# Linking Polycarbonate Plastics to Perfluorobutanesulfonate Emissions in Landfill Leachates via Suspect and Nontarget Screening of Diverse Plastic Products

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Cite This: https://doi.org/10.1021/acs.est.5c09017

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**ABSTRACT:** Driven by strict global regulations on legacy per- and polyfluoroalkyl substances (PFAS), short-chain alternatives such as perfluorobutanesulfonate (PFBS) are increasingly used, resulting in widespread environmental detection, notably at elevated concentrations in landfill leachates, while emission sources remain uncertain. Plastics, recognized as reservoirs of numerous chemical additives with documented but rarely investigated PFAS usage, represent a potential unidentified source. Here, 105 plastic products representing 11 types of polymers were analyzed, with 20 PFAS from 6 classes identified (confidence levels of 3 or better). Polycarbonate (PC) exhibited the highest average total PFAS concentration (713 ng/g), primarily dominated by PFBS (97%), reaching up to 6130 ng/g. PFBS, commonly co-occurred with a monohydrogen-substituted PFBS (H-PFBS), particularly enriched in PC components from electronic products, indicating intentional use as flame retardants. Consistent PFBS to H-PFBS peak area ratios observed in PC samples (~4%) and industrial PFBS products (0.07–3%), along with patent records, suggest H-PFBS as a manufacturing impurity. Simulated leaching experiments in a realistic scenario showed rapid migration of PFBS and H-PFBS into both freshwater and seawater, achieving equilibrium releases up to 39% within 14 days. Annual PFBS emissions from PC plastics to landfill leachates in China were estimated at ~40 kg, contributing ~10% of the total landfill-derived PFAS flux. These findings identify that PC plastics may represent a previously underrecognized contributor to PFBS contamination in landfill leachates, emphasizing the urgent need for safer plastic additives and enhanced waste management.

**KEYWORDS:** per- and polyfluoroalkyl substances (PFAS), perfluorobutanesulfonate (PFBS), polycarbonate, plastic additives, high-resolution mass spectrometry
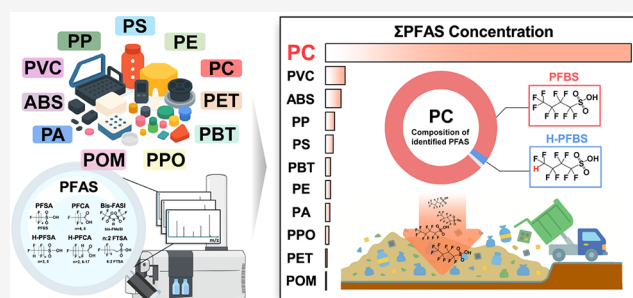
## INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) are a large group of thousands of anthropogenic organic chemicals that are either fully or partially fluorinated.[1] Due to their outstanding properties of chemical resistance and surface activity, PFAS have been widely utilized as surfactants, water/grease repellents, adhesives, and lubricants in a wide variety of products.[2−4] The increasing demand for PFAS production and usage has resulted in their widespread presence in the environment and global wildlife,[5] raising significant concerns regarding their persistence, bioaccumulation, and toxicity.[6−8] To address potential risks, two major PFAS, perfluorooctane-sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), have been gradually phased out by manufacturers due to the global regulations by governments,[9] promoting a shift from long-chain legacy PFAS to other short-chain analogues or emerging PFAS with partially hydrogenated or ether-linking chains.[10] However, many of these PFAS alternatives are now being detected more widely in the environment,[11] and their major applications remain mostly undisclosed due to industrial confidentiality, hindering the implementation of enhanced regulation and the control of emission sources.[12]

Recent studies demonstrate that landfill leachates serve as significant point sources of emerging, short-chain PFAS alternatives to surrounding aquatic environments, largely due to the high mobility and water solubility of these compounds released from discarded solid wastes.[13,14] Among them, perfluorobutanesulfonate (PFBS), a typical short-chain (C4) alternative to PFOS, has become one of the typical

characteristics of PFAS landfill contamination because of its widespread presence and abnormal levels (up to $\mu g/L$) observed in landfill leachates worldwide for years.[13] Although PFBS is commonly found in products such as carpets, leather goods, and cosmetics,[15−17] its low concentrations, along with the relatively low levels of PFBS-related precursors (e.g., perfluorobutane sulfonamide (FBSA) and its derivatives), are insufficient to account for its high abundance frequently observed in landfill leachates.[18,19] Increasing evidence of PFBS toxicity to aquatic organisms, mammals, and amphibians,[20−22] has drawn significant regulatory attention and led to its designation as a substance of very high concern (SVHC) by the European Chemicals Agency (ECHA) in 2020.[23] Moreover, its frequent detection in human breast milk, with rising concentrations over recent decades, raises further concerns about exposure and health risks.[24] Therefore, identifying the unknown emission sources of PFBS in landfill leachates is of great significance for achieving efficient regulation and risk mitigation.

Plastic products are complex chemical mixtures composed not only of the basic polymer matrix but also numerous chemical additives intentionally introduced for achieving specific functions, with more than 10000 identified chemicals to date.[25] Previous studies have shown that many of these additives are not covalently bound to the polymer during manufacturing,[26] facilitating their migration and subsequent release into the surrounding environment following end-of-life disposal. Landfills, as one of the most common waste disposal methods, are the final destination of most plastic materials, as well as the sink for those migratory additives. Meanwhile, improper disposal of plastics can also lead to direct environmental contamination. For example, phthalates, widely used plasticizers and known endocrine disruptors, are frequently found in landfill leachates and surface waters, primarily due to the chemicals' leaching from deteriorating discarded plastic products.[27,28] Given the widespread use of plastics and the substantial volume of associated waste, the high mobility of these additives positions plastics as an important yet often underrecognized potential contributor to environmental contaminant loads, including chemical pollution in landfill leachates.

Although plastics are widely recognized reservoirs of synthetic chemicals, the actual presence of PFAS in general plastic products has not been systematically investigated. In fact, PFAS have been extensively documented in patent records as functional additives during plastic manufacturing processes, serving critical roles. For instance, ammonium perfluorocarboxylate was patented for mold release, methyl perfluoroalkyl ether for foam blowing, and lithium perfluoroalkanesulfonate for static control.[3] However, uncertainties regarding PFAS in plastics still exist, as not all registered patents are implemented in the market. Several studies on PFAS in food-contact materials have indirectly revealed the presence of PFAS such as perfluorocarboxylates, fluorotelomer phosphate diester, and fluorotelomer alcohol (with maximum reported concentrations up to 51.9 ng/g) in plastics by including plastic packages in their sample selections, but the insufficient sample sizes fail to adequately represent the diversity of plastic usage and the various polymer categories.[29−31]

Therefore, to address the insufficient recognition of PFAS in plastics and prevent the potential threats posed by plastics as potential sources of PFAS emissions to environments,

particularly regarding their potential contribution to the special PFAS pattern in landfill leachates (e.g., abnormal levels of PFBS), this study mainly focuses on: (1) comprehensively screening PFAS from currently available plastic products across the most common polymer categories via suspect and nontarget screening strategies using high-resolution mass spectrometry (HRMS); (2) investigating the migration potential of the predominant PFAS identified in plastics; and (3) exploring a potential linkage to those previously unexplained PFAS detections in relation to landfill leachates. This work advances the understanding of a specific plastic type (i.e., polycarbonate) as a previously underrecognized potential contributor to PFBS contamination, particularly in landfill environments, and provides an empirical basis to inform safer plastics regulatory frameworks and promote waste management for mitigating PFAS contamination.

## ■ MATERIALS AND METHODS

**Chemicals and Reagents.** The detailed information on native and mass-labeled PFAS standards is summarized in Tables S1 and S2, including full names, related abbreviations, formulas, CAS numbers, and vendors. All solvents and reagents were purchased from Thermo Fisher Scientific (U.S.A.) with purities of HPLC or higher.

**Sample Collection and Pretreatment.** A total of 105 commercially available plastic products were selected, representing two distinct categories: (1) daily plastic products, in which plastic serves as the primary structural material (e.g., plastic packaging and clothes hangers); and (2) plastic components integrated into electronic products (e.g., the outer plastic casing of wall switches and keyboard caps). All samples were purchased from local and online stores in China in August 2024. The chosen plastic products represented 11 types of the most common plastic polymer: polypropylene (PP, $n = 12$), polystyrene (PS, $n = 9$), polyvinyl chloride (PVC, $n = 16$), polyethylene (PE, $n = 9$), acrylonitrile butadiene styrene (ABS, $n = 12$), polycarbonate (PC, $n = 17$), polyamide (PA, $n = 7$), polyethylene terephthalate (PET, $n = 7$), polybutylene terephthalate (PBT, $n = 6$), polyoxymethylene (POM, $n = 5$), and polyphenylene oxide (PPO, $n = 5$). The detailed information on the plastics analyzed in the current study is listed in Table S3. The sample preparation was conducted immediately in a hood after opening the original commercial packaging. The remaining portions of the plastic products were carefully wrapped in aluminum foil and sealed in clean polypropylene bags to prevent environmental contamination, then stored at room temperature for future use.

The pretreatment method applied in this study was based on the established PFAS extraction practices for polymeric matrices by USEPA.[32] Briefly, each sample was cut into 0.5 × 0.5 cm pieces before analysis. 100 mg of each sample was weighed in a 15 mL polypropylene tube, spiked with surrogate standards, and then extracted using 4 mL of methanol by ultrasonication at room temperature for 1 h. The methanol-extractable fraction typically represents the readily extractable (migratable) PFAS fraction rather than the total PFAS burden, reflecting the subset of compounds with greater environmental mobility, as these chemicals are more readily mobilized by a mild solvent.[31,33] The extraction was repeated three times, and the supernatants were combined to be further concentrated to ~1 mL by nitrogen blow, filtered through a 0.22 $\mu$m syringe filter, and spiked with internal standards before instrumental analysis. For industrial PFBS product analysis, 100 mg of each

product was dissolved in 100 mL of methanol to prepare stock solutions, which were then diluted to 100 ng/mL with methanol and spiked with internal standards prior to instrumental analysis.

**Simulated Leaching Experiments.** Six PC plastic samples with the highest total PFAS concentrations were selected for leaching simulation experiments. A modified leaching test, adapted from the EN 12457 standard series recommended by the European Union's Landfill Directive (1999/31/EC),[34] was employed to simulate aqueous contact of landfilled plastics under meteoric water (rainfall/percolation) conditions, using distilled water to assess the compliance of waste prior to landfill acceptance. The short-time agitation was replaced with a prolonged static immersion period for kinetics modeling,[35] and the solid to liquid ratio was reduced to 1/20. In parallel, artificial seawater was also employed to simulate the most conservative release potential into the marine environment, which was prepared based on a previously reported formula containing 26.73 g/L of sodium chloride, 0.72 g/L of potassium chloride, 1.15 g/L of calcium chloride, 2.26 g/L of magnesium chloride, and 3.25 g/L of magnesium sulfate with a final pH value adjusted to 8.1.[36] Briefly, 1 g of each PC sample was weighed in a 50 mL polypropylene tube filled with 20 mL of distilled water or artificial seawater. To generate an environmentally relevant leaching profile that reflects integrated natural day—night conditions, the tubes were then capped to avoid airborne contamination and exposed on a rooftop in Shenzhen, China (22°36′N, 113°592032E) to introduce thermal changes by natural solar exposure for a period of 14 days (December 4 to 18, 2024). After 6, 12, 24, 48, 96, 168, and 336 h, 0.25 mL of the distilled water or seawater leachate was collected and centrifuged at 10,000 rpm for 5 min, and 0.2 mL of the resulting supernatant was spiked with internal standards. During instrumental analysis, the LC flow was diverted to waste for the first 2 min to remove early eluting salts and prevent ion-source contamination.[37] Over the course of the experiment, no rainfall was observed, and the average outdoor temperature was 17 °C (range from 12.2 to 20.6 °C). The water temperature and ultraviolet index were recorded regularly on the sampling day for monitoring the environmental changes throughout the daytime (more details are summarized in Table S4). All samples were analyzed in duplicate. Leaching blanks (20 mL of either distilled water or artificial seawater without plastic samples) were prepared in parallel using the same polypropylene tubes under identical experimental conditions to monitor potential background PFAS contamination. No PFAS were detected in blank samples throughout the experiment. The release of PFAS from the selected plastic products was then examined by the pseudo-first-order kinetic model as follows:[35]

$$Q_t = Q_e(1 - e^{-k_1 t})$$

where $Q_t$ represents the released chemical amount (ng/g) at time $t$ (h), $Q_e$ is the equilibrium released amount (ng/g), and $k_1$ stands for the release rate constant (h$^{-1}$).

**Instrumental Analysis.** Chemical analysis was conducted by a 1290 Infinity ultrahigh-performance liquid chromatograph system coupled with a 6546 quadrupole time-of-flight high-resolution mass spectrometer (UPLC-QTOF-MS, Agilent, U.S.A.). Detailed chromatographic conditions, MS source

settings, and data acquisition parameters are provided in the Supporting Information (Text S1).

**Suspect and Nontarget Screening of PFAS.** A workflow integrating full scan, iterative DDA, and target DDA was performed in this study to enhance the screening efficiency and to expand the MS$^2$ coverage of ions at various abundance levels (Figure S1). Briefly, the screening process can be summarized into 4 steps: (i) The raw data files from iterative DDA were processed by the open-source software MS-Dial (version 5.3) for peak picking and feature identification using its online MSP spectral library of PFAS, which includes 6936 PFAS MS$^2$ records (ESI-).[38] The matched features with MS$^2$ error ≤ 0.005 Da and MS$^2$ fragments matched ≥ 2 were assigned as identified PFAS candidates. The matched features with only MS$^1$ error ≤ 0.001 Da but with no MS$^2$ spectra acquired were classified into a preidentified PFAS list; (ii) The raw data files of the full scan were processed by the function of Find by Formula (FBF) from Agilent MassHunter Qualitative Analysis software (version 10.0) to match with an emerging PFAS list collected from recent research.[39] Only features demonstrating a match score of ≥ 70 for monoisotopic exact mass, theoretical isotope distribution, isotope spacing consistency (mass accuracy ≤ 5 ppm),[40] and without being identified in prior steps, were retained in the preidentified PFAS list; (iii) MS$^2$ spectra of the suspect features in the preidentified PFAS list were further acquired using target DDA and being analyzed manually for checking characteristic fragment ions, retention time and peak shape by Agilent Masshunter Qualitative Analysis 10.0; (iv) Commercially available standards were then applied to confirm the PFAS candidates in terms of fragmentation pattern and retention time. The annotation rules by Charbonnet et al. were employed to assign confidence levels of identification and only candidates with levels of 3 or better were marked as identified PFAS.[41]

**(Semi-)Quantification of Identified PFAS in Plastic Samples.** The identified PFAS with reference standards (Level 1) were quantified by the full scan mode of UPLC-QTOF-MS using calibration curves with concentrations ranging from 0.1 to 100 ng/mL. PFAS without commercial standards (identification Levels 2−3) were semiquantified using calibration curves of structurally analogous PFAS, following established approaches widely applied in recent PFAS nontarget screening studies, which represent order-of-magnitude estimates (details summarized in Table S5).[42−44] All calibration curves demonstrated good linear correlation ($R^2$ > 0.99) and samples exceeding the upper quantification limits were subjected to systematic dilution until analyte concentrations fell within the established calibration ranges.

**Quality Control and Quality Assurance (QA/QC).** One quality control (QC) sample, prepared by mixing one randomly selected sample from each polymer type, was routinely included in every batch of 20 samples. The instrumental stability was then monitored by evaluating the retention time and peak intensity consistency of a representative ion feature from each QC run throughout the analytical batch. Procedure blanks for plastic extraction ($n = 8$), prepared by performing the complete sample preparation procedure without plastic samples, and matrix-spiked samples ($n = 4$) were included during the instrumental analysis to monitor background contamination and evaluate method accuracy. In the process of PFAS screening, all detected feature peaks were sorted according to the following criteria to avoid background interference: (1) signal-to-noise (S/N) ratio

**Table 1. Per- and Polyfluoroalkyl Substances (PFAS) Identified in Plastic Products**

| Class | ID | Annotation | Formula | Theoretical $m/z$ [M-H] | Observed $m/z$ [M-H] | RT/min | Confidence Level[a] |
|---|---|---|---|---|---|---|---|
| PFSA | 1 | PFBS | $C_4HF_9O_3S$ | 298.9430 | 298.9432 | 10.688 | 1a |
| PFCA | 2 | PFBA | $C_4HF_7O_2$ | 212.9792 | 212.9789 | 7.075 | 1a |
| | 3 | PFOA | $C_8HF_{15}O_2$ | 412.9664 | 412.9662 | 13.446 | 1a |
| H-PFSA | 4 | H-PFBS | $C_4H_2F_8O_3S$ | 280.9524 | 280.9526 | 8.364 | 2b |
| | 5 | H-PFHxS | $C_6H_2F_{12}O_3S$ | 380.9460 | 380.9461 | 10.629 | 3a |
| H-PFCA | 6 | H-PFBA | $C_4H_2F_6O_2$ | 194.9886 | 194.9887 | 6.231 | 3a |
| | 7 | H-PFOA | $C_8H_2F_{14}O_2$ | 394.9759 | 394.9754 | 12.036 | 2a |
| | 8 | H-PFNA | $C_9H_2F_{16}O_2$ | 444.9727 | 444.9728 | 12.748 | 3a |
| | 9 | H-PFDA | $C_{10}H_2F_{18}O_2$ | 494.9695 | 494.9693 | 13.320 | 3a |
| | 10 | H-PFUdA | $C_{11}H_2F_{20}O_2$ | 544.9663 | 544.9672 | 13.863 | 2a |
| | 11 | H-PFDoA | $C_{12}H_2F_{22}O_2$ | 594.9631 | 594.9635 | 14.267 | 2a |
| | 12 | H-PFTrDA | $C_{13}H_2F_{24}O_2$ | 644.9599 | 644.9603 | 14.614 | 2a |
| | 13 | H-PFTeDA | $C_{14}H_2F_{26}O_2$ | 694.9567 | 694.9567 | 14.928 | 2a |
| | 14 | H-PFPeDA | $C_{15}H_2F_{28}O_2$ | 744.9535 | 744.9524 | 15.203 | 2a |
| | 15 | H-PFHxDA | $C_{16}H_2F_{30}O_2$ | 794.9503 | 794.9506 | 15.438 | 2a |
| | 16 | H-PFHpDA | $C_{17}H_2F_{32}O_2$ | 844.9471 | 844.9458 | 15.646 | 2a |
| | 17 | H-PFODA | $C_{18}H_2F_{34}O_2$ | 894.9439 | 894.9443 | 15.808 | 2a |
| | 18 | H-PFNDA | $C_{19}H_2F_{36}O_2$ | 944.9407 | 944.9422 | 15.988 | 3a |
| $n$:2 FTSA | 19 | 6:2 FTSA | $C_8H_5F_{13}O_3S$ | 426.9679 | 426.9669 | 13.370 | 1a |
| Bis-FASI | 20 | Bis-FMeSI | $C_2HF_6NO_4S_2$ | 279.9178 | 279.9176 | 8.795 | 1a |

[a]Confidence level criteria adapted from Charbonnet et al.[41]

> 3, (2) ratio of sample intensity to procedural blank > 3, and (3) peak area > $10^4$, in line with previous nontargeted screening workflows.[42] All quantified PFAS concentrations in samples were corrected by the procedural blank subtraction. The absolute recoveries of the target analytes ranged from 66% to 133%. The recoveries of surrogate standards ranged from 59 ± 2% to 127 ± 2% (mean ± standard errors). For analytes not detected in procedural blanks, method detection limits (MDLs) were determined based on the lowest concentration in the calibration curve that produced an S/N ratio of approximately 3. The S/N was calculated using Agilent MassHunter Qualitative Analysis software (version 10.0), defined as the peak height divided by the measurable peak-to-peak noise baseline in the chromatogram with a ± 10 ppm mass window. For target analytes detected in procedural blanks, MDLs were calculated as three times the standard deviation of the detected levels. The details of MDLs, procedural blanks, matrix spike recoveries, and surrogate recoveries were summarized in Tables S5−S7.

**Data Analysis.** The estimated amount of PFBS that can be released to landfills from disposed PC plastic products related to electronics was calculated using the following formula:

$$M_{\text{PFBS-released}} = M_{\text{e-waste}} \times P_{\text{plastic}} \times P_{\text{PC}} \times P_{\text{landfill}} \times C_{\text{PFBS}}$$
$$\times P_{\text{migration}}$$

where $M_{\text{PFBS-released}}$ (g) represents the estimated amount of PFBS released; $M_{\text{e-waste}}$ (10.1 Mt/y) represents the annual production amount of e-waste in 2019, China;[45] $P_{\text{plastic}}$ (21%) represents the proportion of plastic components in e-waste;[46] $P_{\text{PC}}$ (10%) represents the proportion of PC plastic among all plastic components in e-waste;[47] $P_{\text{landfill}}$ (80%) is the proportion of e-waste disposed in landfills;[48] $C_{\text{PFBS}}$ (1070 ng/g) stands for the average concentration of PFBS in PC products of electronic usages in this study; $P_{\text{migration}}$ (22%) represents the average proportion of PFBS released to distilled water observed in this study.

The differences of each identified PFAS among different types of plastic were presented in terms of detection frequencies and average values ± standard errors. Only PFAS with detection frequencies ≥50% were included in the statistical analysis, and concentrations of PFAS below MDLs were imputed with MDL/2. The Mann−Whitney was used to perform the comparative analysis for PFAS concentrations in plastic samples with different applications after the normality of the data was tested by the Shapiro-Wilk test. Microsoft Excel was used to perform descriptive statistics, and figures were generated using Origin, Adobe Illustrator, and R Studio.

## ■ RESULTS AND DISCUSSION

**Identification of PFAS in Plastic Samples.** Overall, a total of 20 PFAS were identified across 11 types of plastics (Table 1) with confidence levels ≥ 3, among which 5 were further confirmed by authentic standards. The identified PFAS were further classified into 6 classes based on molecular substituents and terminal groups (Figure 1A), including perfluoroalkyl sulfonate (PFSA), perfluoroalkyl carboxylate (PFCA), monohydrogen-substituted polyfluoroalkyl sulfonate (H-PFSA), monohydrogen-substituted perfluoroalkyl carboxylate (H-PFCA), $n$:2 fluorinated telomer sulfonate ($n$:2 FTSA), and bis-perfluoroalkyl sulfonimide (bis-FASI).

*PFSA, PFCA, and n:2 FTSA.* PFBS, PFBA and PFOA, and 6:2 FTSA were first annotated via suspect screening by matching the data obtained from iterative DDA with the online PFAS spectral library connected to MS-Dial. Then their identities were further confirmed as Level 1a by comparing retention times, differences of exact masses, and fragments from $MS^2$ spectra with authentic standards. The relevant $MS^2$ spectra of these PFAS are provided in Figures S2−S4.

*H-PFCA.* Thirteen $MS^1$ features were filtered from the full scan data by matching the general formula of H-PFCA homologues (C4, C8−C19) with mass errors less than 5 ppm, and exhibiting increasing retention times (RTs) correlating with carbon chain lengths. Subsequently, target DDA analyses were employed to acquire their $MS^2$ spectra for manual
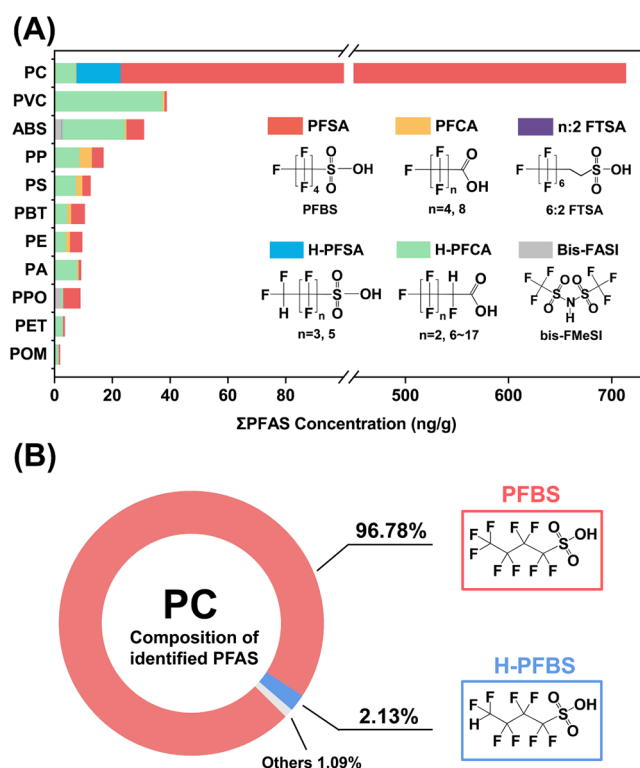
**Figure 1.** (A) Average concentrations of each per- and polyfluoroalkyl substance (PFAS) class detected in different types of plastic samples. (B) Composition of all identified PFAS in polycarbonate (PC).

interpretation (Figure S5). Characteristic fragment ions of $C_2F_5^-$ and $C_3F_7^-$, observed across $MS^2$ spectra, indicated cleavage of the fluorocarbon chain, suggesting the hydrogen substituent was positioned distally from the perfluoroalkyl end. Notably, highly abundant $[M-64]^-$ fragments were detected consistently among most H-PFCAs, corresponding to the neutral losses of $CO_2$ (44 Da) and HF (20 Da). This specific fragmentation pattern strongly implied the substitution of hydrogen at the $\alpha$-position within their molecular structures.[42] In contrast, for the C9 homologue (H-PFNA), the observed fragment at $m/z$ 330.9802 ($C_7F_{13}^-$, 1.51 ppm) with relative abundance of 100%, resulting from a successive loss of $CO_2-CF_2-HF$ (114 Da) from the precursor ion at $m/z$ 444.9728 ($C_9HF_{16}O_2^-$, 0.44 ppm), suggested instead a $\beta$-position hydrogen substitution.[42] These fragmentation patterns observed for the H-PFCA homologues closely matched those reported. Consequently, several homologues were identified at Level 2a by matching with $MS^2$ spectra from published references,[42,49–51] while the remaining ones were assigned to Level 3a due to the absence of available spectral references.

*H-PFSA.* Two H-PFSA homologues, including H-PFBS and H-PFHxS, were identified based on the workflow described for the H-PFCA homologues. The initial identification was supported by the diagnostic fragments associated with sulfonate groups $SO_3^-$ and $FSO_3^-$, clearly observed in the DDA-acquired $MS^2$ spectra of precursor ions at $m/z$ 280.9526 ($C_4HF_8SO_3^-$, 0.71 ppm) and $m/z$ 380.9461 ($C_6HF_{12}SO_3^-$, 0.26 ppm), respectively (Figure S6). For the C4 homologue (H-PFBS), the detected fragment at $m/z$ 180.9899 ($C_4F_7^-$, 3.31 ppm) originated from the sequential neutral losses of HF and $SO_3$, confirming the presence of hydrogen. The observed

fragment at $m/z$ 130.9928 ($C_3F_5^-$, 2.29 ppm) further indicated that the hydrogen substitution occurred at one of the three terminal carbons of the alkyl chain. Similarly, for the C6 homologue, the hydrogen substitution was supported by the detected fragment at $m/z$ 230.9869 ($C_5F_9^-$, 3.03 ppm), along with the recurring $C_3F_5^-$ diagnostic fragment. However, due to insufficient $MS^2$ spectral evidence to confirm the exact position of the substituted hydrogen, H-PFHxS was assigned to Level 3a, while H-PFBS was annotated as Level 2b due to the structural support from a reaction pathway patented previously.[52]

*Bis-FASI.* A short-chain homologue of bis-FASI, bis-trifluoromethane sulfonimide (bis-FMeSI, also known as NTf2 and bistriflimide), was tentatively identified at the RT 8.80 min by matching its precursor ion at $m/z$ 279.9176 ($C_2F_6S_2O_4N^-$, −0.71 ppm). Three major fragments were detected at $m/z$ 77.9653 ($SO_2N^-$, −2.56 ppm), 146.9603 ($CF_3SO_2N^-$, −2.72 ppm), and 210.9220 ($CF_3S_2O_4N^-$, −2.84 ppm), with relative abundances of 18%, 100%, and 14%, respectively (Figure S7). These fragments strongly supported the chemical structure of bis-FMeSI, featuring one central nitrogen atom bonded with two trifluoromethanesulfonyl groups.[53] The identification of bis-FMeSI was further confirmed at Level 1a by matching both the RT and fragmentation pattern to those of a commercially available authentic standard. It is worth noting that this emerging PFAS has been widely used as an electrolyte and antistatic agent, owing to its chemical stability and electrical conductivity, particularly in the production of lithium-ion batteries.[54] The continuous industrial demand and applications have led to its wide presence in various environments, including sediments, surface water, and tap water.[54–56] Nonetheless, this is the first report on its identification in plastic products.

**PFAS Concentrations in Plastics.** Concentrations and detection frequencies (DFs) of the identified 20 PFAS in plastic products are summarized in Table 2. At least one PFAS was found in each type of sample. Four PFAS, including PFBS, H-PFDoA, H-PFTrDA, and H-PFTeDA, were detected in more than half of the samples. For the rest of PFAS compounds, low detection frequencies (6−46%) were observed among all plastic samples. Among 11 types of plastic, PC and PVC were found with the most identified PFAS (18 out of 20), with PFAS detection frequencies ranging from 6−94% and 6−100%, respectively, followed by PP, PS, and PA (16 out of 20). The highest average total PFAS concentrations (mean $\sum$PFAS concentrations) were observed for PC at 713 ng/g (Figure 1A), more than 10 times higher than other polymer types of plastic products, followed by PVC (38.7 ng/g), ABS (30.9 ng/g), and PP (16.7 ng/g).

It is worth noting that PFBS was the predominant PFAS detected in PC plastic samples, contributing around 97% of the total average PFAS concentration (Figure 1B), and was found in all types of plastic, with detection frequencies varying from 44% to 100%. The average concentration of PFBS in PC samples was 690 ng/g, while in other types of plastic it varied from 0.31 to 5.83 ng/g, which motivates us to investigate the specific usage of PFBS in PC plastics. However, there are few reports on the occurrence of PFBS in plastics before. A 2023 study analyzing 18 plastic food packaging samples found the level of PFBS reached 26.6 ng/g in one sample with polymer type undisclosed. The frequent detection of PFBS in food-contact materials has been attributed to its chemical stability and surface activity, which provide water/oil repellency and

**Table 2. Detection Frequencies (DF, %), and Average Concentrations (ng/g) ± Standard Error (SE) of Identified Per- and Polyfluoroalkyl Substances (PFAS) in Plastic Samples of 11 Polymer Types**

| Compounds | PP DF | PP Mean ± SE | PS DF | PS Mean ± SE | PVC DF | PVC Mean ± SE | PE DF | PE Mean ± SE | ABS DF | ABS Mean ± SE | PC DF | PC Mean ± SE | PA DF | PA Mean ± SE | PET DF | PET Mean ± SE | PBT DF | PBT Mean ± SE | POM DF | POM Mean ± SE | PPO DF | PPO Mean ± SE |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| PFBS | 75 | 3.78 ± 2.31 | 44 | 2.62 ± 1.22 | 69 | 0.55 ± 0.20 | 67 | 4.04 ± 3.54 | 50 | 5.83 ± 2.80 | 94 | 690 ± 367 | 100 | 0.69 ± 0.15 | 86 | 0.35 ± 0.19 | 67 | 4.53 ± 4.30 | 60 | 0.31 ± 0.20 | 60 | 5.74 ± 5.10 |
| PFBA | 8 | 4.07 ± 3.99 | 11 | 0.53 ± 0.45 | 19 | 0.44 ± 0.21 | 11 | 1.31 ± 1.23 | 17 | 0.53 ± 0.32 | 6 | 0.14 ± 0.06 | 29 | 0.34 ± 0.19 | 0 | <MDL | 17 | 1.40 ± 1.32 | 0 | <MDL | 0 | <MDL |
| PFOA | 92 | 0.27 ± 0.07 | 44 | 1.64 ± 1.51 | 50 | 0.12 ± 0.02 | 22 | 0.07 ± 0.02 | 17 | 0.07 ± 0.01 | 18 | 0.07 ± 0.02 | 14 | 0.07 ± 0.03 | 0 | <MDL | 0 | <MDL | 0 | <MDL | 0 | <MDL |
| H-PFBS[a] | 8 | 0.03 ± 0.01 | 0 | <MDL | 0 | <MDL | 11 | 0.03 ± 0.01 | 17 | 0.03 ± 0.01 | 41 | 15.2 ± 12.4 | 0 | <MDL | 0 | <MDL | 0 | <MDL | 0 | <MDL | 0 | <MDL |
| H-PFHxS[a] | 0 | <MDL | 0 | <MDL | 0 | <MDL | 0 | <MDL | 0 | <MDL | 35 | 0.15 ± 0.08 | 0 | <MDL | 0 | <MDL | 0 | <MDL | 0 | <MDL | 0 | <MDL |
| H-PFBA[a] | 0 | <MDL | 0 | <MDL | 6 | 18.8 ± 18.7 | 0 | <MDL | 83 | 17.8 ± 5.55 | 12 | 1.85 ± 1.24 | 0 | <MDL | 0 | <MDL | 17 | 1.04 ± 0.96 | 0 | <MDL | 0 | <MDL |
| H-PFOA[a] | 75 | 0.33 ± 0.11 | 44 | 0.13 ± 0.04 | 56 | 0.29 ± 0.08 | 0 | <MDL | 17 | 0.09 ± 0.03 | 29 | 0.14 ± 0.04 | 43 | 0.23 ± 0.13 | 29 | 0.15 ± 0.07 | 100 | 0.52 ± 0.12 | 0 | <MDL | 20 | 0.10 ± 0.05 |
| H-PFNA[a] | 25 | 0.26 ± 0.07 | 11 | 0.22 ± 0.07 | 38 | 0.40 ± 0.13 | 0 | <MDL | 0 | <MDL | 12 | 0.26 ± 0.10 | 14 | 0.30 ± 0.15 | 29 | 0.25 ± 0.07 | 17 | 0.19 ± 0.04 | 0 | <MDL | 0 | <MDL |
| H-PFDA[a] | 8 | 0.02 ± 0.01 | 22 | 0.07 ± 0.03 | 6 | 0.03 ± 0.01 | 0 | <MDL | 0 | <MDL | 18 | 0.11 ± 0.06 | 14 | 0.13 ± 0.11 | 43 | 0.23 ± 0.12 | 17 | 0.05 ± 0.03 | 100 | 0.59 ± 0.18 | 20 | 0.06 ± 0.04 |
| H-PFUdA[a] | 0 | <MDL | 11 | 0.18 ± 0.15 | 44 | 7.03 ± 4.52 | 0 | <MDL | 0 | <MDL | 24 | 0.32 ± 0.16 | 14 | 0.58 ± 0.55 | 29 | 0.23 ± 0.16 | 67 | 0.78 ± 0.37 | 20 | 0.05 ± 0.03 | 20 | 0.13 ± 0.11 |
| H-PFDoA[a] | 100 | 5.07 ± 0.47 | 100 | 3.49 ± 0.56 | 100 | 5.42 ± 1.28 | 44 | 0.81 ± 0.36 | 75 | 1.05 ± 0.31 | 47 | 1.47 ± 0.62 | 71 | 2.19 ± 2.02 | 57 | 0.66 ± 0.32 | 67 | 0.64 ± 0.29 | 40 | 0.45 ± 0.27 | 20 | 0.11 ± 0.08 |
| H-PFTrDA[a] | 100 | 1.90 ± 0.19 | 89 | 2.69 ± 0.81 | 88 | 4.54 ± 1.06 | 100 | 1.80 ± 0.56 | 100 | 2.28 ± 0.45 | 88 | 2.22 ± 0.82 | 71 | 3.30 ± 2.42 | 71 | 1.24 ± 0.45 | 67 | 0.92 ± 0.46 | 40 | 0.25 ± 0.16 | 40 | 0.22 ± 0.17 |
| H-PFTeDA[a] | 75 | 0.30 ± 0.06 | 56 | 0.25 ± 0.14 | 94 | 0.57 ± 0.14 | 44 | 1.16 ± 0.98 | 100 | 0.44 ± 0.07 | 88 | 0.72 ± 0.37 | 71 | 0.82 ± 0.64 | 29 | 0.11 ± 0.06 | 50 | 0.19 ± 0.09 | 40 | 0.07 ± 0.04 | 40 | 0.14 ± 0.07 |
| H-PFPeDA[a] | 100 | 0.29 ± 0.04 | 100 | 0.14 ± 0.02 | 81 | 0.16 ± 0.05 | 33 | 0.03 ± 0.01 | 0 | <MDL | 24 | 0.06 ± 0.03 | 43 | 0.14 ± 0.10 | 29 | 0.05 ± 0.03 | 33 | 0.05 ± 0.02 | 0 | <MDL | 0 | <MDL |
| H-PFHxDA[a] | 92 | 0.10 ± 0.02 | 56 | 0.07 ± 0.02 | 25 | 0.05 ± 0.02 | 0 | <MDL | 0 | <MDL | 0 | <MDL | 14 | 0.06 ± 0.05 | 0 | <MDL | 17 | 0.02 ± 0.01 | 0 | <MDL | 0 | <MDL |
| H-PFHpDA[a] | 50 | 0.04 ± 0.01 | 11 | 0.02 ± 0.01 | 13 | 0.02 ± 0.01 | 0 | <MDL | 0 | <MDL | 6 | 0.03 ± 0.01 | 14 | 0.11 ± 0.10 | 0 | <MDL | 17 | 0.02 ± 0.01 | 0 | <MDL | 0 | <MDL |
| H-PFODA[a] | 83 | 0.09 ± 0.02 | 67 | 0.04 ± 0.01 | 50 | 0.07 ± 0.03 | 0 | <MDL | 0 | <MDL | 6 | 0.01 ± 0.01 | 14 | 0.04 ± 0.03 | 14 | 0.01 ± 0.01 | 17 | 0.01 ± 0.01 | 0 | <MDL | 20 | 0.01 ± 0.01 |
| H-PFNDA[a] | 92 | 0.11 ± 0.02 | 67 | 0.05 ± 0.01 | 38 | 0.05 ± 0.02 | 0 | <MDL | 0 | <MDL | 0 | <MDL | 14 | 0.03 ± 0.02 | 0 | <MDL | 0 | <MDL | 40 | 0.02 ± 0.01 | 20 | 0.02 ± 0.01 |
| 6:2 FTSA | 25 | 0.19 ± 0.08 | 11 | 0.26 ± 0.21 | 31 | 0.12 ± 0.03 | 22 | 0.12 ± 0.05 | 42 | 0.14 ± 0.04 | 47 | 0.17 ± 0.04 | 0 | <MDL | 14 | 0.08 ± 0.03 | 0 | <MDL | 20 | 0.11 ± 0.06 | 0 | <MDL |
| bis-FMeSI | 0 | <MDL | 0 | <MDL | 6 | 0.05 ± 0.01 | 22 | 0.17 ± 0.12 | 25 | 2.60 ± 1.72 | 12 | 0.07 ± 0.02 | 14 | 0.10 ± 0.06 | 14 | 0.05 ± 0.01 | 17 | 0.07 ± 0.02 | 0 | <MDL | 20 | 2.34 ± 2.30 |
| ΣPFAS | 100 | 16.7 ± 0.30 | 100 | 12.4 ± 0.16 | 100 | 38.7 ± 1.58 | 100 | 9.55 ± 0.39 | 100 | 30.9 ± 0.72 | 100 | 713 ± 21.8 | 100 | 9.14 ± 0.21 | 100 | 3.42 ± 0.06 | 100 | 10.4 ± 0.31 | 100 | 1.86 ± 0.06 | 60 | 8.88 ± 0.56 |

[a]Semi-quantifications based on their respective calibration curves of similar chemical structure analogues were applied due to their unavailable commercial standards.
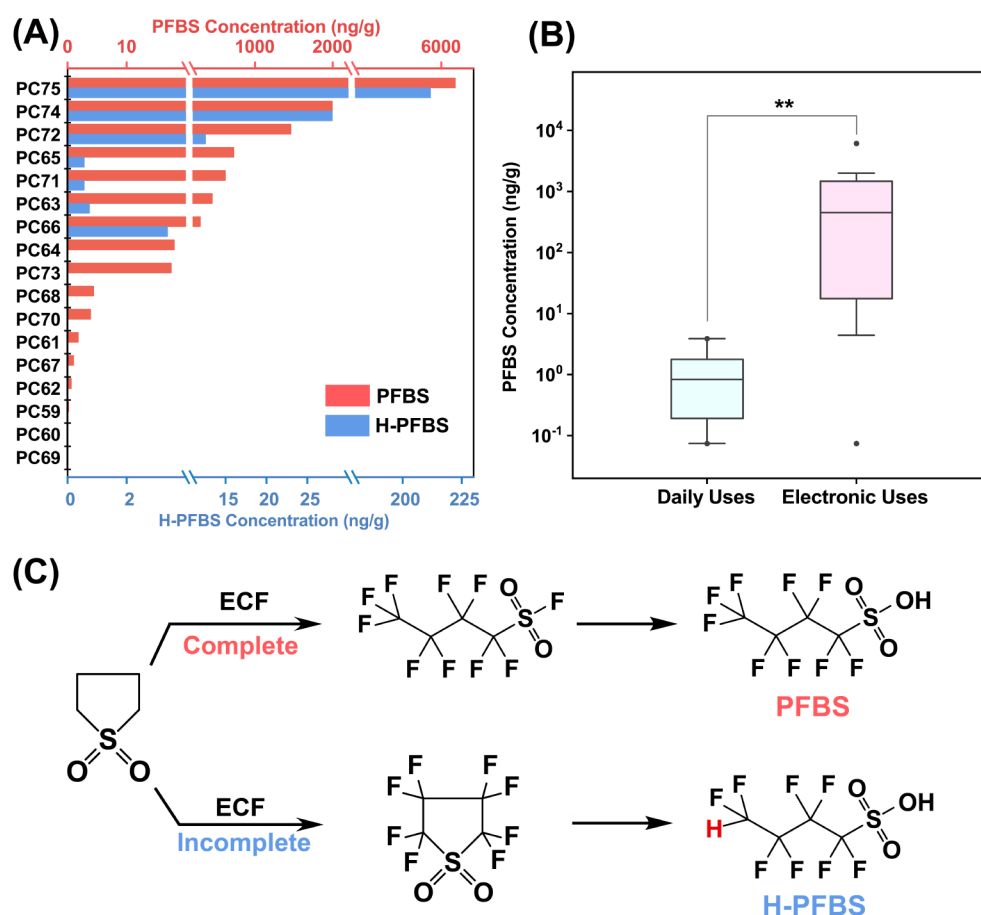
F

**Figure 2.** (A) Concentrations of PFBS and H-PFBS in all PC samples. (B) Concentrations of PFBS from PC plastic products intended for daily uses and electronic uses. Concentrations are shown as boxplots, representing the 25th and 75th percentiles; black lines represent the median, and the whiskers represent the 10th and 90th percentiles. The asterisk indicates a statistical difference at the $p < 0.01$ level based on a Mann−Whitney test. (C) Synthesis pathway of H-PFBS reconstructed from the patent record.[52] ECF represents electrochemical fluorination.

improved product durability.[57,58] These functional benefits suggest a possible intentional use of PFBS in plastic formulations. Additionally, given that no PFBS precursors (e.g., C4-based perfluoroalkyl sulfonamides and derivatives) were detected in the plastic samples, the observed PFBS levels are considered to result from its direct presence in plastic products.

To implement the insufficient academic research on the function of PFBS in plastics, we searched its usage in the publicized patent database and found PFBS plays a crucial role in imparting heat resistance as a flame retardant for resin production (including PC and PET).[59,60] Additionally, a 1997 patent recorded PFBS as an essential ingredient in producing antistatic agents used in plastic resins (including PET, ABS, PC, and PS),[61] while a 2012 patent documented its application as the raw material for the catalyst used in the synthesis of ultrahigh molecular weight PE.[62] Compared to other plastic polymers, PC, in particular, is widely used as a high-performance engineering plastic due to its excellent heat resistance, impact strength, and dimensional stability. These features are often enhanced through the incorporation of functional additives. Therefore, the presence of PFBS in PC is probably linked to its intentional addition to enhance flame retardancy, antistatic properties, and overall durability.

Interestingly, H-PFBS, a hydrogen-substituted PFBS, was found in PC with a relatively high detection frequency (41%) and an average concentration of 15.2 ng/g (Figure 1B), both

notably higher than those in other plastic types. However, unlike other commonly detected PFAS that are widespread in the natural environment, H-PFBS has been primarily observed in specific scenarios influenced by human activities, often co-occurring with PFBS. For example, H-PFBS was once identified along with PFBS in contaminated groundwater impacted by historical aqueous film-forming foams.[63] In the surrounding river stream of a fluorine-chemical manufacturer in China, H-PFBS and PFBS were detected, with both the maximum concentrations exceeding 1 $\mu$g/L.[64] The co-occurrence of PFBS and H-PFBS in PC plastics, consistent with previous observations, suggests that H-PFBS may originate as a byproduct or degradation product of PFBS during the manufacturing activity of PFBS-related products.

PFOA and PFBA are the two major PFCAs detected in plastic products. They were detected in 30% and 11% of the samples, respectively, and with relatively low average concentrations of 0.230 and 0.889 ng/g, respectively. PFOA has been widely applied as an effective mold release agent in manufacturing plastics due to its oleophobic and hydrophobic nature.[3] However, owing to the increasing concerns about its persistence, bioaccumulation, and toxicity, it has been phased out in various manufacturing activities, including plastic production, since it was added to the POPs list.[9] Consequently, the presence of PFBA in plastics could be regarded as a short-chain alternative to PFOA.[65] H-PFCA (C4, C8−C19) were also measured in different types of plastic,
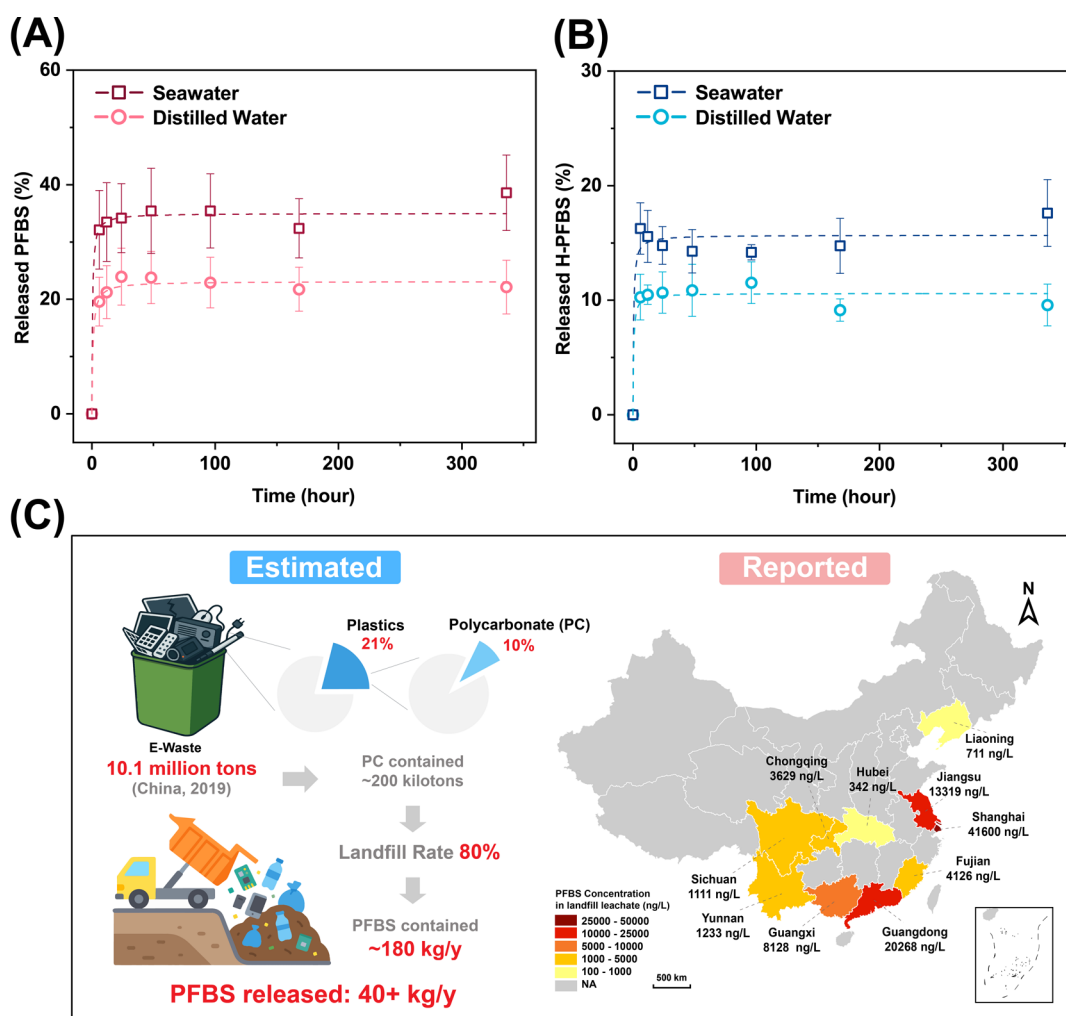
**Figure 3.** Release of PFBS (A) and H-PFBS (B) from polycarbonate (PC) samples after aging simulation in distilled water and seawater. (C) The estimated annual amount of PFBS that can be released into Chinese landfill leachates, and reported PFBS concentrations in landfill leachates across ten provinces of China, reconstructed from earlier studies.[51,75−77]

with average concentrations among all plastic products ranging from 0.0274 to 5.27 ng/g. Previous reports have revealed the application of H-PFCA for facilitating the emulsion polymerization of vinyl fluoride.[66,67] While their roles in nonfluorinated polymers have not been clearly verified, the presence of H-PFCA previously reported in plastic packages may suggest their potential application as processing aids during the manufacturing of other polymers.[30]

6:2 FTSA was detected in 8 polymer types of plastic with detection frequencies ranging from 11−47% and with average concentrations ranging from 0.0824 to 0.257 ng/g. The highest concentration was observed in one sample of PS made for foam board at 1.92 ng/g. It was documented as an additive for plastic material and resin manufacturing.[3] Moreover, bis-FMeSI was found in 8 out of the 11 polymer types but was mainly detected in 2 samples of ABS and 1 sample of PPO. Its concentrations in these two types of samples exceeded 10 ng/g. As an emerging PFAS with growing concerns, bis-FMeSI is primarily applied in lithium-ion battery production,[54,68] but current knowledge of its other applications remains insufficient. Bis(pentafluoroethylsulfonyl)imide (bis-FEtSI), structurally similar homologues to bis-FMeSI, have been patented as antistats in plastic or rubber,[3] which might explain the presence of bis-FMeSI in plastics.

**PFBS and H-PFBS in Individual PC Samples.** Our results found that PFBS and H-PFBS were the major PFAS congeners in PC plastics (Figure 1B), but their concentrations showed a wide variation in individual PC samples, with the concentrations of PFBS ranging from nondetection to 6130 ng/g, spanning over 3 orders of magnitude (Figure 2A). Notably, 7 out of 17 PC samples exhibited PFBS concentrations above 100 ng/g, while 3 of them exceeded 1000 ng/g. The highest PFBS concentration of 6130 ng/g was detected in PC75 (a battery spacer), followed by PC74 at 1995 ng/g (a gasket) and PC72 at 1465 ng/g (an insulating sheet), suggesting that PFBS may be preferentially used in PC products intended for electronic applications. This hypothesis was further supported by categorizing all PC samples based on daily and electronic uses, where PC products related to electronics exhibited significantly higher PFBS concentrations than those intended for daily uses (1070 vs 1.26 ng/g, $p < 0.01$, Figure 2B). Electronic components often operate under elevated temperatures, making thermal resistance a critical requirement for the plastic materials used. Although PC, as a widely used engineering plastic, offers excellent properties such as high hardness and impact strength, it still requires the incorporation of flame retardants to meet the stringent flame resistance standards for use in electronic applications.[69]

Notably, perfluoroalkyl sulfonates were implemented as effective flame retardants for PC in the early 1970s, with potassium perfluorobutanesulfonate (KPFBS) subsequently selected as the primary candidate for further commercial development.[70,71] PFBS shows its flame-resistant effect in PC primarily by suppressing gas-phase combustion through the release of fluorinated and sulfonated species, and by promoting condensed-phase char formation catalyzed by acid generated from its sulfonate group.[70,72] Thus, based on the high concentrations of PFBS in PC products only for electronic uses, along with patent-supported evidence, its intentional addition is most plausibly attributed to enhancing flame resistance.

Additionally, the presence of H-PFBS exhibited a similar trend to PFBS, as H-PFBS was detected in PC samples where high levels of PFBS (>100 ng/g) were also observed (Figure 2A). For instance, the highest concentrations of H-PFBS (212 ng/g) and PFBS (6130 ng/g) were found in the same sample. Besides, although the number of samples with detected H-PFBS was small, the peak area ratios of H-PFBS to PFBS in all PC samples with both compounds detected were consistently below 4%, indicating that H-PFBS occurred at trace levels relative to PFBS (Table S8). This is in line with similarly H-PFBS to PFBS ratios reported in Ruhr and Rhine river samples near fluorochemical industries (3% and 5%, respectively),[64] suggesting that H-PFBS likely originates as an impurity generated during PFBS synthesis. To validate this, commercially available PFBS products for industrial use were purchased for H-PFBS detection in the current study, and the results showed consistently low peak area ratios of H-PFBS to PFBS (ranging from 0.07% to 3%). Additionally, this interpretation is further supported by a 2012 patent,[52] which explicitly documents that H-PFBS can form as an unintended byproduct during PFBS manufacturing, resulting from incomplete fluorination under suboptimal processing conditions (Figure 2C).

**Migration of PFBS and H-PFBS from PC Plastics.** Six PC samples with the highest PFBS concentrations were selected to simulate the environmentally relevant migration of PFBS and H-PFBS from PC plastics under a realistic scenario with day−night natural cycling. The results showed that PFBS and H-PFBS in PC were released rapidly to both distilled water and artificial seawater in the first 6 h and reached the release equilibrium within a single day (Figures 3A,B, S8 and S9), significantly faster than the previously reported leaching of phthalate esters from microplastics to water, which required approximately 10 days to reach equilibrium.[35] After 14 days, average concentrations of PFBS and H-PFBS in the water sample can reach up to 484 and 3.31 $\mu$g/L, respectively, contributing to 22% and 10% of the total methanol-extractable concentration in PC plastics. Elevated concentrations of these two compounds (897 and 6.00 $\mu$g/L) were found in artificial seawater compared to distilled water, with average release percentages increasing to 39% for PFBS and 18% for H-PFBS. Additionally, the process of PFBS and H-PFBS release in water aligned well with pseudo-first-order kinetics, indicating that migration primarily occurs through diffusion from the plastic interface.[73] The lack of correlation between equilibrium PFBS release levels ($Q_e$) and initial addition amounts across different samples suggested that differences in product properties might also influence migration behavior (Table S9). Given that the flame-resistant mechanism of PFBS in PC involves gas-phase radical inhibition and acid-catalyzed char formation during combustion,[72] PFBS functions as an additive-type flame retardant, rather than a reactive one chemically bonded to the polymer matrix. This physical incorporation may explain the rapid leaching behavior observed in our experiments. In addition, the release patterns of PFBS and H-PFBS were highly synchronized, and no trend of increasing H-PFBS accompanied by decreasing PFBS was observed after reaching equilibrium, further suggesting that H-PFBS was likely present as an impurity associated with PFBS rather than its degradation product.

The leaching simulation in distilled water used for assessing landfill-waste regulatory compliance,[34] reflects early stage leachate generation, simulating the infiltration of moisture and rainfall during the initial phase of landfilling.[14,74] Although no exact threshold is currently available to assess the risk of PFBS leaching, the substantial release observed under this conservative condition strongly indicates that PC wastes containing PFBS may be unsuitable for direct landfilling. This highlights the need for appropriate waste classification and preliminary treatment before disposal. Furthermore, the high mobility of PFBS observed in seawater underscores the potential risk of marine contamination in cases of improper plastic waste disposal, reinforcing the importance of implementing effective waste management strategies on PFBS-containing plastics across both terrestrial and coastal environments.

**Link to PFBS Occurrence in Landfill Leachates.** Our findings suggest that PFBS is significantly present in PC plastic products related to electronics with considerable migration potential, which poses challenges to the end-of-life management of e-waste-related plastic materials. In 2019, China generated 10.1 million tons of e-waste, accounting for nearly 20% of the global e-waste production.[45] However, around 80% of the e-waste was disposed of in landfills.[48] Among the complex composition of e-waste, plastics constitute 21% of e-waste by mass,[46] with PC typically comprising approximately 10% of the plastic fraction.[47] Given the substantial uncertainties regarding direct statistical data of PFBS loading and leaching from e-waste plastics in landfills, a conservative estimate based on the occurrence data and leaching results in this study suggests that ~190 kg/y of PFBS could be present in landfilled PC e-waste plastics in China in 2019 (Figure 3C). Of this, roughly ~40 kg/y may be released into the leachates under conservative assumptions. This value may correspond to considerable proportions of the total annual PFAS emission flux from national landfill leachates reported for the top three countries, accounting for 6% of the United States (620 kg/y), 10% of China (419 kg/y), and 20% of Japan (196 kg/y).[14]

PFBS has been widely documented in landfill leachates from China, which further supports our estimated PFBS emission.[13] Over the past decade, high levels of PFBS have been detected in landfill leachates across ten provinces in China (Figure 3C and Table S10). Shanghai reported the PFBS concentration in raw landfill leachates at 41600 ng/L in 2015, accounting for 14% of the reported total PFAS concentration.[75] As documented in several studies, PFBS was frequently detected in landfill leachates from Guangdong and Jiangsu, with average concentrations at 20268 ng/L and 13319 ng/L, respectively, contributing 60% to the summed PFAS in Guangdong and 40% in Jiangsu.[51,75,76] Huang et al. have found that PFBS was dominant (contributions ranging from 8% to 31%) out of 17 PFAS in leachates from Western China in 2022, with concentrations varying from 146 to 13626 ng/L.[77] A recent

study in 2024 revealed that PFBS has become increasingly dominant in landfill leachates from 11 Chinese cities, accounting for an average of 68% of the total detected PFAS, with an average concentration at 4507 ng/L.[76] These unusually high PFBS levels and proportions to total PFAS fluxes in landfill leachates are generally attributed to the considerable amounts of disposed PFBS-containing solid waste, but the precise sources remain uncertain. Among the highly suspected contributors, textiles such as carpets and leather products have drawn attention due to the extensive use of PFBS in waterproof coatings and surface care products.[14,75,78] However, the reported concentrations of PFBS in these materials (ranging from 0.03 to 120 ng/g) appear insufficient to fully account for the levels observed in landfill leachates.[16,19,79] On the other hand, although carpets have been reported to contain C4-based perfluoroalkyl sulfonamide and derivatives, recognized precursors of PFBS, their concentrations are generally several orders of magnitude lower than what would be required to explain the PFBS levels measured in leachates.[18,19,80] Thus, the high PFBS concentrations quantified in PC plastics, together with the broadly consistent proportion of estimated PFBS to total PFAS relative to values reported for landfill leachates, suggest that PFBS-containing PC products represent a plausible emission source contributing to the observed PFBS contamination in landfills.

Additionally, it is noteworthy that two recent studies have reported the co-occurrence of H-PFBS and PFBS in landfill leachates, though the ratio of these two compounds is not available.[50,51] Based on our findings that H-PFBS was identified as a trace impurity of PFBS in PC plastics with high migration potential, we propose that H-PFBS in landfill leachates may similarly originate from PFBS-containing plastic waste.

## ■ ENVIRONMENTAL IMPLICATIONS

This study has several limitations. First, the mild extraction procedure applied in this study was designed to reflect environmentally migratable PFAS, rather than to quantify the total PFAS burden in plastics, and therefore did not incorporate more exhaustive methods (e.g., tetrahydrofuran extraction, TOP assay), which may have led to an underestimation of nonextractable PFAS compounds. Future studies should employ more comprehensive solvent extraction approaches to fully characterize the PFAS profiles in plastics. Second, ultrashort-chain PFAS (e.g., TFA and TFMS), which are not generally recognized to be intentionally added for plastic production and present significant analytical challenges in conventional reversed-phase liquid chromatography methods, were not included in the scope of this study.[81] Third, the migration simulation was not conducted under controlled laboratory conditions (e.g., fixed UV exposure and consistent temperature). Future studies incorporating experiments with well-controlled variables are needed to elucidate the mechanisms by which different aging stressors influence the release of PFBS and H-PFBS from PC plastics. Finally, the estimated PFBS release into landfill leachates was based on the experimental results from a conservative release simulation rather than under landfill conditions (i.e., elevated temperatures and microbial activity), representing a conservative, order-of-magnitude indication of the potential emissions from PFBS-containing PC plastics. The actual release within landfills may be underestimated.

Nonetheless, the high-resolution mass spectrometry-based screening employed in this study successfully identified a broad suite of PFAS compounds in various plastic products, confirming that plastics serve as an important and widespread carrier of PFAS in practical applications, though generally at lower concentrations compared to other consumer products. The nondetection of most legacy PFAS, like PFOS, which was historically patented as a mold release agent, implies the shift toward other PFAS alternatives in industrial plastic processing. However, the high levels of PFBS and its impurity H-PFBS found in PC products used for electronics should raise significant concerns regarding both human health and environmental safety.

Although polymer components in electronics (e.g., socket cases) typically pose minimal health risks to humans via direct contact compared to plastics intended for daily use, PFBS, as a physically embedded additive, may easily migrate into airborne dust.[72,82] This migration poses potential health risks, particularly in indoor environments where electronic equipment is prevalent. These risks become more pronounced during the recycling and dismantling activities on e-waste plastic components, which can further increase PFBS exposure for workers.[83] Furthermore, the recycling and dismantling activities can release a substantial amount of PFBS into the environment, leading to contamination of surrounding ecosystems.[84] The European Chemicals Agency (ECHA) has listed PFBS and its salts on the Candidate List of substances of very high concern (SVHCs) in 2020 based on their characteristics of being persistent, bioaccumulative, and toxic (PBT). The U.S. Environmental Protection Agency (EPA) has also released the final report of the Human Health Toxicity Values for PFBS in 2021, confirming potential effects of thyroid disruption, developmental delays, and kidney impacts from PFBS exposure.[85] Recently, accumulating evidence has highlighted additional concerns on PFBS, including immunotoxicity, neurobehavioral alterations, and hepatotoxic effects linked to metabolic disruption.[86−88] The high PFBS levels observed in PC plastics therefore underscore the urgent need to reassess the necessity of its application, safety, and associated risks.

Considering the persistence and high mobility of PFBS, the abundant PFBS consistently reported in landfill leachates over the past years has lacked a clearly identified source input, which previous research has not adequately explained. The high PFBS levels observed in PC plastics in this study suggest that PC plastics may represent a previously underrecognized potential contributor to such input. Moreover, the rapid migration and high mobility of PFBS pose significant risks to the surrounding environments, such as nearby groundwater and surface waters. To mitigate these potential risks, greater attention should be given to classifying, recycling, and managing plastic waste based on their polymer types and functions, rather than carrying out landfilling indiscriminately. Finally, the co-occurrence of H-PFBS points to manufacturing origins and may serve as a new marker for PFBS-related source tracking.

## ■ ASSOCIATED CONTENT

### Ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.5c09017.

Detailed information on chemicals and reagents; plastic product information; environmental information on leaching experiments; MDLs and blank concentrations; surrogate and matrix spike recoveries; peak ratios of H-PFBS to PFBS for all PC samples; release kinetics parameters; PFBS concentrations in landfill leachates from previous studies; workflow of PFAS screening; chromatograms and MS2 spectra of all identified PFAS; and release kinetics of individual samples (PDF)

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**Notes**

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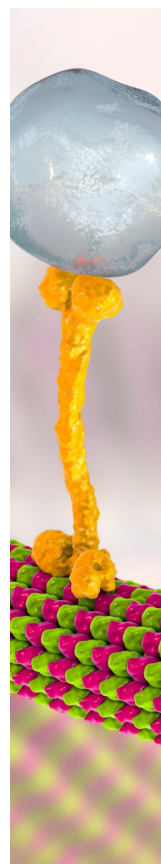
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