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Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Unique *per*- and polyfluoroalkyl substances (PFAS) source suggested by a Lake Trout (*Salvelinus namaycush*) PFAS profile in a temperate lake

Eric Levanduski^a, Susan F. Cushman^b, Lisa B. Cleckner^c, Wayne Richter^{d,1}, Jesse C. Becker^d, Trevor Massey^c, Jacques Rinchard^e, N. Roxanna Razavi^{a,*}

^a Department of Environmental Biology, State University of New York College of Environmental Science and Forestry, Syracuse, NY 13210, United States

^b Department of Biology, Hobart and William Smith Colleges, Geneva, NY 14456, United States

^c Finger Lakes Institute, Hobart and William Smith Colleges, Geneva, NY 14456, United States

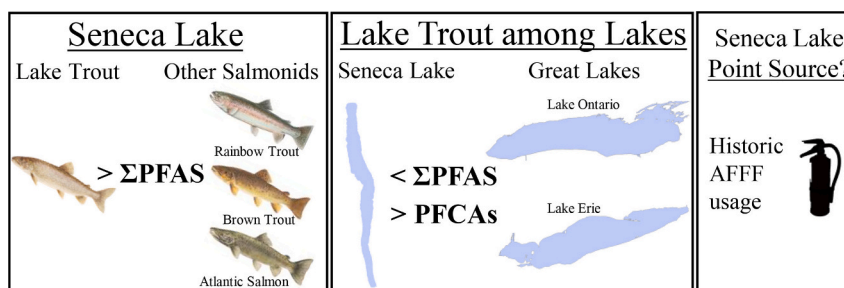
^d Division of Fish and Wildlife, New York State Department of Environmental Conservation, Albany, NY 12233, United States

^e Department of Environmental Science and Ecology, State University of New York Brockport, Brockport, NY 14420, United States

HIGHLIGHTS

- Lake Trout had higher \sum PFAS concentrations than other sportfish.
- 5 PFAS compounds were only found in Lake Trout sportfish (e.g., PFHxA and PFPeA).
- Lake Trout in Seneca Lake had higher \sum PFAS but lower PFOS percent than other lakes.
- PFNA and PFDoA were elevated in Lake Trout from Seneca Lake compared to other lakes.
- Previous use of AFFF in the Seneca Lake watershed is a suspected PFAS source.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Jay Gan

Keywords:

PFOS
Mercury
Bioaccumulation
AFFF
Laurentian Great Lakes
Finger Lakes

ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are detected in pelagic freshwater fish and have deleterious effects on their health. It is unclear if traditional proxies for uptake of contaminants in fish (e.g., length, weight, age) predict fish PFAS concentrations. Here, we observe that summed PFAS concentrations are significantly higher in Lake Trout (*Salvelinus namaycush*) than other sportfish in Seneca Lake, New York. Carbon source (as proxied by $\delta^{13}\text{C}$) predicted variability within species, and trophic level (as proxied by $\delta^{15}\text{N}$) trended among species. A moderate inverse correlation ($r = -0.51$) was found between mercury and summed PFAS in Lake Trout. Summed PFAS concentrations and length, weight, or age were not statistically related, suggesting these characteristics are not reliable proxies for PFAS bioaccumulation. Length, weight, and age were significant predictors for mercury, indicating these drivers may be resulting in differential bioaccumulation in PFAS and mercury. In Seneca Lake, a unique PFAS composition was found for Lake Trout, where PFOS represents a lower proportion of summed PFAS than in other species in Seneca Lake, as well as relative to Lake Trout from other neighboring Finger Lakes. In addition, compared to Lake Erie and Lake Ontario, Lake Trout from Seneca Lake have higher concentrations of PFOA, PFNA, and PFDA, but lower proportions of PFOS. Lake Trout from Seneca Lake have a PFAS composition that consists almost exclusively of perfluoroalkyl carboxylic acids (PFCAs) and perfluorosulfonic acids (PFSAs),

* Corresponding author.

E-mail address: razavi@esf.edu (N.R. Razavi).

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<https://doi.org/10.1016/j.scitotenv.2025.179038>

Received 24 December 2024; Received in revised form 27 February 2025; Accepted 1 March 2025

Available online 11 March 2025

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similar to the composition used in aqueous film forming foams (AFFF) before 2000 at a former military and current Superfund site in the lake's watershed.

1. Introduction

Sportfish are an important component of a lake ecosystem as top predators (Vanni et al., 1997) and provide a means of acquiring healthy protein for humans along with other social and cultural benefits (Lynch et al., 2016; Turyk et al., 2012). However, contaminants such as *per*- and polyfluoroalkyl substances (PFAS) and mercury (Hg) can pose a threat to the health of sportfish, and subsequently to human and wildlife health through consumption (Ankley et al., 2020; Scheuhammer et al., 2007). Both Hg and some PFAS are persistent, bioaccumulative, and toxic substances. Mercury is known to be a ubiquitous neurotoxicant of concern (Kumari and Chand, 2023). In terms of persistence among legacy and emerging contaminants, Cousins et al. (2022) argue that some PFAS (e.g., perfluorooctanesulfonic acid (PFOS) and perfluorononanoic acid (PFNA) are even the most persistent contaminants in existence. Characterizing concentrations of both legacy and emerging contaminants, as well as the beneficial fatty acids, in fish thus remains an important research goal for human and ecosystem health.

PFAS are a group of >9000 man-made fluorinated chemicals with a diverse structure. The group is defined by a common fully fluorinated (perfluoroalkyl substances) or multi-fluorinated (polyfluoroalkyl substances) bond (Buck et al., 2011). PFAS are used widely in consumer and industrial products because of their water-repellent and low surface tension properties (Glüge et al., 2020). Common point sources for PFAS exposure in wildlife include wastewater treatment plants, landfill leachate runoff, and aqueous film forming foam (AFFF) use and disposal (Conder et al., 2021; Paige et al., 2024). Fish exposure to certain PFAS compounds can cause negative health effects, including immunosuppression, inflammation, reduced reproduction, and death at high concentrations (Guo et al., 2019, Suski et al., 2021, Zhong et al., 2020). Bioaccumulation and trophic transfer of some PFAS has been observed, with long chain PFAS considered bioaccumulative ($\text{Concentration}_{\text{biota}}/\text{Concentration}_{\text{water}} > 1$), including PFOS, PFNA, perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUNA), and perfluorododecanoic acid (PFDoA) (Burkhard, 2021). Biomagnification ($\text{Concentration}_{\text{predator}}/\text{Concentration}_{\text{prey}} > 1$) of PFOS was recently observed in Lake Michigan fish across all predator-prey relationships and highest in Lake Trout (*Salvelinus namaycush*) among the species tested (Miranda et al., 2023). Other long-chain PFAS compounds, including PFOA, PFNA, and PFDA were also found to biomagnify in Lake Michigan fishes (Miranda et al., 2023). Direct discharge and atmospheric deposition are two main drivers for increased PFAS concentrations in waterbodies, but few other waterbody parameters have been determined (Podder et al., 2021).

Mercury is a global pollutant that has been detected in freshwater fisheries, including the New York Finger Lakes and its tributaries (Razavi et al., 2019, 2020). Lake Trout from Seneca Lake have previously exhibited total Hg concentrations ranging from 208 to 703 nanograms/g wet weight (ng/g ww; Razavi et al., 2020). In waterbodies without a point source, atmospheric Hg concentrations drive fish Hg concentrations (Gandhi et al., 2014; Harris et al., 2007), and regulations on emissions have been shown to decrease Hg in Great Lakes fish (Zhou et al., 2016). However, Hg bioaccumulation is also strongly affected by water quality parameters including pH and dissolved organic carbon (Brumbaugh et al., 2001), as well as fish diet and growth dynamics (e.g., Lepak et al., 2019), which can decouple Hg trends in fish from atmospheric emissions.

Stable isotope analysis has been used to track trophic transfer of contaminants in biota, due to ease of use and breadth of information provided (Fry, 2006). $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ stable isotopes are used as a proxy for carbon source and trophic level, respectively (Post, 2002). When

taken from different organs, stable isotopes can represent different life stages for the organism, with stable isotopes derived from muscle tissue representing a short-term scope of the carbon source and trophic level of the organism. These analyses can be used to track biomagnification and food web dynamics (Layman et al., 2011). The use of this tool will help further elucidate controls on trophic transfer of PFAS.

To date, no published studies of PFAS concentrations in pelagic fish exist from the Finger Lakes. Our objectives were to analyze PFAS concentrations in Seneca Lake sportfish, and determine if carbon source, trophic level, and/or size (i.e., length and weight) predict PFAS concentrations. We also measured Hg concentrations as a point of comparison to PFAS concentrations observed here. Comparing bioaccumulation of PFAS and a well-studied legacy contaminant can help confirm the accuracy of potential predictor results. The benefit of consuming Lake Trout from Seneca Lake was assessed using fatty acid composition. Finally, we compared PFAS concentration and composition from Seneca Lake to other Finger Lakes and Great Lakes within New York State to better understand trends and possible point sources.

2. Material and methods

2.1. Sampling location

Seneca Lake is one of the largest Finger Lakes (Fig. 1), providing drinking water to approximately 100,000 people in upstate New York (Callinan, 2001). Seneca Lake is also a popular freshwater fishery, especially for trout and other salmonids. The annual National Lake Trout Fish Derby on Seneca Lake draws hundreds of anglers. This study occurred during the 58th year of the Derby, which included >600 participants (Lake Trout Derby, 2022), and is estimated to provide a large economic benefit to the Finger Lakes region through tourism activity (Lynch et al., 2016, Roost., 2024).

Seneca Lake is a moraine lake formed by glacial movement, and includes many native and introduced species, such as trout (Lake Trout; Brown Trout, *Salmo trutta*; and Rainbow Trout, *Oncorhynchus mykiss*), salmon (Landlocked Atlantic Salmon (hereafter Landlocked Salmon), *Salmo salar*), black bass (Largemouth Bass, *Micropterus nigricans*; Smallmouth Bass, *M. dolomieu*), Yellow Perch (*Perca flavescens*), Alewife (*Alosa pseudoharengus*), and Sea Lamprey (*Petromyzon marinus*). The top predators in Seneca Lake include salmon and trout, consuming smaller fishes and invertebrates (NYSDEC, 2025a).

2.2. Fish collection

Sampling was performed at the 2022 National Lake Trout Derby at Seneca Lake. Anglers brought deceased landed fish for length and weight measurements to derby stations and were asked for tissue donations. From each fish, the head and a small (2" x 2" (5.08 cm x 5.08 cm) piece of dorsal muscle) fillet were requested to be donated for analysis. Fillet samples were requested rather than whole-fish as only fillet is typically eaten of the donated species. Only a subsample was taken from the fillet because many anglers planned to consume the remaining fillets. Seventy-three samples were donated (53 Lake Trout, 5 Landlocked Salmon, 9 Brown Trout, and 6 Rainbow Trout) at two locations: Stivers Seneca Marine in Geneva, NY (north end of lake) and Watkins Glen Marina in Watkins Glen, NY (south end of lake) (Fig. 1).

Fillet samples were portioned in halves, one half for PFAS analysis and the other to be freeze dried for Hg and stable isotope analyses. Twenty-nine Lake Trout were analyzed for PFAS and 27 for Hg, and all samples from other species were analyzed for both PFAS and Hg. Not all Lake Trout were analyzed for PFAS, given comparable data are already

available through the New York State Department of Environmental Conservation (NYSDEC).

The sampling protocol used follows the NYSDEC guidelines for PFAS sampling in biota (NYSDEC, 2023a). Samples were obtained by volunteers wearing nitrile gloves, avoiding clothing containing polytetrafluoroethylene materials to reduce risk of contamination. Fish samples were gutted, heads and fillet removed with a washed knife on a marble cutting board, cleaned between samples, and placed into high density polyethylene bags on ice. Samples were then frozen at $-20\text{ }^{\circ}\text{C}$ until analysis, with all PFAS and Hg analyses occurring within 1 year of obtaining the samples.

Analyzed fish results from the NYSDEC, which include Lake Trout from Seneca Lake and comparison waters, were collected in the same manner as described (NYSDEC, 2023a). NYSDEC captured Lake Trout in Lake Erie ($n = 5$) and Lake Ontario ($n = 12$) in 2010, Cayuga Lake ($n = 20$) and Canadice Lake ($n = 22$) in 2019, and Seneca Lake ($n = 24$) in 2020. For Lake Trout concentration comparisons among waterbodies, National Lake Trout Derby data and NYSDEC data were combined for Seneca Lake. Statistical tests compared Lake Trout from Seneca Lake collected by the NYSDEC and the present study, and Lake Trout were deemed comparable by PFAS concentration and therefore combined by summed PFAS concentration (Supplemental Fig. 1). We used the Great Lakes to compare to Seneca Lake, despite being different in size, because they are relatively close in proximity, including within the Lake Ontario watershed, have similar fish species composition, and have published PFAS data to compare to.

2.3. Chemical standards and reagents

Native linear perfluoroalkyl carboxylic acids standards, sulfonates (PFAC-MXF, PFAC-MXG, PFAC-MXH, PFAC-MXI, and PFAC-MXJ), and isotopically labeled internal standards (MPFAC-HIF-ES and MPFAC-HIF-IS) were purchased from Wellington Laboratories Inc. (Guelph, Ontario). ENVI-Carb, methanol, acetonitrile, potassium hydroxide, ammonium acetate, and ammonium hydroxide were purchased from Thermo Fisher Scientific (Waltham, MA). Oasis weak anion exchange solid phase extraction (SPE) cartridges (3 cc, 60 mg sorbent, 30 μm particle size) were purchased from Waters Corporation (Milford, MA).

2.4. Sample analysis

2.4.1. Solid phase extraction (SPE) and liquid chromatography-tandem mass spectrometry (LC-MS/MS)

Analyses were conducted at the SUNY ESF Center for Contaminants of Emerging Concern core facility (Syracuse, NY). EPA Method 533 (US EPA, 2019) for PFAS cleanup modified for fish tissue was followed, while using the draft EPA Method 1633 standards for analysis of 40 PFAS compounds (US EPA, 2021) (Supplemental Table 1). Muscle tissue samples were scaled and homogenized, skin on, in a liquid chromatography-mass spectrometry (LC-MS) grade methanol washed NutriBullet until homogenous. Homogenized fillets ($1.00 \pm 0.05\text{ g}$) from each fish sample received 100 μL of methanol and 20 μL of isotope dilution spike standard, then were vortexed and allowed to equilibrate for 30 min. Potassium hydroxide (9.8 mL) was added to each sample, vortexed, and bath sonicated for 1 h, then rocked horizontally for 16–24 h. Samples were then centrifuged and 2.45 mL of supernatant was placed into a methanol washed 15 mL centrifuge tube containing between 25 and 35 mg of ENVI-Carb and 50 μL of glacial acetic acid, vortexed, and centrifuged. 2 mL of the resulting supernatant was placed into a 250 mL polypropylene bottle containing 98 mL of PFAS free Milli-Q water and 333 μL of glacial acetic acid, and shaken. SPE was performed with a Promochrom SPE-03 system with SPE solvents (LC-MS grade methanol, PFAS free Milli-Q water, 25 mM ammonium acetate in LC-MS grade methanol, and 0.1 % ammonium hydroxide in PFAS free Milli-Q water). Lines were primed and cleaned before and after each run with LC-MS grade methanol, and samples were extracted using weak-anion exchange cartridges and dispensed into methanol washed 20 mL scintillation vials consisting of 2 fractions; the first containing perfluorooctane sulfonamides and the latter all other tested PFAS compounds. The second fraction was evaporated using a nitrogen rotary evaporator until complete dry ($\sim 2\text{ h}$ at $35\text{ }^{\circ}\text{C}$ and 1 Torr), then reconstituted with 1 mL of the second fraction and 20 μL of recovery standard, and stored at $-20\text{ }^{\circ}\text{C}$ until LC-MS/MS procedure.

Tandem Mass Spectrometry was performed according to the draft EPA Method 1633, section 13.0 (US EPA, 2021) on a TSQ Altis Triple Quad LC-MS/MS. In short, performance checks were executed via mass and initial calibrations, then instrument and method blanks, ongoing process and recovery standards, and taurodeoxycholic acid standards were run prior to samples. Recoveries of native and isotopically labeled compounds for calibration must be within 70–130 % and 50–200 % respectively to continue. Four PFAS compounds failed QA/QC and were

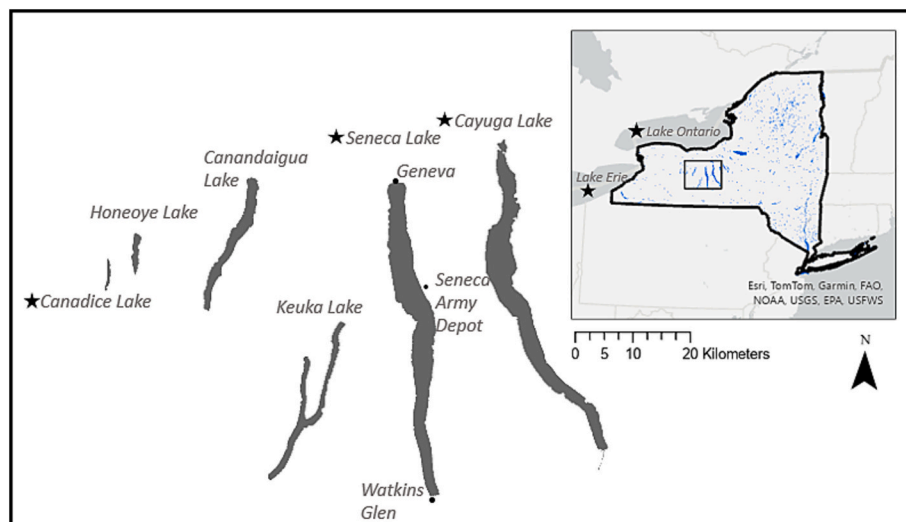


Fig. 1. Fish collection locations in the Finger Lakes region of New York. The two weigh-in stations for the National Seneca Lake Trout Derby were Seneca Stivers Marine in Geneva, NY, and Watkins Glen Marina in Watkins Glen, NY, at the northern and southern ends of Seneca Lake, respectively. Waterbodies with stars next to their names were sampled by the New York State Department of Environmental Conservation (NYSDEC).

not included in further analyses—perfluorobutanoic acid (PFBA), 3-perfluoropropyl propanoic acid (3:3 FTCA), 2H,2H,3H,3H-perfluorooctanoic acid (5:3 FTCA), and 3-perfluoroheptyl propanoic acid (7:3 FTCA). None of these compounds had detectable peaks; however, we note that draft EPA Method 1633 was not optimized for these compounds. Every ten samples, calibration verification standards (range = 100–135 % recoveries, $n = 6$ for each compound) and instrument blanks (0 % detection on all blanks) were run. Percent recoveries were determined for external controls of 1947 Lake Michigan Fish Tissue SRM (NIST) compared to their standards for every run, with a 70–130 % recovery deemed acceptable for use (range = 109–114 % recovery, $n = 6$ for each compound). Samples were analyzed using the TraceFinder v5.1 software, with the criteria outlined in sections 15.1.1–15.1.5 of draft EPA Method 1633 (US EPA, 2021).

2.4.2. Total Hg analyses

Mercury analyses were conducted using thermal decomposition, amalgamation and atomic absorption spectrophotometry (Milestone DMA-80), based on EPA Method 7473 (US EPA, 1998). At least 10 % of samples were analyzed in triplicate (range = 1.3–20.7 % and average = 8.4 % coefficient of variation [CV], $n = 8$). A minimum of a four-point standard curve was run every day of analysis, and blanks were analyzed every five samples. Standards and certified reference materials were analyzed every five samples. For quality assurance/quality control (QA/QC), the recovery of DORM-4 (fish protein, National Research Council of Canada) was 96 % (395 ± 16 ng/g dw, $n = 25$). Fish Hg concentrations were converted to wet weight (ww) for comparisons to risk thresholds using percent moisture calculated for each individual fish after freeze-drying plug samples.

2.4.3. Fatty acid analyses

Twenty Lake Trout samples from Seneca Lake were collected independently of the National Lake Trout Derby on Seneca Lake samples and analyzed for fatty acid compositions in 2017 ($n = 10$) and 2023 ($n = 10$). These samples did not differ by length (Supplemental Fig. 2b) nor by weight (Supplemental Fig. 2c) between years, and did not differ by length (Supplemental Fig. 2d) nor weight (Supplemental Fig. 2e) between fatty acid analysis and Lake Trout Derby samples. Lipids were extracted from muscle using a 2:1 chloroform/methanol solvent containing 0.01 % butylated hydroxytoluene to prevent lipid oxidation according to the method of Folch et al. (1957) and measured gravimetrically. After lipid extraction, a known amount of nonadecanoic acid (19:0, Nu-Check Prep Inc., Elysian, MN) was added to each sample based on the amount of total lipids present (8 mg/50 mg of lipids) to serve as an internal standard. Fatty acid methyl esters (FAMES) were then prepared following the method of Metcalfe and Schmitz (1961). FAMES were separated using an Agilent Technologies 7890 A gas chromatograph system with Agilent Technologies 7693 autosampler and Agilent Technologies 5975C inert XL EI/CI mass selective detector with a triple-axis detector (Agilent Technologies). An Agilent J & WGC column (30 m \times 0.250 mm \times 0.50 μ m thickness) was used with helium as the carrier gas. Run conditions were set following Happel et al. (2017). Individual FAMES were identified using fragment ions and by comparing their retention times to those of authentic standard mixtures with known spectrographic patterns of FAMES. The proportions of individual FAMES relative to the total amount of FAMES detected were determined. Fatty acids are written in International Union of Pure and Applied Chemistry nomenclature—carbon chain length:number of double bonds and the first double bond (n-x) position relative to the terminal methyl group.

2.4.4. Otolith aging

Otoliths were extracted from the heads of fishes and cracked in half to expose age rings. Otoliths were then cleaned using forceps and rings counted on two otoliths for each fish to confirm the age. Otolith ages were measured by two people to secondarily confirm the ages are correct, and if there was a discrepancy, a third person aged and the most

commonly counted age was used. Age was assessed from Lake Trout ($n = 26$), Landlocked Salmon ($n = 4$), Brown Trout ($n = 9$), and Rainbow Trout ($n = 6$) otoliths.

2.4.5. Stable isotope analyses

A small portion of freeze-dried muscle tissue (1.2–1.5 mg) was cut from the muscle fillet subsample used for Hg analysis. Each sample was packaged into tin capsules for $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ signature analysis ($n = 49$). Duplicates were analyzed for 10.2 % of samples ($n = 5$; average RPD for $\delta^{15}\text{N} = 1.0$ %, min = 0.6 %, max = 1.9 %; average RPD for $\delta^{13}\text{C} = 1.7$ %, min = 0.3 %, max = 2.6 %). Analyses were conducted at the Cornell University Stable Isotope Lab (Ithaca, NY) using a Thermo Delta V isotope mass spectrometer and a NC2500 elemental analyzer. Stable isotope values were measured in per mil (‰) using the following equation: $\delta X = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 1000$. For carbon isotopes, X is the ^{13}C , R_{sample} is the ratio of $^{13}\text{C}/^{12}\text{C}$ in the sample, and R_{standard} is the ratio of $^{13}\text{C}/^{12}\text{C}$ in Vienna Pee Dee Belemnite. For nitrogen isotopes, X is the ^{15}N , R_{sample} is the ratio of $^{15}\text{N}/^{14}\text{N}$ in the sample, and R_{standard} is the ratio of $^{15}\text{N}/^{14}\text{N}$ in atmospheric air. Quality control was performed prior to analyses and every 10 samples with methionine to test for the instrument's linearity and response, and animal standards (Deer and CBT) for normalization and precision.

2.5. Statistical analysis

All analyses were conducted in R version 4.3.0 (R Core Team) with tidyverse (Wickham et al., 2019) and ggcorrplot (Kassambara, 2022) packages.

PFAS data are rarely normally distributed due to a high frequency of non-detects, and data did not pass the Shapiro-Wilks normality test. Non-parametric Kruskal Wallis with Wilcoxon post-hoc analysis was used in place of ANOVA for PFAS analyses only. When data were normally distributed, either a Student's *t*-test or ANOVA testing with post-hoc Tukey Honestly Significant Difference testing was used. Significance is shown by asterisks denoted as follows: $p < 0.05$ with *, $p < 0.01$ with **, $p < 0.001$ with ***, and $p < 0.0001$ with ****, with $\alpha = 0.05$ for all analyses. Linear regression analyses were conducted between summed PFAS and other metrics (i.e., $\delta^{15}\text{N}$, $\delta^{13}\text{C}$, length, weight, and age) for both Lake Trout alone and all species together in Seneca Lake, and for Hg to the same metrics for all species. Correlation coefficients were determined by species in Seneca Lake for all continuous variables, and between PFAS and Hg among all species combined. Lake Trout were further analyzed in isolation because they had significantly higher summed PFAS concentrations compared to other sportfish, as described in 3.1.

2.6. PFAS detection limits

Method detection limits and reporting limits for National Lake Trout Derby fish were the same, and all values above the method detection limit were reported here. For samples analyzed by NYSDEC, reporting limits were used. When an individual PFAS result was below the reporting limit, 0 was used when calculating summed PFAS concentration, following Miranda et al. (2023).

3. Results and discussion

3.1. Summary of contaminant concentrations

Samples were above detection limit in 50 % (18/36) of PFAS compounds, with 14 compounds detected in Lake Trout, 9 in Brown Trout, 8 in Landlocked Salmon, and 8 in Rainbow Trout (Fig. 2a). Lake Trout had the highest mean and median concentration of all compounds when detected, and the highest PFOS concentration mean (5.73 ng/g ww), median (3.00 ng/g ww), and maximum (44.16 ng/g ww). PFOS represented a lower percentage of the summed PFAS composition in Lake

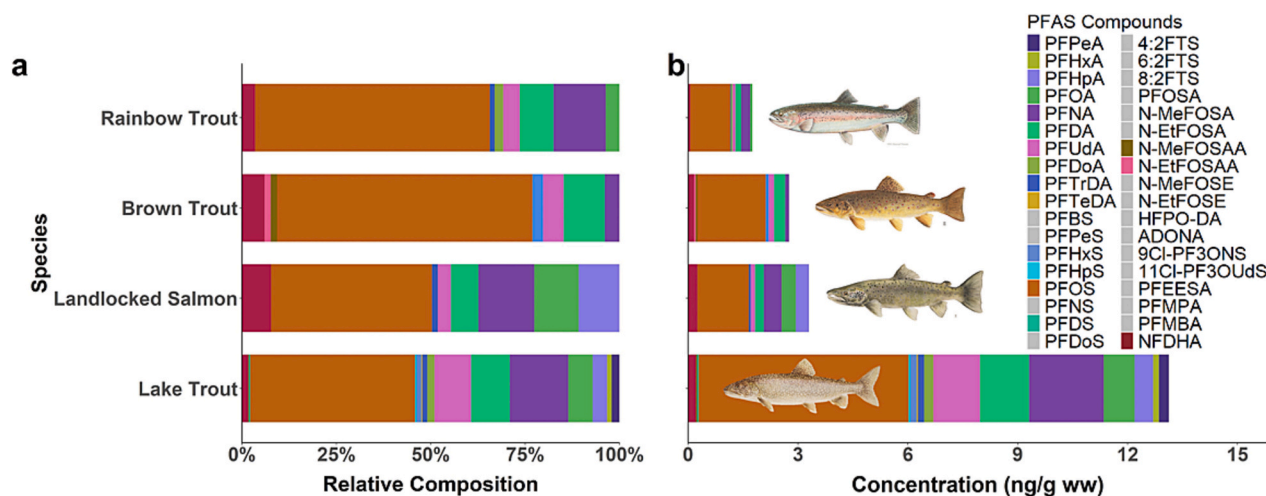


Fig. 2. a. Relative PFAS composition by species from National Lake Trout Derby at Seneca Lake. b. Mean total PFAS for each species. Compounds not detected in any sample are denoted by a gray box in the legend.

Trout (44 %) than in Brown Trout (68 %) or Rainbow Trout (62 %) but was similar to Landlocked Salmon (43 %) (Fig. 2a). PFOS was the most detected compound ($n = 45/49$), and in the highest concentration among all species. PFOA ($n = 29/49$), PFNA ($n = 34/49$), and PFDA ($n = 35/49$) were also commonly detected—similar to other studies on fish in the United States (Barbo et al., 2023, Stahl et al., 2023). Perfluoroalkyl carboxylic acid (PFCA) and perfluorosulfonic acid (PFSA) groups were the most frequently detected, with detections in 15 of 18 compounds, and in the highest concentrations. Perfluorooctane sulfonamidoacetic acids, PFOS precursors and transient degradation intermediates in electrochemical fractionation, were detected infrequently (*N*-methyl perfluorooctanesulfonamidoacetic acid (*N*-MeFOSAA): $n = 3$ and *N*-ethyl perfluorooctanesulfonamidoacetic acid (*N*-EtFOSAA): $n = 1$), potentially from past PFOS production (Buck et al., 2011).

Lake Trout had significantly higher summed PFAS concentrations than the other sportfish species tested (Fig. 2b, Supplemental Fig. 3). In Lake Michigan, Lake Trout had higher summed PFAS concentrations than Rainbow Trout (Miranda et al., 2023), further ascertaining PFAS bioaccumulation to be species specific (Babut et al., 2017; Kaboré et al., 2022; Ren et al., 2022a; Ren et al., 2022b). No previous studies have been conducted comparing Landlocked Salmon or Brown Trout PFAS concentrations to any other species sampled here.

The current New York State Department of Health PFOS fish consumption guideline is 50 ng/g ww for one or fewer meals per month and 200 ng/g ww for “don’t eat” advisory, higher than any concentration observed here (NYSDOH, 2023). Other states, such as Maine, have set their guidelines lower, to 15 ng/g ww for one or fewer meals per month, and 60 ng/g ww as a “do not eat” concentration threshold (Maine CDC, 2023). Maine CDC performed risk assessments based on ASTDR reference doses and found consumption of fish above the 15 ng/g ww threshold resulted in a higher risk of developmental and immunological effects if consumed in greater quantities than recommended (Maine CDC, 2023). All means were below these thresholds, while two Lake Trout samples were higher than the 15 ng/g ww threshold, but none over the 60 ng/g ww threshold. The US EPA has recently published recommended PFOS and PFOA ambient water quality criteria for whole-fish (201 and 6490 ng/g ww, respectively) and muscle tissue (87 and 133 ng/g ww, respectively) (EPA, 2024a; EPA, 2024b), higher than any sample tested here.

Similar to PFAS results, Lake Trout had significantly higher total Hg concentrations compared to Landlocked Salmon and Brown Trout, but not Rainbow Trout (Supplemental Fig. 4). Lake Trout was the only species with a median total Hg concentration higher than the US EPA Hg water quality criterion of 300 ng/g ww (349.10 ng/g ww). No samples

were higher than the NYSDOH fish consumption guideline of 2000 ng/g ww for “don’t eat” fish. Given that both Hg and PFAS bind to proteins, and correlation has been found in other waterbodies (Arinaitwe et al., 2020), we investigated their co-occurrence in Lake Trout. Hg and PFAS were not found to co-occur—total Hg and summed PFAS had a correlation coefficient of -0.51 (Supplemental Fig. 5).

3.2. Summary of diet profile and fatty acid compositions of Lake Trout

Lake Trout rely on Alewife as a dominant diet source in Seneca Lake (Hammers, 2022), and at the time of sampling Round Goby (*Neogobius melanostomus*) had not yet invaded Seneca Lake altering fish diets. Diets of Lake Trout, Landlocked Salmon, Brown Trout, and Rainbow Trout compared during a set of fishing tournaments on Lake Ontario found Lake Trout consumed predominantly Alewife and smelt species, whereas the other three species consumed predominantly Alewife—but also smelt, sculpin, and lower trophic level insects (Brandt, 1986). Alewife are a higher protein fish compared to Rainbow Smelt (*Osmerus mordax*) and Round Goby (Dietrich et al., 2006; Hicks et al., 2019) that are commonly consumed by Lake Trout. Stomach content data in Seneca Lake sportfish are lacking for further comparison and would be valuable in future studies.

Lake Trout from Seneca Lake had average fatty acid compositions of 3.8 % for linoleic acid (18:2n-6) and 3.6 % for α -linolenic acid (18:3n-3) (Supplemental Fig. 2a). These essential fatty acids are precursors of highly unsaturated fatty acids which are involved in the anti-inflammation and immune responses, and fish consumption has been suggested to counteract the negative health effects from PFOS and PFOA exposure (Hamade, 2024). A ratio of oleic acid (18:1n-9) to palmitoleic acid (16:1n-7) over 1 (higher oleic acid percentage) is representative of a more pelagic diet, and higher alewife consumption for Lake Trout (Futia et al., 2019). Lake Trout from Seneca Lake had a ratio of 25.7/7, or 3.67 times higher concentrations of oleic acid, further suggesting a high reliance on Alewife as a diet source. Lake Trout from Seneca Lake had higher average dietary essential fatty acids, namely combined eicosapentaenoic acid (20:5n-3) and docosahexaenoic acid (22:6n-3) per 100 g of lipid (13.8 g) (Supplemental Table 2) compared to Lake Trout from Lake Ontario (10.7 g) and Lake Erie (10.9 g) (Pantazopoulos et al., 2013). The samples analyzed here, when consumed in moderation, were lower than New York State consumption thresholds (NYSDOH, 2023), and are a good source of dietary essential fatty acids (Institute of Medicine, 2005).

3.3. Effects of carbon source, trophic level, age, and size on PFAS accumulation

Carbon source (as proxied by $\delta^{13}\text{C}$) was significantly higher in Rainbow Trout than all other species in Seneca Lake but did not significantly differ among other sampled species (Supplemental Fig. 6c). All species' mean $\delta^{13}\text{C}$ were within 3 ‰, suggesting a similar carbon source (Supplemental Fig. 6a). $\delta^{13}\text{C}$ was found to be a significant predictor for summed PFAS concentrations in Lake Trout ($R^2 = 0.23$)

(Fig. 3a), but a lot of leverage is from a single high concentration point. With this single high concentration point removed, there was no significance observed ($R^2 = 0.07$; Supplemental Fig. 7). More data is needed to assess whether a more benthic carbon source (higher $\delta^{13}\text{C}$) is a driver of higher PFAS concentration in Lake Trout from Seneca Lake. Others have suggested more benthic diet sources have higher PFAS concentrations than pelagic prey (Lescord et al., 2015; Martin et al., 2004), because of sediment contamination and uptake into benthic species such as *Diporeia* (Martin et al., 2004) that are also present in

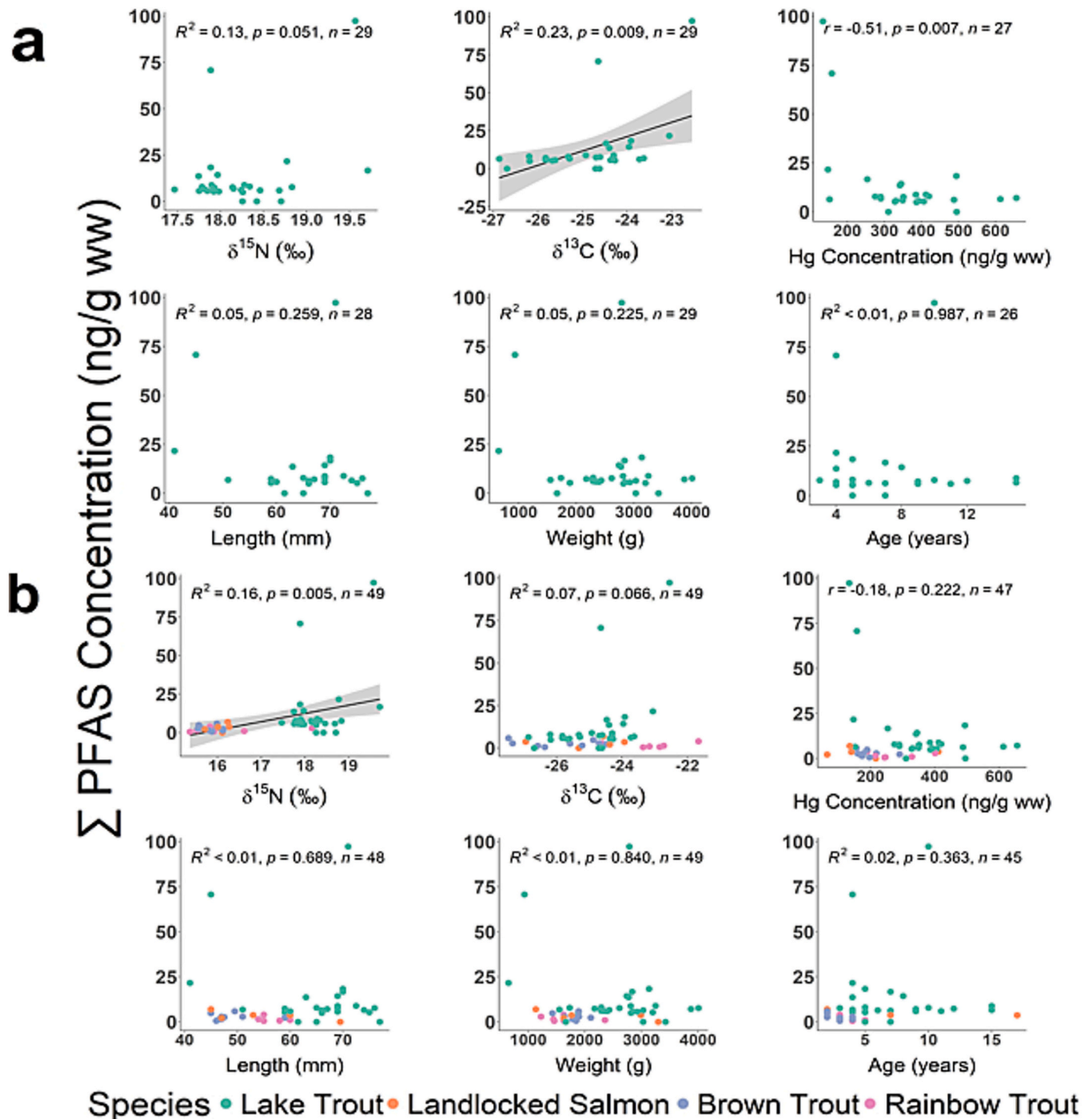


Fig. 3. a. Linear regressions of summed PFAS concentrations predicted by $\delta^{15}\text{N}$, $\delta^{13}\text{C}$, length, weight, and age in Lake Trout and b. in all National Lake Trout Derby on Seneca Lake fish. Coefficients of determination (R^2), p -values, and sample size (n) are listed for each regression, while correlation coefficients (r) are used in their place when appropriate (i.e., Hg).

Seneca Lake (Watkins et al., 2012). *Diporeia* are consumed by pelagic Alewife, a major diet item for Lake Trout in Seneca Lake (Hammers, 2022). A food web base of a benthic species such as *Diporeia* may result in biomagnification of PFAS compounds when consumed by Alewife, and then Lake Trout.

Lake Trout had a significantly higher average trophic level ($\delta^{15}\text{N} = 18.23\text{‰}$) than other species tested in Seneca Lake (15.84–16.24 ‰), likely from consuming higher trophic level prey (Supplemental Fig. 6b). Lake Michigan Lake Trout also exhibited a higher trophic level and summed PFAS concentration compared to other pelagic sportfish tested (Chinook Salmon (*O. tshawytscha*), Coho Salmon (*O. kisutch*), and Rainbow Trout) (Miranda et al., 2023). Lake Michigan Lake Trout and Rainbow Trout had a higher $\delta^{15}\text{N}$ and slightly lower $\delta^{13}\text{C}$ compared to other sportfish species sampled, as well as a higher concentration of summed PFAS (Miranda et al., 2023). Lake Trout had a higher $\delta^{15}\text{N}$ and summed PFAS concentration in Lake Ontario and Lake Huron as well (Ren et al., 2022a; Ren et al., 2022b), and in waterbodies in proximity to AFFF and background sites (Kaboré et al., 2022). Certain PFAS have been found to bioaccumulate and bioconcentrate, such as PFOS and PFDA (Burkhard, 2021), suggesting increased concentrations in Lake Trout compared to other species may be because of consuming higher trophic level prey items. We conducted linear regression analysis to determine if $\delta^{15}\text{N}$ was a predictor of PFAS concentrations in Seneca Lake. No significant results were observed for Lake Trout alone ($R^2 = 0.13$, $p = 0.051$) (Fig. 3a), but a significant relationship was observed among species combined, albeit with low explanation of the variability ($R^2 = 0.16$) (Fig. 3b).

Only Seneca Lake sportfish, not forage fish, were assessed in this study, resulting in a small range of $\delta^{15}\text{N}$ values and similar trophic levels among the four sportfish sampled. In other waterbodies, there were much larger ranges of $\delta^{15}\text{N}$ from the inclusion of lower trophic level species, such as Lake Huron (~2–14 ‰; Ren et al., 2022a), Lake Ontario (~7–14 ‰; Ren et al., 2022b), and Lake Michigan (~7–15 ‰; Miranda et al., 2023). Ren et al. (2022a) reported Lake Trout from Lake Huron having the highest $\delta^{15}\text{N}$ and summed PFAS concentration, but this was not observed in Lake Ontario where Deepwater Sculpin (*Myoxocephalus thompsonii*) had the highest summed PFAS concentrations and similar $\delta^{15}\text{N}$ values to Lake Trout (Ren et al., 2022b). In Lake Michigan, Slimy Sculpin (*Cottus cognatus*) dominated in both summed PFAS concentration and $\delta^{15}\text{N}$ value, both higher than Lake Trout (Miranda et al., 2023).

Length was not a significant predictor for summed PFAS concentration in Lake Trout, nor all species combined (Fig. 3), and did not correlate well with $\delta^{13}\text{C}$ ($r = -0.19$) nor $\delta^{15}\text{N}$ ($r = -0.01$) (Supplemental Fig. 5). Others have also observed no correlation nor prediction between summed PFAS and length (Pan et al., 2018), but Babut et al. (2017) did find individual PFAS correlated positively with length, which was observed in Landlocked Salmon in the current study (i.e., perfluoroheptanoic acid (PFHpA) and PFNA), but not other species. Others have observed similar results in lab studies (Banyoi et al., 2022), with the caveat that many of the studies had concentrations much higher than environmentally relevant concentrations. Increased length is known to predict increased Hg concentrations (Wiener et al., 2002), and this was observed in all species combined ($R^2 = 0.37$) (Supplemental Fig. 8). Length was a predictor for age in Landlocked Salmon ($R^2 = 0.94$) and Rainbow Trout ($R^2 = 0.67$), but not in Lake Trout nor Brown Trout—demonstrating data follow well described patterns (Supplemental Fig. 9). Weight was not found to predict summed PFAS in Lake Trout, nor all species (Fig. 3), but was a significant predictor for Hg concentrations in all species ($R^2 = 0.33$) (Supplemental Fig. 8). Age was also not found to predict summed PFAS concentrations in Lake Trout, nor all species (Fig. 3), but was a predictor for total Hg ($R^2 = 0.14$, Supplemental Fig. 8), as expected. Age has been understudied as a predictor, but the little evidence available shows that age influences PFAS accumulation (Newsted et al., 2017; Ye et al., 2008), contrary to our observations. Our study focused on derby caught species that are typically only weighed in when larger and older, thus skewing our age

range to an older demographic (Lake Trout ages ranged from 3 to 15 years old). Weight has not been observed to affect PFAS concentrations in Largemouth Bass (Babut et al., 2017, Giari et al., 2023, Newsted et al., 2017, Semerád et al., 2021) and has not been tested in any species discussed here. Length, weight, and age were significant predictors for Hg but not summed PFAS, suggesting these characteristics may be important for the differential bioaccumulation in PFAS and Hg observed in their negative correlation ($r = -0.51$), but need to be explored further to determine the mechanism causing bioaccumulation.

Other predictors for PFAS accumulation in fish have been found, including proximity to wastewater treatment plants and atmospheric deposition (Podder et al., 2021), fluorinated precursors in sediment (Langberg et al., 2020), and serum proteome variability (Point et al., 2023). Due to many PFAS compounds being proteinophilic (Han et al., 2003), fish with increased reliance on prey items with higher protein composition are expected to have higher PFAS concentrations. Lake Trout from Seneca Lake generally eat a high protein diet of Alewife (Hammers, 2022) that may be driving PFAS concentrations, but further trophic level dynamics need to be elucidated to determine biomagnification in the lake. Alewife have been suggested as a driver for PFAS concentrations in Lake Ontario Lake Trout because of increased summed PFAS concentrations over Round Goby and Rainbow Smelt, and biomagnification factors were over one for Lake Trout consuming Alewife in PFHxS, PFOS, PFDoA (Ren et al., 2022b).

3.4. Comparing Lake Trout from Seneca Lake to nearby waterbodies

Lake Trout from Seneca Lake had a lower percentage of PFOS than Lake Trout from Canadice Lake, Cayuga Lake, Lake Erie, and Lake Ontario (Fig. 4a). Lake Trout from Seneca Lake had correspondingly higher percentages of other compounds than Lake Trout from the other sampled waterbodies—notably long chain PFCAs (i.e., PFNA and PFDA) (Fig. 4a).

PFAS compositions of Seneca Lake Landlocked Salmon, Brown Trout, and Rainbow Trout were still more comparable to Lake Trout from Seneca Lake than Lake Trout from other waterbodies, suggesting other species in Seneca Lake are affected, but less so than Lake Trout. Differing compositions could be due to a unique point source on Seneca Lake that is affecting Lake Trout from Seneca Lake more than in other waterbodies and other species in Seneca Lake.

Summed PFAS concentrations in Lake Trout from Seneca Lake are significantly higher than the other two Finger Lakes analyzed, Canadice and Cayuga Lakes, but are significantly lower than the Great Lakes we analyzed; Lake Ontario and Lake Erie (Fig. 4b, Supplemental Fig. 10). Lake Trout from Cayuga and Canadice Lakes have comparatively lower PFOS concentrations than Seneca Lake, and PFOS represents a much larger percentage of summed PFAS (Fig. 4a, Supplemental Table 3). For Canadice Lake, only PFOS was detected, in low concentrations, suggesting lower PFAS inputs to the lake, and may be limited to atmospheric deposition impacting all of the sampled lakes (Pfothenauer et al., 2022). PFOS represented 89 % of the summed PFAS concentration in Lake Michigan Lake Trout (Miranda et al., 2023), higher than observed in Lake Trout from Seneca Lake (44 %), and similar to Lake Erie (92 %) and Lake Ontario (99 %).

Some PFAS concentrations reported for Lake Trout in literature are higher than those recorded here, due to the use of whole Lake Trout (Remucal, 2019; Ren et al., 2022a) versus our use of fillet samples. To account for these differences, a general conversion between fillet and whole-body concentrations was used to compare to whole-body concentrations (Levanduski et al., 2024). Miranda et al. (2023) used muscle plugs, similar to our sampling of a small fillet portion, and concentrations are similar—average summed PFAS concentrations were 16.51 ng/g ww in Lake Michigan Lake Trout, compared to 13.12 ng/g ww in Lake Trout from Seneca Lake. Guo et al. (2012) also used fillet samples for Lakes Ontario, Erie, Huron, and Superior, and concentrations were generally higher than others observed here (Supplemental Table 4). We

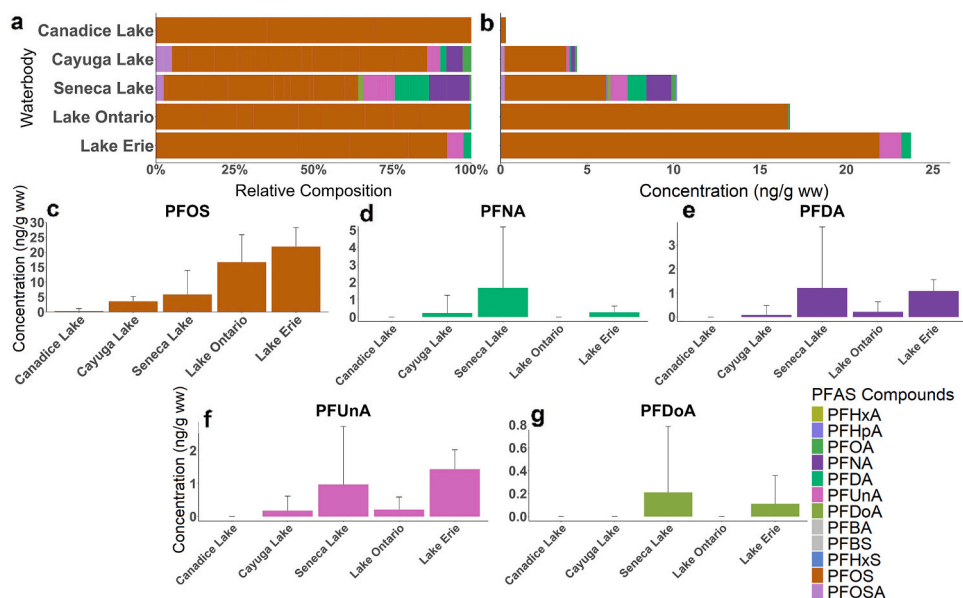


Fig. 4. a. Relative PFAS composition in Lake Trout by waterbody. b. Mean summed PFAS for Lake Trout in each waterbody. c-g. Individual mean PFAS concentrations for 5 commonly detected compounds in Lake Trout among waterbodies. The coloration for each compound is in the legend in the bottom right of the graphic. Compounds not detected in any sample are denoted by a gray box in the legend.

estimated whole-body PFOS concentrations of 53.59 ng/g ww and 43.72 ng/g ww for Lake Erie and Lake Ontario, respectively (Supplemental Table 4). These concentrations fall within the observed range, and ranges quantified by others in Lake Ontario and Lake Erie Lake Trout. Estimated whole-body PFOS concentrations in Lake Trout from Seneca Lake are 23.58 ng/g ww, lower than observed in Great Lakes waterbodies (Supplemental Table 4).

Lake Ontario and Lake Erie have the most potential PFAS sources, due to their large surface areas and high number of industrial sites and urbanization in their watersheds (Remucal, 2019). Despite the higher PFOS and summed PFAS concentrations in these Great Lakes (Fig. 4b), Lake Trout from Seneca Lake have higher concentrations of other long chain PFCA compounds such as PFNA (Fig. 4d), PFDA (Fig. 4e), PFUnA (Fig. 4f), and PFDoA (Fig. 4g). Higher concentrations of long chain PFCA compounds was also observed compared to the other two Finger Lakes sampled (Supplemental Table 3). Long chain PFCA concentrations were higher in Lake Trout from Seneca Lake than from Lake Michigan (Miranda et al., 2023) (e.g., mean PFNA concentrations were 1.68 ng/g ww in Lake Trout from Seneca Lake compared to 0.26 ng/g ww in Lake Michigan Lake Trout). Lake Michigan Lake Trout did not have significantly higher concentrations of PFCA compounds compared to Rainbow Trout, as seen in Seneca Lake, with PFOS representing a larger percentage of summed PFAS in Lake Trout compared to Rainbow Trout in Lake Michigan (Miranda et al., 2023). Lake Michigan Lake Trout exhibited a similar $\delta^{13}\text{C}$ and a higher $\delta^{15}\text{N}$ than Rainbow Trout, the same relationship observed in Seneca Lake (Miranda et al., 2023).

Lake Trout from Lake Erie and Lake Ontario have high concentrations of PFOS relative to the other Great Lakes, and similar to Lake Superior (Supplemental Table 4). Remucal (2019) has reported a general increase in summed PFAS concentration from the western most lakes (Lake Superior and Lake Michigan) to the eastern most lakes (Lake Erie and Lake Ontario). Higher concentrations in the eastern lakes are attributed to increased industrial activity and urbanization on Lake Erie and Lake Ontario. Lake Trout from Seneca Lake had a higher PFOS concentration than Canadice and Cayuga Lakes. Watershed area: surface area is comparable among Cayuga Lake (11.82), Canadice Lake (12.56), and Seneca Lake (9.16), but Seneca and Cayuga have a larger surface area than Canadice. Seneca Lake has many industrial sites within the watershed, including a historic AFFF site. Neighboring watersheds to

Seneca Lake include the largest landfill in New York State. These land uses are both associated with high PFAS contamination (Glüge et al., 2020). Lake Trout from Seneca Lake also have a more diverse PFAS composition suggesting a unique point source contributing to higher concentrations of other PFAS compounds (Fig. 4a).

Lake Trout, Brown Trout, and Landlocked Salmon are all stocked species in Seneca Lake and Cayuga Lake (NYSDEC, 2025a, 2025b). Lake Trout, Brown Trout, and Rainbow Trout are stocked in Canadice Lake (NYSDEC, 2025c), while Lake Trout and Brown Trout are stocked in Lake Ontario (NYSDEC, 2023b), and Rainbow Trout and Brown Trout are stocked in Lake Erie (NYSDEC, 2025d). These stocking efforts are most likely not increasing PFAS concentrations among these samples, as the size at stocking is much smaller than the size at collection for derby caught species and PFAS compounds are expected to be diluted. Capozzi et al. (2023) observed elevated concentrations within hatchery raised fish, but those fish were caught near surrounding point sources.

3.5. A potential point source

The former Seneca Army Depot, a Superfund site that has a history of AFFF activity for fire control training, has been closed since 2000 (US EPA, 2024c; Fig. 1). Compounds used in AFFF mixtures include 3–11 carbon length PFCA and 2–10 carbon length PFSA (Glüge et al., 2020), many of which are detectable in Seneca Lake sportfish, and in highest concentrations in Lake Trout. The only AFFF mixture available at the time for military use (Ansul) was composed solely of PFCA and PFSA compounds (Backe et al., 2013). No fluorotelomerization was conducted at that time to produce AFFF, as some newer AFFF and consumer products use, and fluorotelomers are commonly detected from landfill runoff (Fiedler et al., 2010; Lang et al., 2017). The lack of these compounds suggests that landfill runoff or other modern industrial inputs are not driving PFAS accumulation in Seneca Lake sportfish. The only fluorotelomer compounds detected in Seneca Lake were N-MeFOSAA ($n = 3$) and N-EtFOSAA ($n = 1$), both PFOS precursor compounds as transient degradation intermediates through electrochemical fluorination (Buck et al., 2011), at relatively low concentrations, suggesting that a source for PFAS exposure to sportfish in Seneca Lake is historic AFFF usage.

Groundwater samples from Seneca Army Depot have been tested by

Environmental Working Group in 2018, which found concentrations above 1 ng/g ww for 6 compounds—PFOA (89 ng/g ww), perfluorohexane sulfonic acid (PFHxS; 36 ng/g ww), perfluorohexanoic acid (PFHxA; 14 ng/g ww), PFOS (8.3 ng/g ww), PFHpA (3.2 ng/g ww), and PFBS (1.6 ng/g ww) (EWG, 2024). PFOA, PFHxS, PFHxA, and PFHpA were all detected in higher concentrations in Lake Trout from Seneca Lake than in Lake Trout from Canadice Lake, Cayuga Lake, Lake Erie, and Lake Ontario. This composition of PFAS is similar to the AFFF formulation during the active time of use at Seneca Army Depot (Backe et al., 2013). Although these are commonly detected compounds in water, the lack of detectable concentrations of compounds associated with other point sources—such as landfill leachate—and higher concentrations of detected PFAS compounds than expected from atmospheric deposition, we suspect the Seneca Army Depot to be the unique point source driving PFAS concentrations in Seneca Lake.

4. Conclusions

PFAS concentrations in Lake Trout from Seneca Lake were significantly higher than other sportfish species tested in Seneca Lake, while also being higher than Lake Trout from other Finger Lakes. This study contributes to our understanding of PFAS bioaccumulation by observing that higher PFAS concentrations in Lake Trout were not explained by trophic level (within species), length, weight, nor age. However, Lake Trout appear susceptible to elevated PFAS exposure because of their higher trophic level relative to other sportfish species—likely because of their comparatively higher consumption of Alewife, rather than consumption of invertebrate prey directly. Carbon source was a predictor for summed PFAS concentrations with the relationship significant among all Lake Trout samples, but one high leverage value drove the trends. Hg contamination inversely correlated with summed PFAS concentrations, implying that drivers of Hg uptake, including length and age, are not drivers of PFAS accumulation in Lake Trout from Seneca Lake.

When comparing Lake Trout from Seneca Lake to eastern Lake Erie and western Lake Ontario, PFOS represented a smaller percentage of the summed PFAS composition in Seneca Lake—potentially from fewer industrial inputs. Seneca Lake has a history of AFFF use and is within the airshed of the largest landfill in New York State. Compounds from historic AFFF usage, long chain PFCAs and PFSAs, are the only PFAS observed in equivalent or higher concentrations than the Great Lakes sampled here. The decreased PFOS and increased long chain PFCAs and PFCAs composition suggests the main PFAS source to Seneca Lake is historic AFFF usage from the Seneca Army Depot.

Predictors for Lake Trout's significantly higher concentration could not be fully elucidated, but we propose higher reliance on upper trophic level prey items compared to other sportfish sampled here, as well as impact by the Seneca Army Depot, may be driving concentrations. A complete food web and PFAS analyses of Seneca Lake biota would allow for biomagnification and trophic level dynamics to be derived. Lake Trout from Seneca Lake had detections of PFAS compounds not observed in other sportfish species in Seneca Lake, and reflected a unique source compared to Lake Trout from the Great Lakes and other Finger Lakes. By being a high trophic level species that can accumulate compounds not observed in other sportfish here, Lake Trout are shown to be a good sentinel species for contaminant studies in the future. Point source delineation by testing fish from tributaries connecting Seneca Lake to potential point sources, such as the Seneca Army Depot, would confirm if a point source is polluting Seneca Lake's prized Lake Trout fishery.

CRedit authorship contribution statement

Eric Levanduski: Writing – review & editing, Writing – original draft, Visualization, Software, Formal analysis, Data curation. **Susan F. Cushman:** Writing – review & editing, Project administration, Investigation, Funding acquisition, Data curation, Conceptualization. **Lisa B.**

Cleckner: Writing – review & editing, Methodology, Investigation, Funding acquisition, Conceptualization. **Wayne Richter:** Writing – review & editing, Visualization, Software. **Jesse C. Becker:** Writing – review & editing, Validation, Methodology. **Trevor Massey:** Writing – review & editing, Methodology, Investigation, Formal analysis. **Jacques Rinchar:** Writing – review & editing, Methodology, Investigation, Data curation. **N. Roxanna Razavi:** Writing – review & editing, Writing – original draft, Supervision, Resources, Project administration, Methodology, Funding acquisition, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

Funding for this work came from the NYS Center of Excellence in Healthy Water Solutions, Seneca Lake Pure Waters Association, and Hobart and William Smith Colleges. Thank you to all Seneca Lake Pure Waters Association volunteers for help processing fish during the National Lake Trout Derby on Seneca Lake, all undergraduates for help processing fish and prepping for analyses including ESF undergraduates (Jeremiah Tennant, Annabelle Vogl, Sarita Perez, Sydney Burgy) and Hobart and William Smith Colleges undergraduates (Taline Almasian, Emma Brachfeld, Emma McLean). Thank you to Mike Satchwell for help with SPE and LC-MS/MS. Thank you to Ben Carson and NYSDEC for aging otoliths. Thank you to students from Dr. Rinchar's lab at SUNY Brockport for lipid and fatty acid analyses.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2025.179038>.

Data availability

Data will be made available on request.

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