were PFSA (Fig. 1). Prior research indicates that 60–70% of the targeted PFAS in municipal organic and biosolid composts were PFCAs and PFSA, together termed perfluoroalkyl acids (PFAAs).^{13,28} In the yard waste compost, short-chain PFAAs, such as perfluorobutanoic acids (PFBA), perfluorohexanoic acids (PFHxA), and perfluorobutane sulfonic acids (PFBS), were detected at high concentrations. Specifically, concentrations of $11.15 \pm 1.5 \mu\text{g kg}^{-1}$ PFBA, $1.34 \pm 0.07 \mu\text{g kg}^{-1}$ PFHxA, and $1.53 \pm 0.12 \mu\text{g kg}^{-1}$ PFBS were detected (Fig. 1). Correspondingly, previous studies reported high concentrations of PFBA (0.15 – $12.04 \mu\text{g kg}^{-1}$), PFHxA (1.06 – $1.39 \mu\text{g kg}^{-1}$), and PFBS (0.79 – $7.63 \mu\text{g kg}^{-1}$) in commercial and municipal organic waste compost^{28,30} (Table S1†). Long-chain PFAS, including PFOA ($0.44 \pm 0.04 \mu\text{g kg}^{-1}$), linear PFOS ($0.86 \pm 0.08 \mu\text{g kg}^{-1}$), branched PFOS ($0.43 \pm 0.06 \mu\text{g kg}^{-1}$), and PFHpS ($1.15 \pm 0.045 \mu\text{g kg}^{-1}$), were also detected in the yard waste compost (Fig. 1). Notably, US chemical manufacturers voluntarily phased



Fig. 1 Concentrations of seventeen different PFAS from six classes in yard waste compost. Among these, only perfluoro-3-methoxy propanoic acid (PFMPA) was detected in the PFECA class, and 11-chloroeicosafuoro-3-oxaundecane-1-sulfonic acid (11-Cl-PF₃OudS) was detected in the PFESA class. The average and standard deviation for each compound were calculated from six measurements, and extended details are available in Table S9.† Perfluoroalkane sulfonyl fluorides (PASFs) represent three sulfonamido compounds.



out PFOA and PFOS in 2002 and 2015, respectively.³⁷ Similar to our findings, studies identified long-chain PFAAs (PFCAs > C7 and PFSAs > C5) in various composts^{13,29} (Table S1†). Additionally, considerable concentrations of 6:2 fluorotelomer sulfonic acid (6:2 FTS, $0.30 \pm 0.02 \mu\text{g kg}^{-1}$) and *N*-ethyl perfluorooctane sulfonamido acetic acid (EtFOSAA, $0.12 \pm 0.008 \mu\text{g kg}^{-1}$) were identified (Fig. 1). Fluorotelomer sulfonic acids can transform into PFAAs in composts.³⁰ This conversion involves the oxidation of carbon-fluorine bonds within fluorotelomer-based PFAS, yielding PFAAs.^{30,38} Hence, some detected PFAAs in compost might stem from fluorotelomers' transformation. Additionally, fluorotelomer alcohols (FTOHs) can transform into FTCAs during yard waste composting, similar to the transformation of 6:2 FTOH into 5:3 FTCA and 4:3 FTCA during sewage sludge composting.³⁹

3.2. Plastics and PFAS co-occurrence

LDPE and PET were identified as prevalent plastics in yard waste compost (Fig. S2†). This is because PET water bottles often end up in yard waste, and many individuals dispose of yard waste using LDPE bags. When examining the carbonyl index, vinyl index, and crystallinity before and after composting, negligible chemical degradation was observed (Fig. S3†); however, these plastics can act as carriers of adsorbed PFAS in the environment.^{40–43} We identified seven PFAS in the incoming

plastics and ten in the compost-separated plastics (Fig. 2). The incoming LDPE contained more total PFAS ($7.41 \pm 0.41 \mu\text{g kg}^{-1}$) than the incoming PET ($1.35 \pm 0.1 \mu\text{g kg}^{-1}$) (Fig. 2). This discrepancy is because LDPE, with its high flexibility, rough surface, and strong hydrophobicity, adsorbs more organic pollutants than PET.^{44–46} Additionally, the shape of PET bottles remains largely unchanged, limiting the surface area available for PFAS adsorption. PFAS concentrations varied between incoming and compost-separated LDPE and PET, suggesting PFAS enrichment on plastics during composting. The total PFAS in compost-separated LDPE and PET increased to $8.66 \pm 0.81 \mu\text{g kg}^{-1}$ and $5.44 \pm 0.56 \mu\text{g kg}^{-1}$, respectively. Notably, the mass of PFAS per kg in the combined LDPE and PET plastics is about 23.8% lower than the mass found in a kg of compost. Both types of plastics showed a dominance of short-chain PFAS (Fig. 2). Besides the original PFAS in incoming LDPE, PFHxA ($0.38 \pm 0.01 \mu\text{g kg}^{-1}$), MeFOSAA ($0.1 \pm 0.02 \mu\text{g kg}^{-1}$), and EtFOSAA ($0.18 \pm 0.03 \mu\text{g kg}^{-1}$) were adsorbed during composting (Fig. 2). This enrichment may be due to the composting process creating microscopic cracks in plastics, enhancing their surface area (Fig. S4†).^{47,48}

3.3. Vertical distribution of PFAS in a compost storage pile

The compost PFAS concentration at the surface level was significantly lower compared to the PFAS level at 2 ft depth,



Fig. 2 Ten out of forty analyzed PFAS were detected in low-density polyethylene (LDPE) and polyethylene terephthalate (PET) separated from incoming and composted yard waste. Extended forms of the PFAS compounds are available in Table S9.†





Fig. 3 The ratio of PFAS concentrations at 2 ft depths to the surface in the yard waste compost. To calculate the ratio, PFAS concentrations at 2 ft depths were divided by the PFAS concentrations at the surface for composts A, B, and C. Nineteen out of the forty analyzed PFAS were detected in these compost samples.

indicating potential volatilization loss or downward movement with moisture. The total PFAS concentration at 2 ft depth was 2.5, 2.1, and 2.5 times of the total PFAS concentrations at the surface level for composts A, B, and C (Fig. 3, S5 and S6[†]). Similarly, the total PFAS concentration at 1 ft depth was 57.65% greater than the surface level in compost A (Fig. S5 and S6[†]). Additionally, the concentration of two dominant PFAS classes, such as PFCAs and PFSAs, at 2 ft depths were 2.8–3.6 and 1.4–1.7 times of the surface level concentrations (Fig. 3). In compost A, at 2 ft depth, PFBA had the highest concentration ($2.95 \pm 0.08 \mu\text{g kg}^{-1}$), trailed by PFBS ($1.88 \pm 0.1 \mu\text{g kg}^{-1}$); a trend mirrored in composts B and C (Fig. S7–S9[†]). Of the short chain PFCAs, PFBA and PFPeA concentrations at 2 ft depth were approximately 5.9–9.3 times of the surface level concentrations in composts A–C (Fig. 3). Downward movement of short-chain PFAS was reported in soil columns because of the high water solubility from their terminal functional groups' water affinity.^{49,50} Thus, the precipitation (*e.g.*, rainfall, snow) at the compost site will directly affect the downward movement of water-soluble PFAS in the compost piles. The compost moisture

content ranged from 28.1 to 29.9% at the surface and 41.3 to 44.8% at 2 ft depths, similar to the PFAS distribution trend with depth (Table S3[†]). The ratio of PFAS concentration at 2 ft depths to the surface was positively correlated with the water solubility of PFAS (Fig. 4A and Table S10[†]), which suggests that the water solubility of PFAS impacted their downward movement in the compost pile.

Besides downward movement with moisture, volatile loss and transformation might explain PFAS reduction at the pile's surface. Literature reveals that short-chain PFAS (*e.g.*, PFBA, PFPeA, PFHxA) with high vapor pressure are more volatile than their long-chain counterparts (Fig. S10[†]). These short-chain PFAS compounds sorb to water vapor, increasing their long-range transport potential.^{51–53} A strong positive correlation between the vapor pressure of PFAS and the ratio of PFAS concentration at 2 ft depth to the surface in the compost samples was observed (Fig. 4B and Table S11[†]). This suggests the possibility of volatile loss of PFAS with high vapor pressure from the compost surface, leading to reduced PFAS levels at the surface. PFAA precursors such as fluorotelomer sulfonates and sulfonamides are semi-volatile due





Fig. 4 Linear regression analysis between the ratio of PFAS concentrations at 2 feet depth to surface level and their water solubility (A) and vapor pressure (B). To calculate the ratio, PFAS concentrations at 2 ft depth were divided by the concentrations at the surface for composts A, B, and C. The median value of water solubility and vapor pressure of PFAS were used for the analysis and plots. Median value for each PFAS were derived by consolidating data from 82 peer-reviewed sources for water solubility and 77 peer-reviewed sources for vapor pressure (Fig. S10†).

to their large molecular size and strong intermolecular forces, limiting evaporation.^{51,54} These precursors have low water affinity due to their longer carbon chains, making them more likely to sorb on solids.^{55,56} However, we found that semi-volatile PFAA precursors such as FOSA-1's concentrations at the surface level were 40–60% less compared to the 2 ft level, EtFOSAA was 45–60% less, and 6 : 2 FTS was 25–80% less at the surface than at 2 ft (Fig. 3 and S7–S9†). Besides the downward movement with moisture and volatile loss, this high percent loss from the surface could be attributed to the biological transformation of PFAA precursors into short-chain PFAAs at the top compost layer (0–10 cm) under oxygen-rich conditions.⁵⁷ Studies indicate that this biotransformation is more prevalent in surface soil than in deeper layers.^{57–59}

Long-chain PFAS were less mobile in the compost pile than short-chain PFAS and PFAS precursors. The change of long-chain PFAS concentrations at the compost surface was notable, but less than the change in short-chain and precursor PFAS concentrations (Fig. 3). The distribution of long-chain PFAS is influenced by both moisture and DOM in the solid medium.^{49,61–67} Interestingly, DOM at 2 ft depth (7.82–14.21 g kg⁻¹) was significantly ($\alpha \leq 0.05$) greater than the DOM at the compost surface (3.57–4.78 g kg⁻¹), showing a strong correlation with the total PFAS at varying depths (Tables S12, S13 and Fig. S5†). However, there was no significant variation in the total carbon content across depths in composts A–C (Tables S2, S14 and Fig. S6†). The fate of PFAS is also influenced by their structural variations, such as isomers. Research indicates that linear PFAS isomers tend to adhere to soil and sediments, whereas branched isomers are more prone to movement.⁶⁰ This difference is linked to the greater polarity of branched isomers relative to linear ones. Our findings revealed that branched PFOS levels were 2.5 times greater at 2 ft depth than at the surface, suggesting water-assisted downward movement (Fig. 3). Conversely, linear PFOS levels were similar at both depths, indicating sorption by compost. PFAS binding affinity on organics can also be affected by other chemical factors such as functional groups, aromaticity, and humification.^{50,68–70} Furthermore, biogeochemical elements like iron deposits on compost can also influence PFAS adsorption, especially with iron oxides binding short-chain PFAS *via* surface complexation or ion exchange.^{50,71} Redox gradients alter the distribution and form of iron oxides with depth, which could affect PFAS adsorption variability.^{72–74} Although theoretically, stronger surface adsorption of PFAS might occur by biogeochemical factors that could slow down PFAS's downward movement, this study did not show such evidence.

Composts at greater depths had notably more moisture than surface layers (Table S3†). Depth and moisture content were interdependent, significantly impacting the concentration of PFAS species at 0 and 2 ft depths (Table S15†). Statistical analyses showed significant correlations among certain short-chain PFAS (*i.e.*, PFBA, PFBS, PFPEA, PFHxA, and PFHpA) and between them and compounds like 6 : 2 FTS, 8 : 2 FTS, and FOSA-1, suggesting a shared origin (Fig. 5). Likewise, long-chain PFAS (*i.e.*, PFNA, PFDA, and PFUDA) were significantly correlated with one another and with certain short-chain compounds and precursors (*i.e.*, PFHpA, 8 : 2 FTS, and EtFOSAA), hinting at a common origin (Fig. 5). However, PFASs showed no distinct correlation in yard waste compost, indicating different sources. Prior research has noted strong correlations among various PFAS in liquid and solid mediums, pointing to shared origins.^{75–78} A leading contributor of PFAS in yard waste is plastics, which often end up in yard waste due to the lack of public awareness and monitoring. Paper, paper products, and packaging materials can also act as sources of PFAS in yard waste compost. Moreover, the water used to maintain the moisture content of yard waste can act as a source of PFAS in compost. The proximity of the compost site near the Fargo landfill increases the likelihood of PFAS entering the compost through atmospheric deposition, particularly due to the





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