

# **Elevated PFAS Precursors in Septage and Residential Pump Stations**

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compounds, including short-chain alternatives, highlights shifting contamination patterns. Findings emphasize the need for tailored analytical frameworks and pretreatment technologies to mitigate PFAS risks across decentralized and centralized wastewater systems. Integrating precursor analysis is critical for accurate risk assessment, as targeted PFAS measurements underestimate contamination. These results provide new insights into PFAS behavior in residential wastewater, guiding future mitigation efforts.

KEYWORDS: PFAS, TOP assay, municipal wastewater, septage, septic systems

# INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) are widely used in household and industrial applications due to their resistance to heat, water, and oil.<sup>1-3</sup> In residential settings, PFAS can enter wastewater streams through various household products and activities. Common sources include stain-resistant carpets, nonstick cookware, waterproof clothing, food packaging, personal care products, and cleaning agents (Table S1). These products often contain PFAS precursors which are released during routine domestic activities such as laundry, dishwashing, and cleaning.<sup>8,9</sup> Unlike industrial wastewater, residential wastewater is typically assumed to contain lower PFAS concentrations, but recent evidence suggests that household sources can contribute significant loads of PFAS precursors.<sup>10</sup> Even centralized wastewater treatment plants exhibit limited PFAS removal efficiencies globally, with studies reporting minimal reductions in terminal PFAS and precursors during treatment.<sup>11</sup> This poses challenges for both centralized and decentralized wastewater management systems, as PFAS and their precursors often bypass conventional treatment processes.<sup>12</sup>

Residential wastewater management systems, including septic systems and pump stations, serve as critical nodes where PFAS precursors accumulate and undergo potential transformation.<sup>13</sup> Septic systems, which are commonly used in rural and suburban areas without access to centralized

treatment, are designed primarily to manage organic waste and pathogens, with limited capacity to remove synthetic chemicals like PFAS.<sup>14</sup> As a result, PFAS and their precursors often persist in septic effluent, posing a significant risk of leaching into surrounding soils and groundwater.<sup>15</sup> In contrast, residential pump stations, which collect wastewater from multiple households for transport to centralized treatment facilities, can provide a composite profile of PFAS precursor contamination in residential wastewater.<sup>16</sup> Comparing these systems offers valuable insights into the distribution and behavior of PFAS precursors in decentralized versus centralized wastewater settings.

Despite the critical role of septic systems in wastewater management, serving approximately 60 million people in 21% of households in the United States, there is limited data on their contribution to PFAS contamination.<sup>17</sup> As the number of septic systems is increasing with ~1.5 million systems are being installed each year, septic systems represent a significant

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Figure 1. Graphical illustration of the different scenarios and sample types captured within the study area.

pathway for PFAS precursors to enter the environment.<sup>15,17</sup> Similarly, residential pump stations, while serving as intermediary systems in centralized treatment networks, have not been extensively studied. This study aims to bridge these knowledge gaps by systematically quantifying PFAS precursor concentrations in both wastewater from septic systems and residential pump stations. Through a comprehensive sampling effort targeting 12 pump stations over a one-month period and 17 distinct septage samples, this research seeks to compare PFAS precursor levels between these systems and identify hotspots of contamination. The analytical approach for this study involved targeted PFAS analysis, covering a comprehensive range of 70 PFAS compounds, along with pretreatment using the Total Oxidizable Precursor (TOP) assay. These findings provide valuable insights to support treatment operators in addressing the complexities of PFAS contamination, enhancing risk assessment processes, and developing effective management strategies.

## METHODS AND MATERIALS

**Municipal Sewer District.** The targeted district collects and treats an average of 185 million gallons a day (MGD) of wastewater from different sources, including ~232,000 house-holds in a 290 square mile area conveyed through 3000 miles of sewer pipe. For the sewer-shed examined in this study (Figure 1), number of septic systems is ~20,000 with daily trucked wastewater to the plant of ~145,000 Liters. The number of residential-only pumpstations is 89 with daily pumped wastewater to the plant of ~26 × 10<sup>6</sup> Liters.

**Sampling Plan and Analytical Methods.** Grab samples were collected over a 4-week period between February and March 2024. A stratified sampling approach was employed to ensure geographic and demographic representativeness (Table S2). Twelve pumpstations were sampled twice with one month time gap to capture potential temporal variations. A detailed sampling plan is described in Section S2. Detailed descriptions of PFAS analytical methods are listed in Section S3 in the Supporting Information (SI) file. Briefly, samples were analyzed by Modified EPA 537 Method at commercial lab (Eurofins) for 70 analytes via targeted analysis using LC-MS/

MS, isotope dilutions or extracted internal standard calibrations were included. Samples were also treated using the TOP Assay technique following the method developed by Houtz and Sedlak.<sup>18</sup> The results before and after TOP Assay pretreatment were compared to evaluate PFAS precursors in both pump station and septage samples. Method blanks showed no detectable PFAS in pre-TOP samples. Post-TOP blanks contained trace PFPrA (<5 ng/L), attributed to oxidation of spiked M2-4:2 FTS surrogate. Method detection limits (MDLs) ranged from 0.5-5 ng/L. Isotope dilution analyte recoveries were 70-130% for 92% of compounds. Section S4 describes water quality parameters including biological oxygen demand (BOD), chemical oxygen demand (COD), dissolved organic carbon (DOC), total suspended solids (TSS), nitrate, nitrogen, ammonia and oil and grease. Water quality data measured at pump stations and in septage were compared to evaluate differences between station types.

# RESULTS AND DISCUSSION

Differences in Organic and Inorganic Contaminants. Septage samples exhibited significantly higher concentrations of organic and inorganic contaminants than pump stations (Table S5), reflecting the accumulation of pollutants in septic systems (Figure 2A). Elevated levels of DOC (168.6  $\pm$  151.6  $mg/L vs 140.1 \pm 140.9 mg/L$ ), BOD (2279  $\pm 1004 mg/L vs$  $202 \pm 34 \text{ mg/L}$ , COD (8266  $\pm 3187 \text{ mg/L}$  vs 632  $\pm 364$ mg/L), and TSS (8170  $\pm$  3509 mg/L vs 194  $\pm$  124 mg/L) highlight the lower water quality of septage. High BOD and COD levels indicate substantial organic loads, creating anaerobic conditions that may slow precursor degradation. While DOC levels were similar between matrices (p = 0.752), septage retained more particulate-bound contaminants, increasing PFAS sorption potential.<sup>19</sup> The greater organic complexity of septage contributes to precursor persistence and incomplete oxidation, complicating downstream PFAS treatment.<sup>20</sup> These findings emphasize the need for tailored management strategies to address PFAS in decentralized systems and reduce pollutant release risks.<sup>14,21</sup> Detailed implications are discussed in Section S5.



**Figure 2.** Box-and-whisker plot showing the differences between septage and pump stations in [A] DOC, BOD, COD, and TSS concentrations. [B] Total targeted PFAS pre- and post-TOP assay. Boxes represent the interquartile range with the median indicated by the horizontal line and mean values as blue squares. Whiskers extend to 1.5 times the interquartile range, and diamonds represent actual measured concentrations for each parameter. All raw data are summarized in File S2.

Total Targeted PFAS and TOP Assay Analysis. The analysis of total targeted PFAS concentrations demonstrated a clear distinction between septage and residential pump stations (Figure 2B), with septage showing significantly higher (Table S6) median concentrations (176.2 ng/L) compared to pump stations (21.9 ng/L). This disparity reflects the contrasting operational characteristics of these systems and their roles in wastewater management. These concentrations result in daily average targeted PFAS loads of  $2.6 \times 10^7$  ng and  $5.7 \times 10^8$  ng from septage and pump stations, respectively. Septic tanks, by design, retain wastewater over extended periods, allowing for the accumulation of PFAS and their precursors.<sup>13</sup> In contrast, pump stations function as transitional systems, rapidly transporting wastewater to centralized facilities, resulting in lower overall PFAS concentrations. Anomalously high PFAS levels in a subset of pump station samples, comparable to those in septage, point to the influence of specific household discharges and potential temporal variations as discussed in the next section. Household-specific factors such as the frequency

of cleaning activities, use of PFAS-containing products, and variability in consumer behavior have been identified as significant contributors to PFAS variability in residential wastewater.<sup>10</sup> Similarly, the role of household consumer goods, such as water-repellent fabrics and nonstick cookware, was also previously highlighted as key sources of PFAS in wastewater from decentralized systems.<sup>13</sup>

The TOP assay results revealed a substantial increase in PFAS concentrations post-treatment for both septage and pump stations. Median post-TOP concentrations were 687.5 ng/L for septage and 84.2 ng/L for pump stations, with the former showing a more pronounced increase. This indicates that septage harbors significantly higher concentrations (p < p0.001) of PFAS precursors compared to pump stations, consistent with findings that anaerobic conditions, typical of septic tanks, inhibit precursor degradation and promote their persistence.<sup>13,22</sup> The elevated precursor loads in septic systems position them as major point sources of PFAS to surrounding soils, groundwater, and WWTPs.<sup>8,15</sup> Particularly, the high precursor concentrations in septage have critical implications for WWTP operations.<sup>23</sup> When septic wastewater reaches WWTPs, oxidative processes such as aeration and chemical treatments can convert these precursors into terminal PFAS, increasing the measurable PFAS load in both treated effluent and biosolids.<sup>12</sup> This transformation amplifies PFAS burdens within the facility, complicating effluent compliance and increasing the environmental risk associated with biosolid land applications. While pump stations exhibit lower precursor concentrations due to shorter retention times and more dynamic flow conditions, they still contribute to the overall precursor load entering WWTPs, albeit to a lesser extent.<sup>8</sup>

These findings highlight the necessity of integrating precursor analysis into PFAS monitoring frameworks to better understand and manage contamination risks. Reliance on targeted PFAS measurements alone underestimates the true extent of PFAS contamination, as precursors represent a substantial proportion of the total PFAS potential. Expanding monitoring to include precursors would enable a more accurate assessment of risks and inform targeted mitigation efforts.<sup>10</sup> Furthermore, targeted interventions such as pretreatment systems for septic tanks (e.g., aerobic treatment or disinfection) and improved sludge management practices could help mitigate the risks associated with decentralized systems.

**Temporal Differences at Pump Stations.** The analysis of PFAS concentrations at 12 pump stations, sampled twice with a one-month interval, revealed significant temporal variability (p  $\ll 0.001$  via a two-tailed t test with equal variances) in both pre- and post-TOP assay results (Figure S1). This variability was evident even at the same locations, underscoring the dynamic nature of residential wastewater systems and the challenges in characterizing PFAS contamination reliably. Pre-TOP assay samples showed a higher frequency of nondetects compared to post-TOP assay results, which highlight the limitations of targeted PFAS analysis alone. The oxidative conditions of the TOP assay uncovered substantial hidden precursor contributions,<sup>24</sup> emphasizing the risk of false negatives when precursors are not accounted for. Similar observations have been reported in monitoring other persistent organic contaminants, such as pharmaceuticals and personal care products, where episodic inputs and incomplete analytical coverage have led to underestimated concentrations in environmental samples.<sup>25,26</sup> The variability observed in

PFAS concentrations can be attributed to several temporal and spatial factors, as described in Section S6.<sup>27</sup> Mitigating these temporal variations necessitates innovative sampling and management strategies.

Speciation of Targeted PFAS and Precursors. This study identified 27 out of 70 monitored PFAS compounds across septage samples and pump stations (Figure 3), a



**Figure 3.** Concentrations of detected PFAS pre- and post-TOP assay in samples from [A] pump stations and [B] septic tanks. Error bars represent the mean  $\pm$  standard deviation. All raw data are summarized in File S2.

significant increase compared to many previous investigations, which often limit analyses to fewer compounds. For instance, Lin et al. (2024) targeted 40 PFAS compounds in their investigation of residential wastewater contributions to WWTPs in the San Francisco Bay Area and found that even with a comprehensive list, a significant portion of PFAS precursors remained undetected prior to the TOP assay.<sup>10</sup> The extended analyte list in the present study enabled a more nuanced understanding of PFAS dynamics, revealing critical differences in precursor profiles and concentrations between decentralized and centralized systems. The most frequently detected compounds were perfluorooctanesulfonic acid (PFOS) (detected in 65% of samples) and perfluorobutanoic

acid (PFBA) (detected in 60% of samples (File S1). Shortchain PFAS, such as PFBA and perfluoropentanoic acid (PFPeA), were more prevalent than long-chain compounds. Similarly, carboxylic acids were more prevalent and abundant than sulfonic acids.<sup>28</sup> A notable finding was the detection of diPAPs (dialkyl perfluoroalkyl phosphates), key PFAS precursors, in both septage and pump station samples. DiPAPs, frequently used in consumer products like paper coatings and textiles, are known to degrade into terminal PFAS under oxidative conditions. Septage samples consistently exhibited higher concentrations of diPAPs than pump stations, likely due to their high solids concentration, anaerobic conditions and extended retention times, which promote precursor persistence.<sup>29,30</sup> This aligns with findings from Thompson et al. (2023), who highlighted the ubiquity of diPAPs in household products that contribute to wastewater contamination.4,31 Previous research on municipal sources (Table S1) helps explain the expected PFAS concentrations. While drinking water typically contains low PFAS, certain household products can contain up to 1,000,000 ng/g of PFAS and their widespread use and disposal could contribute to PFAS accumulation in septic systems.

The post-TOP assay results further emphasized the critical role of precursors. Both septage and pump station samples showed significant increases (p < 0.001) in PFAS concentrations after oxidation and consistent decreases in targeted PFAS precursors (e.g., FTCAs and diPAPs), with septage samples displaying a more pronounced rise (Figure 3B). In comparison to Lin et al. (2024),<sup>10</sup> where residential wastewater from diverse neighborhoods showed PFAS levels averaging 287 ng/L post-TOP assay, the concentrations in septage in this study were almost 4 times higher (1176.5 ng/L). This disparity underscores the need to differentiate between centralized and decentralized systems in PFAS management strategies. Furthermore, the dominance of short-chain PFAS in both pump stations and septage mirrors the findings of Lin et al.,<sup>10</sup> reflecting the industry shift away from long-chain compounds due to regulatory restrictions.<sup>32,33</sup> These observations further support previous reports on speciation and concentrations PFAS and precursors transformation between the influents and effluents of WWTPs.<sup>34</sup> While not a focus of this research, it is essential to highlight that only the high-strength septage is hauled to treatment facilities. The more dilute, but still contaminated, liquid fraction of septic system effluent is released untreated to surface and groundwater resources.

The challenges of analyzing complex wastewater matrices were evident in this study, with distinct oxidation behaviors observed between PFAS precursors. FTCAs were fully oxidized during the TOP assay, whereas diPAPs showed incomplete oxidation in some septage samples (Figure 3). This incomplete oxidation is likely due to the high organic load and chemical complexity of septic effluent, which creates significant matrix interference.<sup>24</sup> The elevated organic content increases oxidation demand (Figure 1A), reducing the efficiency of the TOP assay in transforming diPAPs into terminal PFAS. These oxidative challenges were more severe than those reported by other complex matrices like landfill leachates,<sup>8</sup> as septic effluent integrates a broader range of household-derived organic pollutants that intensify matrix effects. This highlights a critical trade-off in applying the TOP assay to complex samples. Dilution can reduce matrix effects but risks lowering precursor concentrations below detection limits, while preserving matrix complexity can lead to incomplete oxidation and underestimation of PFAS potential. To overcome these challenges, pretreatment methods such as preoxidation or advanced filtration could enhance TOP assay performance.<sup>24</sup> The differing oxidation behaviors of FTCAs and diPAPs also highlight variability in precursor susceptibility to oxidative degradation, influenced by molecular structure and matrix interactions. Addressing these limitations through tailored pretreatment and analytical techniques will improve the accuracy of precursor quantification in complex systems like septic effluent, supporting more effective mitigation strategies for PFAS management.

Environmental Implications. The findings of this study underscore the critical role of residential wastewater systems, both decentralized (septic systems) and transitional (pump stations), in contributing to PFAS contamination in the environment. Septic systems, with their high retention times and organic load, act as significant reservoirs for both terminal PFAS and their precursors. The detection of diPAP precursors coupled with indications of incomplete oxidation in TOP analysis raises questions about whether septic effluent may represent a latent and underreported source of PFAS, warranting further investigation into its potential to leach into surrounding soils and groundwater. This aligns with previous reports that demonstrated the prevalence of PFAS in residential wastewater contributions to municipal systems but extends the understanding by highlighting the unique challenges posed by septic systems. The disproportionate PFAS concentrations observed in septic effluent raise concerns about localized contamination, particularly in areas with shallow aquifers or permeable soils. The anaerobic and chemically diverse conditions in septic tanks may not only preserve precursors but may also facilitate their transformation into terminal PFAS, amplifying long-term environmental risks. Groundwater contamination from decentralized systems has been documented extensively, with high PFAS levels in private wells near septic systems.<sup>35</sup> This study highlights the need for frameworks to incorporate decentralized systems into PFAS management strategies, as these systems remain largely unregulated compared to centralized treatment facilities.

Pump stations, while exhibiting lower overall PFAS concentrations, introduce episodic contamination spikes, complicating WWTP operations. Variability in household discharges affects influent PFAS loads, with precursors converting into terminal PFAS during treatment, increasing effluent and biosolid burdens. As PFAS regulations evolve, greater attention must be given to both decentralized and transitional systems in wastewater management. The detection of 27 PFAS compounds, including short-chain PFAS, underscores shifting contamination patterns. The dominance of short-chain PFAS aligns with industry transitions away from long-chain variants due to regulatory restrictions. This presents a dual challenge: addressing legacy contamination while mitigating emerging risks associated with short-chain PFAS persistence and mobility. These findings emphasize the necessity of expanding PFAS monitoring frameworks to include precursors. Conventional targeted PFAS analysis underestimates contamination, as demonstrated by the significant post-TOP increases observed. Incorporating precursor analysis will improve risk assessments and inform regulatory decisions. Addressing these challenges requires coordinated efforts to ensure effective PFAS management across decentralized, transitional, and centralized wastewater systems.

## ASSOCIATED CONTENT

## **Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.5c00246.

File S2: All raw data for this study (XLSX)

Additional details on PFAS residential sources, sampling plans, PFAS analysis, other water quality analysis, statistical analysis and extended discussion on water quality and PFAS temporal variations (PDF)

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#### Notes

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