

Per- and Polyfluoroalkyl Substances in Toilet Paper and the Impact on Wastewater Systems

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INTRODUCTION

With increasing frequency, scientists and policy makers rely on measurements from society's wastewater as sentinels of disease transmission,¹ drug use,² and chemical exposure and environmental release.³ Per- and polyfluoroalkyl substances (PFAS), ubiquitous chemicals now documented throughout the natural environment,⁴ wildlife,⁵ human blood,⁶ and breastmilk,⁷ have been similarly documented in wastewater,⁸ particularly the residual sewage sludge from biological wastewater treatment.^{9,10}

As PFAS occur in numerous household and personal care products,^{11,12} the measurement of PFAS in wastewater residuals comes as no surprise, but as much of the world's sewage sludge is land applied,^{13,14} the regulatory community now wrestles with how best to address the human health and environmental ramifications of PFAS discharges. Understanding the sources of PFAS and their respective contribution to wastewater will be key to prioritizing action—possible sources include the soaps, shampoo, and makeup that we wash down the drain,¹¹ clothes and related cleaning products in our laundry,¹⁵ and discharges from commercial and industrial operations.^{16,17}

Here, we evaluate a perhaps unexpected contributor of PFAS to our wastewater, an input anticipated at every wastewater treatment facility—toilet paper. PFAS have been documented in papers products¹⁸ and are a known wetting agent additive used to increase the efficiency of the pulping process.¹⁹ The use of PFAS by paper mills has already been identified as a source of environmental contamination²⁰ and could result in residual PFAS in toilet paper. Furthermore,

toilet paper is often manufactured from recycled paper fibers (which often can contain PFAS).^{21,22} We obtained packaged toilet paper from four world regions (Africa, North America, South and Central America, and Western Europe) and assayed the concentration of 34 PFAS. Additionally, we analyzed the same suite of PFAS in sewage sludge from eight wastewater treatment plants in Florida, United States (US), to complement the existing body of knowledge on PFAS in sewage sludge, and these data, along with estimates of average toilet paper usage and sludge generation, allowed us to assess the relative contribution of toilet paper to the PFAS loading, at least for the dominant detectable species of PFAS, in wastewater sewage sludge.

MATERIALS AND METHODS

Sample Collection and Extraction of Toilet paper. Samples of toilet paper were collected from November 2021 to August 2022 by a volunteer network of students and professors; individuals were given instructions to document the location of toilet paper collection, the toilet paper recycled content, brand, and a picture or physical label of the toilet paper if possible (see Figure S1.). Samples were either collected as entire rolls or 10 g subsamples. Samples were

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Figure 1. Concentrations of 6:2 diPAP and relative distribution of detected PFAS in toilet paper. (A) Concentration of 6:2 diPAP in toilet papers sampled from different world regions. (B) Concentration of 6:2 diPAP in toilet papers labeled as containing recycled paper compared to nonrecycled toilet papers. (C) Proportion of each PFAS detected across all toilet paper samples relative to the total mass of \sum_{6} PFAS. The proportion of each PFAS is as follows: 91% (6:2 diPAP), 3.7% (8:2/6:2 diPAP), 1.3% (8:2 diPAP), 3.3% (PFHxA), 0.6% (PFOA), and 0.4% (PFDA).

stored in sealed high-density polyethylene (HDPE) containers for no more than 180 days at 25 $^{\circ}$ C.

Samples were extracted following a modified method published by Timshina et al.²³ In short, triplicate 1 g subsamples of toilet paper were transferred to 50 mL falcon tubes and spiked with 40 μ L of an isotopically labeled internal standard mixture (see Tables S1 and S2). Extraction was repeated three times in the following manner: after adding 10 mL of 0.3% methanolic ammonium hydroxide, samples were agitated on a rotisserie rotator for 45 min, centrifuged for 10 min at 4000 rpm, and supernatants transferred to 50 mL centrifuge tubes. Extracts were evaporated to approximately 1 mL in a Biotage TurboVap before supernatants from the 3-fold extraction were combined. For purification, combined extracts were vortexed for 30 s with 50 mg ± 10 mg of ENVI-Carb graphitized activated carbon, centrifuged for 10 min, and transferred to new 15 mL tubes. The final extracts were evaporated to 1 mL and aliquoted for analysis. Extracts were stored no more than 30 days at -20 °C until analysis (see Table S4 for PFAS concentrations in toilet paper). Extraction efficiencies were measured using pooled samples spiked with mass-labeled PFAS (additional information about the QC process and reported extraction efficiencies can be found on page S4 and Table S10 of the SI).

Sample Collection and Extraction of Wastewater Sludge. Wastewater sludge samples were collected from eight facilities in Florida, US, between June 2021 and August 2021. Samples were collected in 23 L HDPE buckets using stainless steel shovels washed with methanol prior to and between collections. Samples were transported to the laboratory, aliquoted into 2 L HDPE bottles, and stored at -20 °C until analysis. The sample extraction method is a modified version of a previously reported solid matrix extraction method.²⁴

Prior to extraction, wastewater sludge samples were homogenized by rotating for 20 min at 70 rpm and air-dried in a fume hood for a period of 3 days. A subsample of air-dried wastewater sludge was taken to complete dryness at 110 ± 5 °C according to ASTM D2216²⁵ to determine the moisture content for calculating PFAS concentrations on a per dry mass basis. Air-dried wastewater sludge samples were extracted in triplicate and divided into 10.0 ± 0.1 g subsamples, added to a 50 mL polypropylene tube, and spiked with 50 μ L of an isotopically labeled PFAS IS mixture. Then, 8.5 mL of 0.3% ammonium hydroxide in methanol was added to the sample, and the mixture was vortexed for 1 min, sonicated for 30 min, and rotated in an end-over-end fashion for 30 min. The sample was centrifuged at 4000 rpm for 10 min, and the supernatant was removed using a pipet. An additional 8.5 mL of 0.3% ammonium hydroxide in methanol was added to the remaining sample, and the vortex, sonication, rotation, centrifugation, and supernatant removal steps were repeated. The combined extracts (17 mL) were concentrated to 10 mL under a gentle stream of high purity nitrogen gas (Biotage TurboVap II) at 35 °C. Extracts were then stored for less than 30 days at -20 °C until analysis. Evaporated extracts were aliquoted into 200 μ L

| Country | Median 6:2 diPAP concentrations in wastewater sludge (ng/g) | Average toilet paper usage per capita (kg-person/year) | Wastewater sludge generated per capita (kg-person/year) | Fraction of 6:2 diPAP in wastewater sludge attributed to toilet paper usage (%) | |
|--|---|---|--|---|--|
| United States ^{27,31,32} | 82 | 26 | 27 | 3.7 | |
| Canada ^{26,31,32} | 85 | 26 | 28 | 3.5 | |
| China ^{30,31,33} | 12 | 7 | 28 | 6.9 | |
| Australia ^{32,34,35} | 32 | 13 | 18 | 7.2 | |
| France ^{29,31,36} | 1.7 | 10 | 21 | 89 | |
| Sweden ^{28,31,36,37} | 5.6 | 15 | 25 | 35 | |
| a The methodology for determining the fraction of 6:2 diPAP in wastewater sludge attributed to toilet paper usage is presented in the SI. | | | | | |

Table 1. Fraction of 6:2 diPAP in Wastewater Sludge Expected to Come from Toilet Paper^a

polypropylene autosampler vials for analysis by liquid chromatography and tandem mass spectrometry (see Tables S5–S8 for concentrations of detected PFAS in wastewater sludge samples).

LC-MS/MS Analysis. Toilet paper and wastewater sludge extracts were analyzed via targeted ultrahigh pressure liquid chromatography-tandem mass spectrometry (UHPLC-MS/MS) using a Thermo Vanquish UHPLC system (Waltham, MA, US) coupled to a Thermo Quantis triple quadrupole mass spectrometer (operated with electrospray in negative ionization mode and scanning via scheduled selected reaction monitoring (SRM)).

A Gemini C18 (100 mm \times 2 mm; 3 μ m) column from Phenomenex was used for chromatographic separation. Water [A] and methanol [B] both containing 5 mM of ammonium acetate were used as the mobile phases. The gradient elution was set as 0-3 min 10% B, 3-4.5 min 10%-35% B, 4.5-12.5 min 35%-95% B, 12.5-12.51 min 95%-99% B, 12.51-19 min 99%, and then equilibrated to initial conditions in 30 min. The temperature of the autosampler was 4 °C. The flow rate was 0.5 mL min⁻¹, and injection volume was 10 μ L. Water, methanol, and ammonium acetate used in the study were all Optimal grade purchased from Fisher Scientific. Scheduled SRM mode (monitoring two transitions, if possible) was used to detect and quantify each PFAS (native and mass-labeled species are shown in Tables S1 and S2 of the SI). The most intense transition was used for quantification, while the second transition was used to confirm identification (if applicable). For PFAS where no isotopically labeled standard was available, an alternative labeled standard, either similar by structure or retention time, was selected for quantitation. Table S2 displays the mass spectrometric scan parameters (and transitions) for all targeted PFAS, while Table S3 displays additional LC-MS/ MS parameters. Additional information regarding the QA/QC protocol, extraction efficiencies for each matrix, and data analysis are available on pages S4-S5 of the SI.

RESULTS AND DISCUSSION

Of the 34 PFAS analyzed, six were detected in the toilet paper samples: perfluorohexanoic acid (PFHxA), perfluorooctanoic acid (PFOA), perfluorodecanoic acid (PFDA), 6:2 fluorotelomer phosphate diester (6:2 diPAP), 6:2/8:2 fluorotelomer phosphate diester (6:2/8:2 diPAP), and 8:2 fluorotelomer phosphate diester (8:2 diPAP). By far the largest single PFAS among those detected was 6:2 diPAP, representing 91% \pm 8% of the total mass of PFAS on average. A previous study identified elevated diPAP concentrations in paper mill wastes streams, suggesting that diPAPs could be introduced during production.²⁰ Analysis of variance (ANOVA) between regions, and a *t* test between recycled and nonrecycled samples, found that concentrations of 6:2 diPAP did not differ among regions nor those identified with recycled content (p > .05). These comparisons, along with the distribution of PFAS detected among toilet paper samples, are shown in Figure 1 (A) - (C).

Samples of wastewater sludge were collected and characterized for PFAS to explore the magnitude of the potential PFAS loading into wastewater systems from toilet paper. DiPAPs were similarly among the most abundant PFAS measured in wastewater treatment sludges, though they represented a smaller contribution than in the toilet paper itself. The sum of three diPAPs (6:2, 62:8:2, and 8:2 diPAP) in the eight sludge sources tested contributed $54\% \pm 15\%$ of the \sum_{34} PFAS, on average. Concentrations of 6:2 diPAP ranged from 27.2 to 750 ng/g with an average concentration of 141 ng/g (PFAS concentrations can be found in Tables S5–S8 of the SI). Two studies on North American sewage sludge similarly identified diPAPs as the most abundant PFAS measured with mean concentrations of 6:2 diPAP at 164 and 121 ng/g.^{26,27} Interestingly, reported results from Europe and China found diPAPs to contribute less to the total mass of PFAS present in wastewater sludge, with studies from Sweden, France, and China reporting mean concentrations of 2.0, 2.9, and 11.6 ng/g for 6:2 diPAP, respectively, which represented 3%, 2%, and 8% of the total mass PFAS measured. $^{28-30}$

The use of toilet paper, and whether it is flushed down the toilet or disposed of with trash, varies regionally as a function of cultural norms, economic means, and sanitation infrastructure. Reported per capita toilet paper use in regions such as Japan, the US, Canada, and Western Europe ranges from 15 to 25 kg/person-year, while estimated use in Latin America, China, and Africa ranges from 2 to 10 kg/person-year.³¹ To estimate per capita mass of 6:2 diPAP entering wastewater systems from toilet paper, a mean 6:2 diPAP concentration of 3.2 ng/g from the 21 samples analyzed was applied to the range of reported toilet paper usage rates. This results in an annual 6:2 diPAP contribution from toilet paper of 6.4 to 80 μ g/person-year.

We next examined the contribution of toilet paper to the total 6:2 diPAP found in wastewater sludge using region-specific assumptions (concentration in sludge, per capita toilet paper use, and per capita sludge generation; Table 1). The estimated contribution from toilet paper was lowest in the US (3.7%) and Canada (3.5%) and highest in Sweden (35%) and France (89%). The larger contribution observed in the two European data sets stems from the lower reported 6:2 diPAP sludge concentrations compared to those measured in North America (by an order of magnitude). Comprehensive PFAS data on regional wastewater slude sources are remarkedly limited, but in those nations where multiple data sets were available (US and Sweden), each nation's data were of the

same magnitude (median of 7.0 and 1.8 ng/g in the Swedish studies and 53 and 100 ng/g in the US studies).

Wastewater Implications. Our results suggest that toilet paper should be considered as a potential significant source of PFAS entering wastewater treatment systems. The lower estimated 6:2 diPAP contribution derived from the North American data sets, despite the greater reported use of toilet paper in this region, suggests that other 6:2 diPAP sources, such as cosmetics,¹¹ textiles, and food packaging,³⁸ deserve attention, especially given the apparently smaller contribution of these sources based on data sets from France and Sweden. Previous work from D'eon et al.²⁶ suggests that diPAPs are a prevalent North American chemical contaminant. Perhaps regional consumer product choices and discard practices can inform strategies to reduce wastewater borne PFAS loads. This reduction in PFAS is critical, since wastewater effluent and sludge are commonly reused for irrigation and/or land application;³² research has already shown that these two pathways pose a risk for human and environmental exposure to PFAS.³

Complicating this discussion, however, is the fact that the dominant PFAS family observed in toilet paper and wastewater treatment sludge, the diPAPs, are precursor species and have the capacity to be transformed into terminal PFAS. Terminal species-such as PFHxA, PFOA, or PFDA-are formed from the biologically mediated transformation of 6:2 diPAP and other diPAP homologues,4,40 and these chemicals are the growing targets of regulatory attention due to the expanding body of knowledge regarding their human health and environmental impacts.41 Additional research is needed to explore whether toilet paper might be a greater contributor to total PFAS in North American wastewater and if the diPAPs from toilet paper might be transforming through the wastewater collection and treatment system. Furthermore, PFAS known as triPAPs and larger analogs have been detected in the environment.⁴² These compounds may also be present in toilet paper; however, determining the quantity of these species is difficult due to the lack of available standards and the limitations of the total oxidizable precursor (TOP) assay for diPAP-containing and/or high organic matter matrixes.^{27,43}

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.3c00094.

Sample collection information, list of materials, previous studies, instrument parameters, extraction efficiencies, and complete data sets (PDF)

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Notes

The authors declare no competing financial interest.

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