

Article

Holistic Estuarine Monitoring: Data-Driven and Process-Based Coupling of Biogeochemical Cycles of Per- and Polyfluoroalkyl Substances

Fatih Evrendilek *  and Gulsun Akdemir Evrendilek 

University of Maine Cooperative Extension, Orono, ME 04469, USA; gulsun.akdemir@maine.edu

* Correspondence: fatih.evrendilek@maine.edu

Abstract

Better understanding the fate and transport of estuarine per- and polyfluoroalkyl substances (PFASs) requires coupling multiple matrix-specific biogeochemical roles, rather than relying on a single-matrix approach. We therefore evaluated sediment and biological matrices (blue mussels (BMs), *Mytilus edulis*; and hardshell clams (HSCs), *Mercenaria mercenaria*) as complementary indicators of PFAS contamination across three locations over a 240-day period following a spill event. A three-tiered analytical approach was applied: Tier 1 used non-parametric statistics to assess the broad-spectrum detection patterns for a total of 40 PFASs ($n = 47$ samples); Tier 2 employed generalized regression (adaptive Elastic Net), random forest, and artificial neural networks to model the concentrations of the most frequently detected PFASs (PFOS, PFOA, PFHxA, and PFOSA) ($n = 188$ observations); and Tier 3 implemented a system dynamics model to mechanistically couple the PFOS and 5:3 FTCA fate. The results suggest that the sediment acted as a long-term sink for legacy long-chain compounds (99.3%, primarily PFOS), while the biota, particularly BMs, acted as sensitive recorders of acute pulses and hydrophilic precursors, uniquely accumulating 5:3 FTCA during spring pulses ($p < 0.001$). All the models identified the matrix type as the dominant driver of the most prevalent PFAS concentrations. A reliance on sediment monitoring alone may fail to capture the majority of the active contamination burden sequestered in the biota, suggesting that effective risk assessment necessitates an integrated view.

Keywords: PFAS fate and transport; environmental monitoring; blue mussels; hardshell clams; sediment



Academic Editors: Hugo Valdés and Aldo Saavedra

Received: 27 December 2025

Revised: 19 January 2026

Accepted: 21 January 2026

Published: 22 January 2026

Corrected: 6 February 2026

Copyright: © 2026 by the authors.

Licensee MDPI, Basel, Switzerland.

This article is an open access article distributed under the terms and conditions of the [Creative Commons Attribution \(CC BY\)](https://creativecommons.org/licenses/by/4.0/) license.

1. Introduction

The source characteristics (type, magnitude, and release rate) of per- and polyfluoroalkyl substances (PFASs) initiate contamination [1], while their subsequent fate and transport in aquatic ecosystems are driven by compartment-specific interactions. Although sediment has traditionally been monitored for legacy long-chain acids (e.g., PFOS), its ability to reflect water column dynamics and bioavailable fractions remains contested [2]. This reliance on a single-matrix-specific approach creates a critical liability within the context of remediation strategies and socio-economic-ecological system (SEES)-level risk assessment [3,4].

On 19 August 2024, Maine's largest PFAS-based aqueous film-forming foam (AFFF) spill (5500 L AFFF + 190,000 L water) at Brunswick Executive Airport produced surface

water PFAS concentrations that exceeded draft EPA criteria [5], from the spill site to the Harpswell Cove estuary within 11 days [6]. AFFF formulations are complex mixtures containing both perfluoroalkyl acids (PFAAs) and their polyfluoroalkyl precursors, such as fluorotelomer sulfonates (e.g., 6:2 FTS). In the environment, these precursors can undergo aerobic biotransformation over timescales of days to weeks to form stable terminal PFAAs, including perfluoroalkyl carboxylates (e.g., 5:3 FTCA from 6:2 FTS) and sulfonates (e.g., PFOS). This dynamic transformation cascade complicates source attribution and fate prediction, as the measured PFAS profile in any given compartment reflects both the original mixture composition and in situ transformation processes, influenced by redox conditions, microbial activity, and transport times.

Following this rapid downstream transport, the present study addresses this knowledge gap by integrating abiotic (sediment) and functionally distinct biotic (hardshell clams (HSCs) vs. blue mussels (BMs)) matrices through a holistic approach analyzing broad-to-PFAS-specific patterns. The selection of BMs and HSCs as biotic sentinels is grounded in their distinct exposure pathways and physiological traits. BMs are epibenthic filter-feeders that primarily sample the water column, providing an integrated signal of dissolved and particulate PFASs over short timescales (days to weeks). In contrast, HSCs are infaunal bivalves that interface with both the sediment porewater and overlying water, leading to an exposure dominated by sediment–water exchange and potentially integrating contamination over longer periods. Differences in PFAS partitioning—with long-chain sulfonates (e.g., PFOS) favoring protein-rich tissues and short-chain compounds remaining more bioavailable—can lead to divergent accumulation patterns between these species, making their comparison particularly informative for understanding compartment-specific PFAS fate.

Accurate quantification of PFASs across environmental matrices is inherently challenged by the potential for laboratory and field contamination and the frequent occurrence of analyte concentrations near method detection limits. Consequently, rigorous quality assurance and quality control (QA/QC) protocols (e.g., the use of isotopically labeled internal standards, procedural and field blanks, and recovery assessments) are fundamental for data integrity. These considerations are particularly critical when interpreting datasets with high rates of non-detects (NDs), a common feature in environmental PFAS monitoring. To robustly address this analytical reality, the present study adopts a tiered analytical framework. The initial tier (Tier 1) prioritizes the analysis of detection patterns and frequencies, a strategy that mitigates the influence of concentration-level uncertainty and provides a foundation for the subsequent data-driven (Tier 2) and process-based (Tier 3) modeling. Furthermore, this approach sets practical limits for detailed statistical and mechanistic modeling, logically restricting such analyses to the subset of analytes with sufficient detection frequency to support reliable concentration estimates.

We hypothesized that (1) sediment and biota (BMs/HSCs), as different ecosystem compartments, occupy distinct biogeochemical roles, with the sediment serving as a stable reservoir for hydrophobic long-chain sulfonates (PFOS) and the biota capturing pulses of hydrophilic short-chain precursors (e.g., 5:3 FTCA) and carboxylates; (2) the matrix type is the primary determinant of variance in the concentrations of most prevalent compounds (e.g., PFOS, PFOA, PFHxA, and PFOSA); and (3) a system dynamics model can simulate the coupled dynamics of persistent PFOS and transient precursors, explaining the observed matrix-specific patterns. To test these hypotheses, we employed a three-tiered approach combining non-parametric statistics, data-driven predictive modeling, and mechanistic system dynamics simulation. From a regulatory and risk perspective, estuarine bivalves are not only effective sentinel organisms but are also directly relevant to seafood safety and human exposure. Tissue concentrations in species like BMs and HSCs, when coupled with

rigorous QA/QC, provide critical data for consumption advisories and risk assessments. By comparing the PFAS burdens in these edible species with those in sediment, this study addresses the need to understand how contaminant dynamics in (a)biotic compartments translate to exposure in seafood, a key component of SEES-level risk management. This study provides the first integrative assessment combining non-parametric tests, machine learning, and mechanistic modeling to elucidate PFAS dynamics across (a)biotic compartments in a post-spill estuary.

2. Materials and Methods

2.1. Study Design and Data Structure

We collected samples from three locations (S1, S2, and S3) across eight dates (17 October, 7 November, and 21 November 2024; 16 January, 12 March, 9 April, 7 May, and 11 June 2025) after the August 2024 Brunswick spill. In straight-line distance, stations S1, S2, and S3 are located approximately 3.5 km southwest, 5.0 km south-southwest, and 5.9 km southwest of the spill site, respectively. In terms of east–west alignment, S2 is the most easterly station, followed by S1, with S3 being the furthest west. The PFASs in the sediment, BMs, and HSCs were analyzed by following EPA Method 1633. The sediment samples were freeze-dried and homogenized; the biological tissues were homogenized wet. All extracts were cleaned by using solid-phase extraction. The analysis was performed by liquid chromatography (LC-40D XS; Shimadzu Scientific Instruments; Kyoto, Japan) with tandem mass spectrometry (LCMS-8050 triple quadrupole; Shimadzu Scientific Instruments; Kyoto, Japan) (LC-MS/MS) using negative electrospray ionization. Separation was achieved on a C18 column (50 mm × 2.1 mm, 1.7 µm particle size) maintained at 40 °C. The mobile phases consisted of (A) 2 mM ammonium acetate in 5:95 acetonitrile/water and (B) 100% acetonitrile, delivered at a flow rate of 0.4 mL/min under a gradient elution program. A suite of 40 target PFAS analytes was monitored. Quantification was performed by using a corresponding set of isotopically labeled internal standards, spiked prior to extraction. The calibration curves (0.01–100 ng/mL) exhibited linearity ($R^2 > 0.99$). The method detection limits were compound- and matrix-specific. QA/QC included the analysis of procedural blanks with each batch. No target PFASs were detected above the limit of quantification in these blanks. Recovery of internal standards met the method acceptance criteria (commonly 70–130%). The duplicate analyses demonstrated acceptable precision (<20%).

A three-tiered analytical framework was applied to address the distinct research questions at different scales of PFAS complexity. Tier 1 (broad-spectrum patterns; $n = 47$ samples) included all 40 targeted PFASs and their sum (Σ PFAS) across the three matrices (sediment, BMs, and HSCs) to assess the detection distributions and counts. Tier 2 (prevalent compound-specific patterns; $n = 188$ observations) involved PFOS, PFOA, PFHxA, and PFOSA, which exhibited sufficient detection frequency (>25%) for robust predictive modeling of their concentrations. Tier 3 (systems dynamic modeling) focused on the simulation of PFOS and 5:3 FTCA dynamics.

To evaluate the PFAS identity as an independent factor, the dataset was reshaped from a wide format in Tier 1 ($n = 47$ physical samples) to a long format in Tier 2 ($n = 188$ observations), enabling generalized linear mixed (GLMM), generalized regression (GR), random forest (RF), and artificial neural network (ANN) models to quantify the interaction effects, such as matrix × PFAS. For Tier 2, the latitude (Lat) and longitude (Lon) recorded in decimal degrees were used instead of the categorical location variable. Similarly, the day of year (DOY) was transformed into cyclical coordinates ($\text{DOY}_{\sin}$, $\text{DOY}_{\cos}$) using a harmonic transformation to account for seasonal periodicity without artificial discontinuities (e.g., between 31 December and 1 January). Each PFAS species was treated as an independent analytical unit for detection and co-occurrence analyses; inference about the spatiotemporal

effects was restricted to matrix- and compound-specific contrasts to avoid pseudoreplication. The non-detects were treated as missing data to avoid bias in the detection-based analyses and preserve conservative lower-bound estimates.

Given the extreme sparsity (93% NDs; non-detects/total points = $1750/1880 \times 100 \approx 93.19\%$), substitution-based imputation was avoided to prevent artificial signal inflation; all concentration-based modeling was restricted to PFASs with sufficient detection frequency ($>25\%$). This conservative approach avoided imputation bias but did not utilize information from censored values; future studies with larger sample sizes may benefit from formal censored data regression techniques (e.g., Tobit model) for concentration estimation. The PFAS concentrations were expressed in ng/g wet weight (WW) for the biota (BMs and HSCs) and ng/g dry weight (DW) for the sediment. This reporting convention is conservative, as converting the biota to DW would typically increase the PFAS concentrations by a factor of 5–6, thus amplifying the observed divergence between the biotic and abiotic matrices. Accordingly, all cross-matrix comparisons should be interpreted as conservative with respect to biotic exposure. The detection frequencies were calculated as the percentage of samples within a group (matrix or location) containing at least one analyte above the limit of detection.

2.2. Data Analyses

2.2.1. Tier 1: Broad-Spectrum (Total PFASs) Detection Patterns

For Tier 1, the high sparsity (93% NDs) and non-normal distribution of the data (Shapiro–Wilk $p < 0.05$) violated parametric assumptions, requiring robust non-parametric methods for inference. A categorical response analysis (CRA) was employed to quantify how the three categorical factors (matrix, location, and DOY) and their interactions affected the detection rates (DRs) and patterns of PFASs ($n = 47$ physical samples). All proportions were reported with 95% confidence intervals (CIs), with significance assessed using q -values of 0.05 after a Benjamini–Hochberg False Discovery Rate (FDR) correction and uncorrected p -values. One-way non-parametric analyses were conducted using Kruskal–Wallis and Steel–Dwass post hoc tests to detect significant differences in the Σ PFAS among the three factors by controlling for Type I errors ($n = 47$). Co-occurrence patterns were analyzed using Spearman’s correlations (ρ).

2.2.2. Tier 2: Drivers of Prevalent PFAS Concentrations

For Tier 2, the data analyses were restricted to the four most frequently detected analytes (DRs: 89.4% PFOS, 46.8% PFOSA, 36.2% PFHxA, and 25.5% PFOA) to satisfy degrees of freedom requirements and avoid singular matrices caused by excessive NDs, with the PFAS identity included as a categorical variable. To predict their concentrations, the performance of artificial neural network (ANN; $n = 93$), random forest (RF; $n = 93$), and generalized regression with adaptive Elastic Net (GR_{AEN} ; $n = 188$) was compared using the coefficient of determination (R^2) and root mean square error (RMSE) on the validation data. The GR_{AEN} was specified with a LogNormal response distribution and validated using the Akaike Information Criterion corrected for small samples (AIC_c). This approach performs handling of multicollinearity as well as shrinking the coefficients of non-informative terms to zero, thus providing a parsimonious and interpretable model. Detailed hyperparameter specifications for the RF, ANN, and GR_{AEN} are provided in Supplementary Table S1. The relative predictor importance was quantified using Monte Carlo-based independent resampling ($n = 5000$). The LogNormal generalized linear mixed model (GLMM) with random effects (47 SampleIDs) was adopted to determine significant fixed effects for spatial coordinates (Lon, Lat), temporal components (DOY_{sin} , DOY_{cos}), matrix, PFAS identity, and their factorial interactions ($n = 188$). Statistical analysis was

performed using JMP Pro software (Version 19.0, JMP Statistical Discovery LLC, Cary, NC, USA).

2.3. Tier 3: System Dynamics Modeling

For Tier 3, to mechanistically interpret the observed matrix-specific PFAS patterns, we implemented a system dynamics model in STELLA® (Version 8.1, ISEE systems, Inc., Lebanon, NH, USA) representing the coupled fate of fluorotelomer precursors (FTCA) and PFOS across estuarine water, sediment, BMs, and HSCs. The model was calibrated to the observed field concentrations using a combination of literature-derived rate constants and site-specific parameter adjustments to ensure mechanistic plausibility while fitting the empirical trends. The model explicitly resolves (i) short-lived, pulse-driven FTCA and (ii) persistent PFOS dynamics at a temporal resolution of 6 h for 600 d, using mass-balance stock–flow ordinary differential equations.

State variables included FTCA and PFOS accumulations in water, BMs, HSCs, and active and deep sediments. We structured PFAS dynamics into three kinetic pools based on observed turnover times: (1) active pool (≤ 3 months): FTCA in water and BMs, representing pulse-responsive, rapidly cycled contamination; (2) slow pool (3 months–10 yr): PFOS in biota and surface sediment, reflecting slower biota depuration, sorption-controlled retention, and sediment–water exchange; and (3) passive pool (> 10 yr): PFOS in deep sediment, representing burial-dominated sequestration. This hierarchy organizes PFAS dynamics according to compound-, hydrometeorological event-, organism-, and sediment-specific processes. Model parameters, state variables, differential equations, and calibration data are detailed in Tables S2 and S3.

3. Results and Discussion

To quantify PFAS contamination across the (a)biotic compartments, Tier 1 presents the broad-spectrum detection and distribution patterns of 40 PFASs in Section 3.1. Tier 2 identifies the spatiotemporal dynamics of the four most detected PFASs (Section 3.2) and drivers through data-driven models (Section 3.3). Tier 3 integrates the empirical findings to couple the biogeochemical roles of the compartments (Section 3.4) and simulates their coupled dynamics via a mechanistic model (Section 3.5). The paper then addresses its scope and limitations (Section 3.6). Finally, a synthesis of insights and their implications for monitoring, risk assessment, and remediation are presented (Section 3.7).

3.1. Broad-Spectrum (Σ PFAS) Detection Patterns and Drivers

The sediment exhibited a significantly lower detection probability for PFASs than the biota (Table 1), with a DR of 3.89% (95% CI: 2.70–5.56%; $q = 0.0281$; $p = 0.0047$), a 52.9% reduction compared to the combined BMs (DR: 7.92%; 95% CI: 6.16–10.12%; $q > 0.3351$; $p = 0.168$)/HSCs (DR: 8.86%; 95% CI: 6.55–11.89%; $q > 0.1919$; $p = 0.064$) baseline. The Σ PFAS concentrations differed among the matrices (Kruskal–Wallis $p = 0.0384$), with the pairwise comparisons showing no significant differences (Steel–Dwass $p = 0.0538$ for sediment vs. HSCs; 0.9174 for sediment vs. BMs; and 0.0746 for HSCs vs. BMs). The sediment (mean = 2.34 ± 1.56 ng/g DW) showed lower detection but concentrations comparable to or slightly higher than the BMs (mean = 4.80 ± 6.06 ng/g WW) and HSCs (mean = 1.31 ± 0.89 ng/g WW). To enable a direct mass comparison, the biota WW concentrations can be converted to a DW basis by using a standardized conversion factor of 5, consistent with the reported moisture contents for marine bivalves (typically 75–85% water; corresponding to dry-weight factors of 4–6.7) [4]. This order-of-magnitude estimate illustrates the potential scale of discrepancy between the matrices, but should be interpreted

as indicative of a qualitative difference rather than a precise partitioning, as the exact value is sensitive to the chosen conversion factor (Table 1).

Table 1. Statistical summary of Σ PFAS concentrations by matrix.

Matrix	Sample Size (n)	Mean (ng/g)	Median (ng/g)	SD	Max (ng/g)	Detection Frequency (%)	Dominant PFAS Identity
BMs	18	4.80	2.33	6.06	22.94 †	7.9	5:3 FTCA (precursor)
HSCs	11	1.31	1.35	0.89	3.70 ‡	8.9	Mixed profile
Sediment	18	2.34	2.01	1.56	6.15 §	3.9	PFOS (legacy)

† The maximum concentration in the BMs (22.94 ng/g WW; 12 March 2025; S1) defines the ecosystem's active state, driven by the acute pulse of 5:3 FTCA. ‡ The maximum concentration in the HSCs (3.70 ng/g WW; 17 October 2024; S1) occurred independently of the pulse state, reflecting a mixed background variability. § The maximum concentration in the sediment (6.15 ng/g; 9 April 2025; S1) defines the ecosystem's legacy sink state, characterized by historical accumulation of long-chain PFOS that remains stable regardless of seasonal water column pulses.

Although the spatial heterogeneity inference was limited by the number of locations ($n = 3$), S3 (mean: 1.30 ± 0.80 ng/g) appeared to have significantly lower Σ PFAS contamination than S1 (mean: 6.81 ± 7.11 ng/g) and S2 (mean: 2.86 ± 2.52 ng/g) (Kruskal–Wallis $p = 0.0008$; Steel–Dwass $p = 0.0026$). This pattern is suggestive of a contamination gradient but should be interpreted with caution due to the limited replication and potential unmeasured confounders (e.g., local sediment composition and organism physiology). The highest and lowest detection rates were S1 (8.50%; 95% CI: 6.15–11.64%; $q = 0.683$; $p = 0.138$) and S3 (5.50%; 95% CI: 4.12–7.30%; $q = 0.683$; $p = 0.228$), respectively. While the detection probability was spatially uniform across the sites, the Σ PFAS magnitude showed a spatial pattern, with S3 as a contamination coldspot.

Temporally, the Σ PFAS contamination exhibited minimal seasonal variation in the detection probability, with DOY71 (mean = 6.25 ± 8.19 ng/g) and DOY16 (mean = 0.75 ± 0.82 ng/g) as the highest and lowest Σ PFAS concentrations, respectively (Kruskal–Wallis $p = 0.2734$; Steel–Dwass $p = 0.5170$). The highest and lowest detection rates were DOY291 (9.17%; 95% CI: 6.13–13.49%; $q = 0.97$; $p = 0.12$) and DOY16 (2.50%; 95% CI: 1.07–5.72%; $q = 0.39$; $p = 0.024$), respectively. Notable three-way (matrix $\times$ location $\times$ DOY) interaction effects were observed, with BMs $\times$ S2 $\times$ DOY 71 and HSCs $\times$ S1 $\times$ DOY 291 exhibiting detection rates of 17.5% (95% CI: 8.75–31.95%; $q > 0.05$; $p = 0.0072$) that quadrupled the sediment baseline and doubled the matrix averages.

3.2. PFAS-Specific Concentration Patterns

Figure 1 illustrates the distinct spatiotemporal dynamics of the four most frequently detected PFASs (PFOS, PFOSA, PFHxA, and PFOA) across the three compartments and three sampling locations over a 240-day period following the spill event. The sediment profile was dominated by long-chain compounds (99.3%, primarily PFOS; Σ PFOS/ Σ PFAS = $(41.819/42.141) \times 100 \approx 99.3\%$), reflecting hydrophobic adsorption to organic carbon. Conversely, the BMs profile was dominated by precursors (85.1%, primarily 5:3-FTCA), while the HSCs exhibited a mixed profile of 32% short-chain and 53% long-chain compounds. The exclusive sequestration of 5:3 FTCA in the BMs, particularly during the spring pulse period (e.g., March–June 2025), defined an active pulse state (Tables 1 and 2).

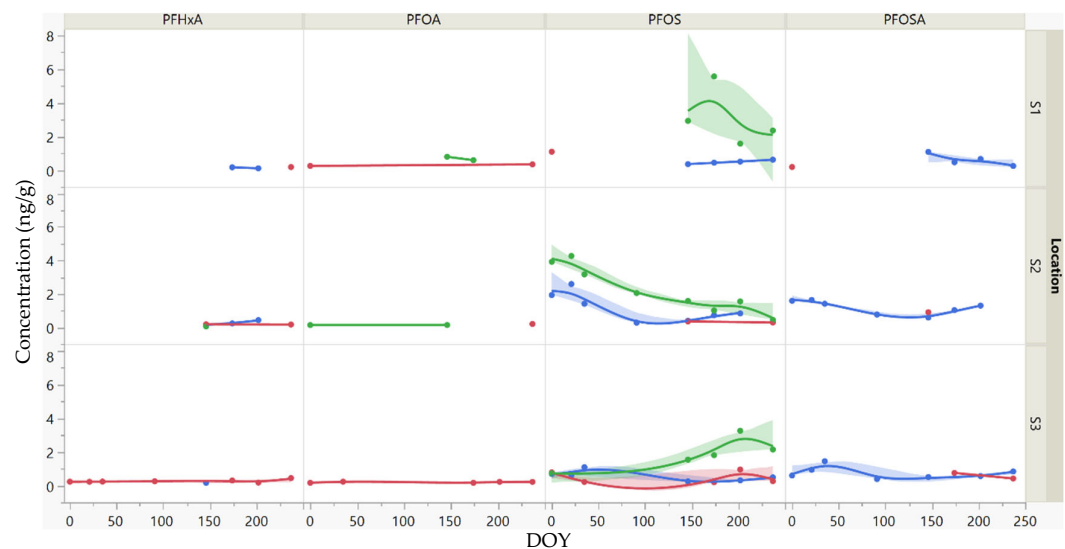


Figure 1. Spatiotemporal dynamics of the four dominant PFASs across the three environmental compartments and three sampling locations in the Harpswell Cove estuary (ME, USA) ($n = 188$). The 240-day period following the August 2024 Brunswick contamination event spanned 17 November 2024 (DOY = 0)–11 June 2025 (DOY 237). S1 and S3 are the closest and farthest points from the spill site, respectively. The visualizations are based on penalized B-splines (P-splines) without imposing any parametric constraints, thus avoiding overfitting while preserving biogeochemically meaningful fluctuations. The shaded regions (95% confidence intervals) quantify uncertainty in the estimated spatiotemporal trajectories, with the bandwidths reflecting sampling density and measurement precision. Blue, red, and green lines: BM, HSC, and sediment, respectively.

Table 2. Prevalent PFASs by ecological state and spatiotemporal zone ($n = 47$).

Ecological State	Matrix	Location	Sampling Date	Detected PFASs	Chain Class	Conc.* (ng/g)
Active pulse	BMs	S1	11 June 2025	5:3-FTCA	Precursor	11.7
				6:2 FTS	Precursor	0.856
				Sum (pulse)		12.556
Legacy sink	Sediment	S2	9 April 2025	PFOS	Long (C8)	5.56
				PFOA	Long (C8)	0.588
				Sum (sink)		6.148
Background level	HSCs	S3	Various	PFHxA	Short (C6)	0.301
				PFOS	Long (C8)	0.779
				Sum (mean)		1.31

* Concentrations for the active pulse and legacy sink states represent the maximum observed values from a single sample. For the background level, the individual PFAS concentrations are representative high values from specific samples (S3, 9 April 2025 for PFHxA; S3, November 2024 for PFOS), while the total (1.31 ng/g) is the matrix-wide mean Σ PFAS for the HSCs ($n = 11$). The active pulse reflects the highest single-sample total (Σ PFAS) in the BMs driven by precursor compounds. The legacy sink indicates the highest sediment PFOS concentration, reflecting historical accumulation. The background level represents the mixed, low-level exposure profile typical in the HSCs.

The PFOS dynamics varied with distance from the spill. At the proximal sites (S1 and S2), the sediment’s and BMs’ PFOS concentrations generally decreased over time. For example, the sediment’s PFOS at S2 declined from a maximum of approximately 4.0 ng/g at $t = 0$ (17 November 2024) to <1.0 ng/g by day 240, indicative of rapid initial dissipation. Conversely, the distal site (S3) exhibited a lagged response, with the sediment’s PFOS rising from near detection limits (<1.0 ng/g) to approximately 2.5 ng/g by around day 200. The

distance gradient ($S1 < S2 < S3$) appeared to reflect the downstream transport and delayed accumulation of PFOS at S3.

The spatial dynamics did not follow a uniform linear decay from the closest (S1) to the distal limit (S3). Sediment contamination was highest at S1 and S2 during the initial sampling phase, and decreased at S3. However, by day 200, the spatial gradient inverted for the sediment's PFOS, with its S3 concentrations exceeding those at the rapidly flushing S2 station. Despite the comparable initial concentrations of PFOS in the sediment at S1 and S2 (~ 4.0 ng/g), the BMs exhibited an initial uptake (~ 2.0 ng/g) at S2 nearly four times higher than that at S1 (~ 0.5 ng/g). This indicates that the magnitude of sediment contamination alone did not predict the BMs' uptake efficiency along the proximal sites.

The three compartments displayed divergent accumulation capacities (Figure 1). The sediment served as the primary reservoir for long-chain sulfonates (PFOS), exhibiting the highest concentrations (up to ~ 6.0 ng/g at S1) and the most dynamic spatiotemporal fluctuations. By acting as the primary receptor for sulfonamides and sulfonates, the BMs displayed pulse-like dynamics, particularly for PFOS and PFOSA, reflecting rapid equilibrium with changing environmental conditions. The HSCs exhibited high stability and low accumulation relative to the other matrices. Across all the PFAS species and locations, the HSCs' concentrations remained consistently low (< 0.5 ng/g) and showed a temporally minimal response to the environmental pulses observed in the sediment and BMs.

The PFASs' dominance and behavior varied markedly by chain length and functional group. PFOS was the dominant contaminant by mass, exhibiting concentrations 3–6 times higher than the other PFAS species and showed the strongest coupling to sediment. PFOSA displayed unique transient dynamics in the BMs, particularly at S2, where its concentrations followed a convex trajectory, namely, rising to a peak at day ~ 50 before depurating. PFHxA and PFOA were the minor contributors to the total PFAS mass. PFHxA showed detectable but low-level pulses in the biota, while PFOA was frequently below detection limits or stable at low concentrations across all the matrices.

Given the spill location east of all the sampling stations, the primary plume was inferred to have traveled west–southwest. According to a Spearman's correlation (ρ) analysis (Figure 2), the positive longitude–BMs' PFHxA correlation ($\rho = 0.833$; $p = 0.0394$) indicates that increased uptake of this dissolved-phase, short-chain compound by the water-filtering BMs peaked at more easterly stations (S2 and S1), aligning with the west–southwest advection of the dissolved plume from the spill site and stronger initial exposure at those stations. The negative longitude–HSCs' PFHxA correlation ($\rho = -0.644$; $p = 0.0444$) favored higher uptake by sediment-dwelling HSCs at more westerly stations (S3 and S1). This biotic dichotomy reflects the species-specific transport of PFHxA: water column-filtering BMs vs. sediment/porewater exposure-regulated HSCs uptakes in more easterly and westerly stations, respectively.

The significant positive correlation between PFOSA and PFOS ($\rho = 0.710$; $p = 0.0014$) within the BMs suggests the in situ biotransformation of the precursor into the terminal product or co-exposure from a common source along the transport path. The negative seasonal (DOY_{sin}) correlation for the BMs' PFOS ($\rho = -0.559$; $p = 0.0195$) reflects an overall decline over the study period, likely due to depuration or seasonal flushing. The combination of the strong co-occurrence of PFOS and PFHxS ($\rho = 1.0$; $p < 0.0001$) and mutual exclusivity between PFHxS and PFHxA, PFOSA and PFHxA, and PFOSA and PFOA ($\rho = -1.0$; $p < 0.0001$) in the HSCs reveals an uptake pattern by the sediment-dwelling HSCs where either sulfonates (PFOS and PFHxS) or carboxylates/precursors (PFHxA, PFOA, and PFOSA) are accumulated.

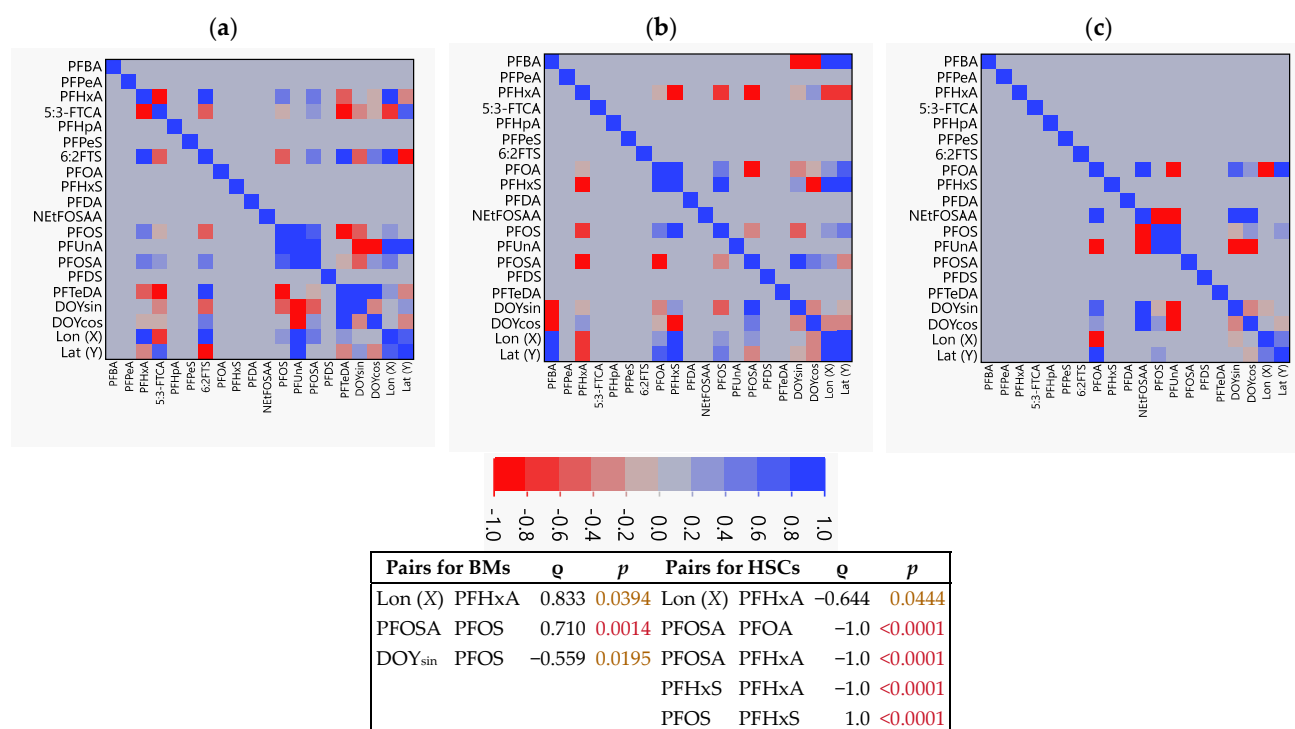


Figure 2. Spearman's correlation (ρ) heatmap among PFASs and associated variables and their correlated pairs at $p < 0.05$ according to (a) BMs (far left panel), (b) HSCs, and (c) sediment (far right panel) ($n = 47$).

3.3. Data-Driven Modeling of Prevalent PFAS Concentrations

To move from broad detection patterns to specific concentration drivers, we modeled the four prevalent compounds (PFOS, PFOA, PFHxA, and PFOSA; $n = 188$). As shown in Table S1, the ANN and RF models achieved higher training and validation R^2 values than the GR_{AEN} , but were not used for inferential conclusions due to their limited interpretability and potential overfitting under the modest sample size. Instead, the GR_{AEN} was selected for the final inference as it provided a sparse, interpretable set of coefficients while directly accounting for multicollinearity, essential for identifying stable drivers (generalized $R^2 = 0.60$; $AIC_c = 78.5$) (Tables S1 and S4). The relative predictor importance analysis by the ANN, RF, and GR_{AEN} converged, identifying the matrix (total effect = 0.628) and PFAS identity (total effect = 0.311) as the first and second strongest drivers of concentration variance, respectively (Table S5). The total effect represented the proportion of variance in the PFAS concentration attributable to each predictor, estimated via Monte Carlo-based independent resampling ($n = 5000$).

To explain the concentration variance in this well-mixed estuary, the GR_{AEN} model found a significant overall effect for both the matrix (Wald $\chi^2 = 18.9$; $p < 0.0001$) and PFAS types (Wald $\chi^2 = 40.5$; $p < 0.0001$), but zeroed out the coefficients for the spatiotemporal terms, except for the DOY_{sin} (the periodic seasonal component: Wald $\chi^2 = 4.4$; $p < 0.0353$). This finding emphasizes the significance of the matrix-specific biogeochemical roles over spatial proximity to sources for these PFASs.

The model coefficients showed that the PFAS concentrations were significantly lower in both the BMs (estimate = -0.83 ; $p < 0.0001$) and HSCs (estimate = -1.01 ; $p < 0.0001$) than in the sediment as the baseline. Similarly, PFHxA (estimate = -1.16 ; $p < 0.0001$) and PFOA (estimate = -1.22 ; $p < 0.0001$) showed significantly lower concentrations than PFOSA as the baseline, while PFOS did not differ significantly from PFOSA ($p = 1.0$). The DOY_{sin} showed a significant negative effect on the concentration (estimate = -0.19 ; $p = 0.035$). The negative DOY_{sin} coefficient and the exclusion of DOY_{cos} suggest a seasonal pattern with

no secondary peak or trough in mid-summer or mid-winter, with the PFAS concentrations plunging around April, rising through autumn, peaking in October, and declining through winter, potentially linked to the hydrometeorological dynamics. After accounting for the matrix and PFAS types, the exclusion of the spatial coordinates indicates that broad-scale geographic trends were not pronounced in this dataset.

However, the non-linear models (RF and ANN) attributed greater importance to the interaction terms (e.g., interaction effect = 0.133 for RF) than the GR_{AEN} (interaction effect = 0.029) (Table S5). While the system was overwhelmingly governed by the main compositional factors of the matrix and PFAS types, non-linear complexities were penalized in the parsimonious linear model. The GLMM analysis revealed significant fixed effects for the spatial (Lon, Lat) and seasonal (DOY_{sin}) components, highlighting structured spatiotemporal variability. More importantly, the GLMM model identified a significant interaction effect between the matrix and PFAS types (e.g., BMs–sediment $\times$ PFOA–PFOSA; $p = 0.0078$), indicating that the contamination profiles were matrix- and PFAS species-dependent.

The divergence in how the models attributed importance to the interaction terms reflects a trade-off between complexity and interpretability. The RF and ANN captured higher-order interactions (e.g., matrix $\times$ PFAS $\times$ time $\times$ space). However, the GR_{AEN} was selected for inferential conclusions because it provided a stable, interpretable set of coefficients, essential for identifying the dominant drivers in this spatially limited dataset while avoiding overfitting and accounting for multicollinearity. The complex interactions identified by the RF and ANN are not discounted; they are explored mechanistically and dynamically in Tier 3, which resolves such patterns (e.g., seasonal pulses and species-specific uptake). Thus, the tiered framework applies the right tool for each analytical purpose, ensuring that both non-linear interactions and their practical relevance are addressed.

3.4. Coupling Matrix-Specific Biogeochemical Roles

The distinct biogeochemical signatures of the matrices underscore the need for coupling the compartment-specific behaviors of PFASs: the sediment acted as a legacy sink for long-chain compounds, the BMs served as a transient reservoir for precursors, and the HSCs reflected a mixed, low-level background exposure profile. As illustrated in Figure 2, the precursor cluster (e.g., 5:3 FTCA and 6:2 FTS) appeared exclusively in the BMs, while the background cluster (legacy PFOA and PFOS) appeared primarily in the sediment and HSCs.

This integrated analysis indicates that the PFAS fate in the estuary is governed by PFAS-specific properties interacting with matrix-specific exposure pathways. The longitudinal correlations highlight how mobile compounds (PFHxA) follow the surface plume trajectories affecting BMs, while benthic processes affecting HSCs create opposing spatial gradients and class-specific accumulation patterns. Together, these findings underscore the necessity of multi-compartmental monitoring and modeling to capture the complex spatiotemporal dynamics of PFAS contamination following diffuse and point-source releases.

S1 (3.5 km moderately east of the spill) received the strongest initial pulse for most PFASs, including the highly mobile short-chain PFHxA. For the long-chain sediment-bound PFOS, S1 showed the highest initial concentrations, with subsequent downstream redistribution leading to delayed accumulation at S3. PFOA, with intermediate sorptivity, was largely restricted to the proximal S1. S2 (5.0 km more easterly but farther south) showed the highest PFHxA in the BMs; strong water column-driven precursor–product dynamics, such as PFOSA; and led to slightly delayed or attenuated signals for PFOS. S3 (5.9 km most westerly) had the weakest and most delayed water column signals and showed elevated PFHxA in the HSCs but became a sink for sediment-redistributed PFOS. The significant interaction effect between the matrix and PFAS types found in the GLMM underscores

the need for a process-based (mechanistic) understanding of biogeochemically coupled interactions and feedback loops among multiple PFAS pools, influxes, and outfluxes.

3.5. Biogeochemically Coupled Cycles of PFOS and 5:3 FTCA

The system dynamics model provides a mechanistic explanation for the empirically divergent behaviors observed among the matrices (Figure 3). As detailed in Tables S2 and S3, the model was parameterized with field-calibrated rate constants and state variables. The spring 2025 5:3 FTCA pulse detected exclusively in the BMs (Figure 3) emerges naturally from the model as a consequence of (i) seasonally pulsed FTCA input, (ii) rapid BMs filtration uptake ($k_{up_FTCA_BM} \gg k_{up_FTCA_HSC}$), and (iii) rapid FTCA loss via degradation and depuration. Figure 3-upper panel quantifies this dichotomy: the 5:3 FTCA concentration in the BMs exhibited a sharp, high-amplitude spike synchronized with the spring water pulse, whereas the HSCs' response was minimal and delayed. This orders-of-magnitude difference in their sensitivity visually underscores why BMs serve as early-warning indicators for acute pulses, while HSCs would fail to capture such events.

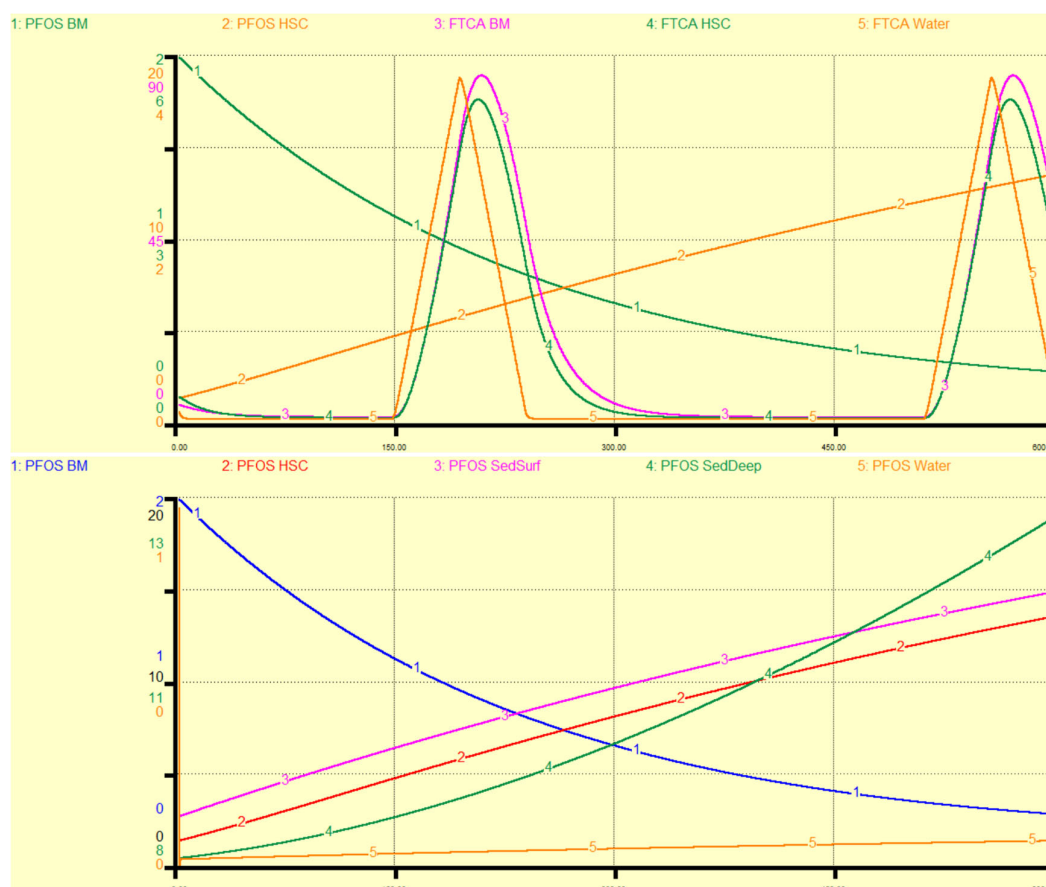


Figure 3. Simulation of dynamics of PFOS (lower panel) and 5:3 FTCA (upper panel) accumulations in estuarine water, sediment, HSCs, and BMs over 600 d (October 2024–June 2026).

In contrast, the PFOS dynamics are dominated by sediment–water exchange and extremely slow biological depuration. This decoupling of exposure pathways is visually evident in Figure 3-lower panel. The simulated PFOS concentration in the HSCs showed a steady, near-linear increase over the 600-day simulation, consistent with a slow, persistent source from sediment desorption and porewater. However, the PFOS in the BMs plateaued and gradually declined, reflecting a pool more closely tied to the initial water column pulse and its own extremely slow depuration, while the water column PFOS remained low and stable. PFOS acted as a time-integrated product of transient FTCA inputs, converting

short-lived precursor pulses into sustained contamination. This explains why the PFOS concentrations increased gradually across the monitoring period and why the HSCs continued accumulating PFOS even as the BMs' concentrations declined. The opposing temporal trends of PFOS in the BMs vs. HSCs are reproduced only when sediment-mediated uptake is included for the HSCs, confirming that benthic coupling is the dominant driver of long-term PFOS exposure.

PFOS accumulation in the BMs was modeled as direct uptake from the water column with extremely slow depuration, whereas the HSCs received PFOS from both the water and sediment. Inferred from the statistical decoupling of BMs and HSCs concentrations, the coefficient ($\alpha = 0.3$) reflected this mixed exposure pathway [7]. PFOS exchange between water and active sediment is governed by reversible sorption–desorption, while burial transfers PFOS irreversibly into deep sediment. This structure captured the long-term memory effect observed in the sediment's PFOS concentrations, independent of short-term water column pulses.

The 5:3 FTCA in the estuarine water was driven by a combination of baseline input and a pulsed input representing post-spill mobilization. The spring 5:3 FTCA pulse observed in the BMs corresponded to increased freshwater runoff and remobilization of AFFF-derived residues observed during March–June 2025 (DOY 71–162). The 5:3 FTCA accumulation in the BMs and HSCs was represented as first-order uptake from the water column, with depuration and mortality losses. These represented the rapid but transient accumulation of 5:3 FTCA in the BMs, consistent with their filter-feeding ecology, and the weaker, delayed response of the HSCs.

The system dynamics model demonstrates that the 5:3 FTCA release on 19 August 2024 in Brunswick was rapidly expressed as a transient pulse captured by the BMs, but was subsequently transformed into persistent PFOS contamination governed by sediment storage, HSCs uptake, and slow biological depuration, thus sustaining seafood exposure risk despite the declining water column concentrations. The model simulations indicate that advisory decisions based solely on water or sediment PFOS concentrations would underestimate the biologically relevant PFAS exposure risk during periods dominated by water column precursors. The BMs responded rapidly to the 5:3 FTCA and early PFOS increases, serving as early-warning indicators during pulse conditions (e.g., spring 2025). However, as the coupled 5:3 FTCA–PFOS system exhibited a transition from pulse-dominated water column dynamics to sediment-controlled persistence, consistent with the delayed equilibration and strong compartmental memory in estuarine systems, the HSCs emerged as the critical long-term exposure vector, retaining elevated PFOS for months after the water concentrations stabilized. In other words, short-lived 5:3 FTCA releases generate prolonged PFOS exposure in edible bivalves. This mechanistic insight supports a tiered advisory framework: (1) BMs for short-term advisories following episodic releases; and (2) HSCs for persistent advisories tied to sediment legacy contamination.

3.6. Model Limitations

The present study is a case-based, process-informed framework, not a universal model. Thus, the system dynamics model presented in Tier 3 is intended to provide an illustrative mechanistic consistency for the observed matrix-specific patterns, rather than to serve as a validated predictive tool. As such, the model is calibrated to be consistent with the empirical data and existing literature; however, its predictions are subject to the uncertainties discussed below. This illustrative model does not include formal sensitivity or uncertainty analyses (e.g., Monte Carlo simulation). Future iterations of this model should prioritize formal sensitivity and uncertainty analyses to quantify the robustness of the inferred processes to parameter uncertainty.

Several simplifying assumptions of our model warrant consideration for future refinement. First, all compartments were treated as spatially well mixed, neglecting fine-scale hydrodynamic gradients and microhabitat heterogeneity. This assumption was supported by the lack of significant spatial effects in the empirical data, but may underestimate localized exposure near sources. Second, uptake, depuration, and transformation processes were represented using first-order rate constants, which captured the dominant dynamics but did not resolve potential non-linearities, such as saturation kinetics or density-dependent feeding behavior. Third, the model did not explicitly resolve intermediate transformation products beyond FTCA and PFOS, and instead aggregated precursor degradation into an effective conversion pathway. Fourth, burial into deep sediment was treated as irreversible over the modeled timeframe, consistent with observed PFOS persistence, but potentially conservative with respect to long-term remobilization. Finally, since the parameter values were informed by the literature ranges and calibrated to observed trends rather than independently estimated for the site, the uncertainty propagation was not quantified. Despite these limitations, the model robustly reproduces the observed divergence among the matrices and provides a mechanistic explanation for patterns that cannot be inferred from statistical analyses and data-driven models alone.

3.7. Implications for Monitoring, Risk Assessment, and Remediation

The integration of Tiers 1–3 suggests that PFAS dynamics in the estuary are governed by biogeochemically distinct ecosystem compartments and their roles. The sediment acted as a slow but long-term sink for hydrophobic PFOS [8,9], whereas the BMs captured the transient pulses of mobile precursors and carboxylates (e.g., 5:3 FTCA and PFHxA) in the water column [10]. The co-occurrence of the precursor 6:2 FTS and its degradation product 5:3 FTCA in BMs during the spring is consistent with a recent, runoff-driven event [11], with transformation timescales (days to weeks) too short for equilibrium partitioning into sediments [9]. This pattern mirrors the seasonal PFAS dynamics observed in other temperate estuaries [12–14]; however, alternative explanations (e.g., mixture composition, transport lags, resuspension, and unmeasured intermediates) may also contribute.

These compartment-specific behaviors suggest a multi-layered understanding with implications for monitoring and risk assessment. First, the overriding influence of PFAS persistence should be considered alongside intra-annual biogeochemical cycles. Second, quantifying PFAS contamination requires moving beyond source apportionment to model spatiotemporally dynamic states, interdependencies, and (in)vulnerabilities. Third, an integrated monitoring–modeling approach is essential; reliance solely on detection rates, concentration-based metrics, or a single matrix would miss critical SEES-level dynamics. As evidenced by the strong negative correlations [15], non-redundancy of HSCs with BMs emphasizes the sampling of diverse ecosystem compartments. Regulatory frameworks prioritizing sediment monitoring alone are at elevated risk of Type II errors regarding emerging precursors. A PFAS-sampling matrix and spatiotemporal resolution should be concurrently considered to jointly optimize resource allocation, uncertainty reduction, and the trade-offs between the selection of spatiotemporal resolution and its early-warning capabilities for SEES-level risks, such as (sea)food safety; public health; and air, water, and soil quality.

4. Conclusions

While focused on a specific post-spill estuary, this integrated approach provides a case-based, process-informed framework for assessing PFAS dynamics. This study suggests that matrix-specific biogeochemical roles rather than simple spatial proximity to sources play an important role in PFAS dynamics. The sediment, BMs, and HSCs captured distinct

contamination pools, supporting multi-compartmental monitoring and modeling where the (1) sediment tracks long-term legacy trends (e.g., PFOS); (2) BMs serve as sentinels for acute water column pulses (e.g., 5:3 FTCA and mobile carboxylates); and (3) HSCs provide integrative measures of mixed exposure pathways (e.g., porewater and sediment–water exchanges). Future monitoring should prioritize functional sampling diversity across ecosystem compartments to capture the full estuarine PFAS exposome and inform effective strategies. The predictive application of the system dynamics component to other estuarine systems requires relaxing its simplifying assumptions (e.g., well-mixed compartments, first-order kinetics, and lack of formal uncertainty propagation) and site-specific calibration.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/pr14020391/s1>, Table S1: Comparison of performance and specifications of random forest (RF), artificial neural network (ANN), and generalized regression with adaptive Elastic Net (GR_{AEN}) models for predicting concentrations of the most prevalent PFAS (PFOS, PFOA, PFHxA, and PFOSA) as a function of PFAS and matrix types, DOY_{sin}, DOY_{cos}, Lat, and Lon *; Table S2: State and rate variables and parameterization of the model developed in this study for estuarine PFAS dynamics; Table S3: Differential equations and flow processes for the system-dynamics model of PFOS and 5:3 FTCA in estuarine compartments; Table S4: Parameter estimates on the log-scale from the generalized regression with adaptive Elastic Net (GR_{AEN}) model for the most prevalent PFAS ($n = 188$); Table S5: Relative predictor importance for the four prevalent PFAS concentrations (PFOS, PFOA, PFHxA, and PFOSA) according to the Generalized regression with adaptive Elastic Net (GR_{AEN}), random forest (RF), and artificial neural network (ANN) models. Refs. [16–35] are cited in Supplementary Materials.

Author Contributions: F.E.: Conceptualization, methodology, data acquisition, predictive modeling, interpretation, writing—original draft, supervision, project administration, funding acquisition. G.A.E.: Conceptualization, resources, writing—review and editing, supervision, project administration, funding acquisition. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by USDA-NACA (Project no: 5410758-1890). The authors thank Liam McNerney for his assistance with field campaigns and the Brunswick homeowners for providing access to water sources on their properties. The authors also thank the anonymous reviewers for their constructive comments, which significantly improved an earlier version of this manuscript. Any remaining errors or omissions are the sole responsibility of the authors. The findings and conclusions in this publication are those of the authors and should not be construed to represent any official USDA, EPA, or U.S. Government determination or policy.

Data Availability Statement: Further data inquiries should be directed to the corresponding author.

Conflicts of Interest: The authors declare no conflicts of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

Abbreviations

The following abbreviations are used in this study:

5:3-FTCA	5:3 fluorotelomer carboxylic acid
6:2 FTS	6:2 fluorotelomer sulfonate
AIC _c	Akaike Information Criterion corrected
ANN	Artificial neural network
AFFF	Aqueous film-forming foam
BMs	Blue mussels (<i>Mytilus edulis</i>)
CI	Confidence interval
CRA	Categorical response analysis
DOY	Day of year

DOY _{cos}	Cosine-transformed day of year (seasonal harmonic)
DOY _{sin}	Sine-transformed day of year (seasonal harmonic)
DR	Detection rate
DW	Dry weight
FDR	False discovery rate (Benjamini–Hochberg correction)
GLMM	Generalized linear mixed model
GR _{AEN}	Generalized regression with adaptive Elastic Net
HSCs	Hardshell clams (<i>Mercenaria mercenaria</i>)
Lat	Latitude
Lon	Longitude
<i>n</i>	Sample size
NDs	Non-detects
PFASs	Per- and polyfluoroalkyl substances
PFHxA	Perfluorohexanoic acid
PFHxS	Perfluorohexanesulfonic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonate
PFOSA	Perfluorooctanesulfonamide
<i>p</i> -value	Statistical significance
<i>q</i> -value	Adjusted <i>p</i> -value after FDR correction
<i>R</i> ²	Coefficient of determination
RF	Random forest
RMSE	Root mean square error
S1, S2, S3	Sampling locations in the Harpswell Cove estuary
Spearman's ρ	Spearman's correlation coefficient
STELLA [®]	Software for system dynamics modeling
ΣPFAS	Sum of all 40 targeted PFAS concentrations
WW	Wet weight

References

1. Buck, R.C.; Franklin, J.; Berger, U.; Conder, J.M.; Cousins, I.T.; de Voogt, P.; Jensen, A.A.; Kannan, K.; Mabury, S.A.; van Leeuwen, S.P.J. Perfluoroalkyl and polyfluoroalkyl substances in the environment: Terminology, classification, and origins. *Integr. Environ. Assess. Manag.* **2011**, *7*, 513–541. [\[CrossRef\]](#)
2. Ahrens, L.; Bundschuh, M. Fate and effects of poly- and perfluoroalkyl substances in the aquatic environment: A review. *Environ. Toxicol. Chem.* **2014**, *33*, 1921–1929. [\[CrossRef\]](#)
3. Brendel, S.; Fetter, É.; Staude, C.; Vierke, L.; Biegel-Engler, A. Short-chain perfluoroalkyl acids: Environmental concerns and a regulatory strategy under REACH. *Environ. Sci. Eur.* **2018**, *30*, 9. [\[CrossRef\]](#)
4. Eriksson, U.; Kärrman, A.; Rotander, A.; Mikkelsen, B.; Dam, M. Perfluoroalkyl substances (PFASs) in food and water from Faroe Islands. *Environ. Sci. Pollut. Res.* **2013**, *20*, 7940–7948. [\[CrossRef\]](#)
5. Federal Register. Draft National Recommended Ambient Water Quality Criteria for the Protection of Human Health for Perfluorooctanoic Acid, Perfluorooctane Sulfonic Acid, and Perfluorobutane Sulfonic Acid. *Fed. Regist.* **2025**, *90*, 1–50. Available online: <https://www.federalregister.gov/documents/2024/12/26/2024-30637/draft-national-recommended-ambient-water-quality-criteria-for-the-protection-of-human-health-for> (accessed on 1 December 2025).
6. Hannan, M.; Evrendilek, F.; Leclair, D.; Choudhary, M.; Mensah, K.; Aeppli, C.; Venkatesan, A.K.; Apul, O.G. Aftermath of a major firefighting foam spill in Brunswick, Maine: Spatiotemporal dynamics of per-and polyfluoroalkyl substances in the downstream surface waters. *J. Hazard. Mater. Lett.* **2025**, *6*, 100150. [\[CrossRef\]](#)
7. Lenihan, H.S.; Peterson, C.H.; Byers, J.E.; Grabowski, J.H.; Thayer, G.W.; Colby, D.R. Cascading of habitat degradation: Oyster reefs invaded by refugee fishes escaping stress. *Ecol. Appl.* **2001**, *11*, 764–782. [\[CrossRef\]](#)
8. Ahrens, L.; Taniyasu, S.; Yeung, L.W.; Yamashita, N.; Lam, P.K.; Ebinghaus, R. Distribution of polyfluoroalkyl compounds in water, suspended particulate matter and sediment from Tokyo Bay, Japan. *Chemosphere* **2010**, *79*, 266–272. [\[CrossRef\]](#) [\[PubMed\]](#)
9. Munoz, G.; Desrosiers, M.; Duy, S.V.; Labadie, P.; Budzinski, H.; Liu, J.; Sauvé, S. Environmental occurrence of perfluoroalkyl acids and novel fluorotelomer surfactants in the freshwater fish *Catostomus commersonii* and sediments following firefighting foam deployment at the Lac-Mégantic railway accident. *Environ. Sci. Technol.* **2017**, *51*, 1231–1240. [\[CrossRef\]](#)

10. Ng, C.A.; Hungerbühler, K. Bioaccumulation of perfluorinated alkyl acids: Observations and models. *Environ. Sci. Technol.* **2014**, *48*, 4637–4648. [[CrossRef](#)]
11. Wang, N.; Liu, J.; Buck, R.C.; Korzeniowski, S.H.; Wolstenholme, B.W.; Folsom, P.W.; Sulecki, L.M. 6: 2 Fluorotelomer sulfonate aerobic biotransformation in activated sludge of waste water treatment plants. *Chemosphere* **2011**, *82*, 853–858. [[CrossRef](#)]
12. Teymoorian, T.; Munoz, G.; Duy, S.V.; Vaudreuil, M.A.; Liu, M.; Liu, J.; Sauvé, S. A province-wide mapping of per- and polyfluoroalkyl substances (PFAS) in surface waters of the St. Lawrence River watershed, Québec, Canada. *Sci. Total Environ.* **2025**, *1000*, 180429. [[CrossRef](#)] [[PubMed](#)]
13. Widdows, J.; Donkin, P.; Staff, F.J.; Matthiessen, P.; Law, R.J.; Allen, Y.T.; Thain, J.E.; Allchin, C.R.; Jones, B.R. Measurement of stress effects (scope for growth) and contaminant levels in mussels (*Mytilus edulis*) collected from the Irish Sea. *Mar. Environ. Res.* **2002**, *53*, 327–356. [[CrossRef](#)]
14. U.S. Environmental Protection Agency (EPA). *Drinking Water Health Advisory for Perfluorooctane Sulfonate (PFOS)*; Office of Water Document 822-R-16-004; U.S. EPA: Washington, DC, USA, 2016.
15. Widdows, J.; Burns, K.A.; Menon, N.R.; Page, D.S.; Soria, S. Measurement of physiological energetics (scope for growth) and chemical contaminants in mussels (*Arca zebra*) transplanted along a contamination gradient in Bermuda. *J. Exp. Mar. Biol. Ecol.* **1990**, *138*, 99–117. [[CrossRef](#)]
16. Chen, H.; Reinhard, M.; Nguyen, V.T.; Gin, K.Y.H. Reversible and irreversible sorption of perfluorinated compounds (PFCs) by sediments of an urban reservoir. *Chemosphere* **2016**, *144*, 1747–1753. [[CrossRef](#)] [[PubMed](#)]
17. Zhao, Z.; Xie, Z.; Tang, J.; Sturm, R.; Chen, Y.; Zhang, G.; Ebinghaus, R. Seasonal variations and spatial distributions of perfluoroalkyl substances in the rivers Elbe and lower Weser and the North Sea. *Chemosphere* **2015**, *129*, 118–125. [[CrossRef](#)] [[PubMed](#)]
18. Conder, J.M.; Hoke, R.A.; Wolf, W.D.; Russell, M.H.; Buck, R.C. Are PFCAs bioaccumulative? A critical review and comparison with regulatory criteria and persistent lipophilic compounds. *Environ. Sci. Technol.* **2008**, *42*, 995–1003. [[CrossRef](#)]
19. Lee, H. Models, muddles, and mud: Predicting bioaccumulation of sediment-associated pollutants. In *Sediment Toxicity Assessment*; CRC Press: Boca Raton, FL, USA, 2018; pp. 267–293.
20. Riisgård, H.U. On measurement of filtration rates in bivalves—The stony road to reliable data: Review and interpretation. *Mar. Ecol. Prog. Ser.* **2001**, *211*, 275–291. [[CrossRef](#)]
21. Hong, S.; Khim, J.S.; Wang, T.; Naile, J.E.; Park, J.; Kwon, B.O.; Song, S.J.; Ryu, J.; Codling, G.; Jones, P.D.; et al. Bioaccumulation characteristics of perfluoroalkyl acids (PFAAs) in coastal organisms from the west coast of South Korea. *Chemosphere* **2015**, *129*, 157–163. [[CrossRef](#)]
22. Arnot, J.A.; Gobas, F.A. A review of bioconcentration factor (BCF) and bioaccumulation factor (BAF) assessments for organic chemicals in aquatic organisms. *Environ. Rev.* **2006**, *14*, 257–297. [[CrossRef](#)]
23. Barron, M.G. Bioaccumulation and bioconcentration in aquatic organisms. In *Handbook of Ecotoxicology*; Hoffman, D.J., Rattner, B.A., Burton, G.A., Jr., Cairns, J., Jr., Eds.; Lewis Publishers: Boca Raton, FL, USA, 1995; pp. 652–666.
24. Norkko, A.; Thrush, S.F.; Cummings, V.J.; Gibbs, M.M.; Andrew, N.L.; Norkko, J.; Schwarz, A.M. Trophic structure of coastal Antarctic food webs associated with changes in sea ice and food supply. *Ecology* **2007**, *88*, 2810–2820. [[CrossRef](#)]
25. Higgins, C.P.; Luthy, R.G. Sorption of perfluorinated surfactants on sediments. *Environ. Sci. Technol.* **2006**, *40*, 7251–7256. [[CrossRef](#)]
26. Zareitalabad, P.; Siemens, J.; Hamer, M.; Amelung, W. Perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS) in surface waters, sediments, soils and wastewater—A review on concentrations and distribution coefficients. *Chemosphere* **2013**, *91*, 725–732. [[CrossRef](#)]
27. Baskaran, M.; Miller, C.J.; Kumar, A.; Andersen, E.; Hui, J.; Selegue, J.P.; Creech, C.T.; Barkach, J. Sediment accumulation rates and sediment dynamics using five different methods in a well-constrained impoundment: Case study from Union Lake, Michigan. *J. Great Lakes Res.* **2015**, *41*, 607–617. [[CrossRef](#)]
28. Armitage, J.M.; MacLeod, M.; Cousins, I.T. Modeling the global fate and transport of perfluorooctanoic acid (PFOA) and perfluorooctanoate (PFO) emitted from direct sources using a multispecies mass balance model. *Environ. Sci. Technol.* **2009**, *43*, 1134–1140. [[CrossRef](#)]
29. Smaal, A.C.; Schellekens, T.; van Stralen, M.R.; Kromkamp, J.C. Decrease of the carrying capacity of the Oosterschelde estuary (SW Delta, NL) for bivalve filter feeders due to overgrazing? *Aquaculture* **2013**, *404*, 28–34. [[CrossRef](#)]
30. Grizzle, R.E.; Langan, R.; Howell, W.H. Growth responses of suspension-feeding bivalve molluscs to changes in water flow: Differences between siphonate and nonsiphonate taxa. *J. Exp. Mar. Biol. Ecol.* **1992**, *162*, 213–228. [[CrossRef](#)]
31. Peterson, C.H.; Beal, B.F. Bivalve growth and higher order interactions: Importance of density, site, and time. *Ecology* **1989**, *70*, 1390–1404. [[CrossRef](#)]
32. Labadie, P.; Chevreuil, M. Partitioning behaviour of perfluorinated alkyl contaminants between water, sediment and fish in the Orge River (nearby Paris, France). *Environ. Pollut.* **2011**, *159*, 391–397. [[CrossRef](#)]

33. Gustafsson, Ö.; Haghseta, F.; Chan, C.; MacFarlane, J.; Gschwend, P.M. Quantification of the dilute sedimentary soot phase: Implications for PAH speciation and bioavailability. *Environ. Sci. Technol.* **1996**, *31*, 203–209. [[CrossRef](#)]
34. Liu, J.; Wang, N.; Buck, R.C.; Wolstenholme, B.W.; Folsom, P.W.; Sulecki, L.M.; Bellin, C.A. Aerobic biodegradation of [14C] 6: 2 fluorotelomer alcohol in a flow-through soil incubation system. *Chemosphere* **2010**, *80*, 716–723. [[CrossRef](#)] [[PubMed](#)]
35. Hawkins, A.J.S.; Pascoe, P.L.; Parry, H.; Brinsley, M.; Black, K.D.; McGonigle, C.; Moore, H.; Newell, C.R.; O’Boyle, N.; Ocarroll, T.; et al. Shellsim: A generic model of growth and environmental effects validated across contrasting habitats in bivalve shellfish. *J. Shellfish Res.* **2013**, *32*, 237–253. [[CrossRef](#)]

Disclaimer/Publisher’s Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.