Per– and Polyfluoroalkyl Substances (PFAS): An Emerging Environmental and Human Health Concern for the Great Lakes?

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The annual value of Great Lakes recreational, tribal, and commercial fisheries exceeds $7 billion and is estimated to support more than 75,000 jobs. These fisheries are an enormously important resource for the Great Lakes, but one that is confronted by multiple human-induced stressors including chemical contaminants that can result in fish consumption advisories and potential restrictions on harvest. Current fish consumption guidelines are generally informed by legacy contaminants [i.e., heavy metals and polychlorinated biphenyls (PCBs)] that are well monitored in fish. However, mounting evidence suggests that those guidelines should also consider emerging contaminants, although quantification of these contaminants is limited. Prominent among these emerging and harmful contaminants are **per- and polyfluoroalkyl substances (PFAS)**, a family of man-made fluorinated organic chemical compounds highly resistant to degradation, making them extremely persistent in the environment and able to bioaccumulate in wildlife such as freshwater fish (Figure 1).

Figure 1. PFAS concentrations in locally caught freshwater fish (from Barbo et al. 2023, Env. Res. 220, 115165)

PFAS have been detected in both marine and freshwater fishes (Miranda et al. 2021), including some species of Great Lakes fish, but data are limited both spatially and temporally for Lake Michigan. Furthermore, the extent and level of PFAS pollution in Great Lakes food webs remains unknown despite the potential for negative impacts on ecological and human systems via fish tissue consumption, the most common avenue for human exposure. Our research addressed this data gap by quantifying the concentration of PFAS in Great Lakes sportfish, and their associated prey fish, to assess potential contaminant pathways resulting in human exposure.

RESEARCH OBJECTIVES

Aim 1: We evaluated dietary routes for PFAS exposure from prey to predator fish in Lake Michigan by quantifying the concentrations of different PFAS compounds in fish along with measuring stable C and N isotopes to infer trophic position.

Aim 2: We assessed the relationship between total fluorine and PFAS concentrations, using a novel technology called Particle-Induced Gamma-ray Emission (PIGE) spectroscopy, to determine the full extent of the PFAS problem in Lake Michigan food webs.

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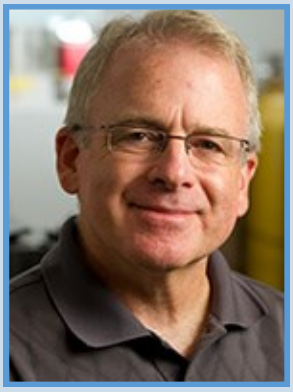
RESEARCHER PROFILES



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Co-Principal Investigator: **Dr. Daniele de A. Miranda** is an assistant research professor in the Department of Biological Sciences at the University of Notre Dame.

MAJOR CONCLUSIONS & SIGNIFICANCE

- In Lake Michigan, we found that PFAS were present in all 204 fish sampled from the lake, but at differing concentrations and composition. In general, total PFAS were lower in prey fish than in predator fish (Figure 2), suggesting that bioaccumulation is occurring via dietary routes (Miranda et al. 2023). Overall, the legacy contaminant PFOS was the most abundant PFAS in fish but many other compounds were also detected (Figure 2). Even though PFAS were detected in all Lake Michigan fish, the concentrations are generally lower than those observed in similar fish from other Great Lakes such as Ontario and Erie.

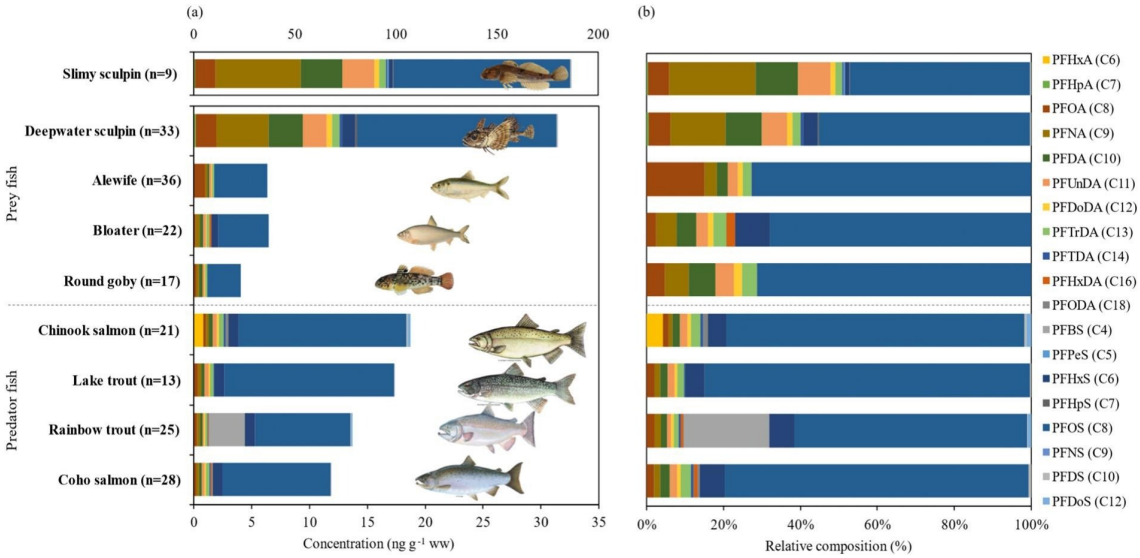

Figure 2. Summed concentrations of 19 PFAS compounds (ng g^{-1} wet weight (ww)) for muscle tissue measured in predator and prey fish from Lake Michigan, USA, expressed as (a) absolute and (b) relative PFAS composition normalized to 100% (n = 204).

- In tributaries to Lake Michigan, we discovered, for the first time for this contaminant, that PFAS were being "off-loaded" from spawning female salmon to their embryos during the process of egg development (Conard et al. 2022). Overall, PFOS was ~100X higher in eggs than in female muscle tissue. This phenomenon can represent a risk not only for embryos but for animals that rely on fish eggs as a food source and suggests that salmon migrations may be transferring PFAS across ecosystems.
- In a global analysis, we evaluated the biomagnification of PFAS in aquatic food webs of the ocean and freshwaters using published literature (Miranda et al. 2022). We found that food chain length, geographic location, sampling methodologies, tissues analyzed, and distance from known direct PFAS inputs have a major impact on the biomagnification of these compounds.

WHAT DOES THIS MEAN FOR INDIANA?

We observed widespread occurrence of PFAS in Lake Michigan forage fish and salmonids, suggesting that these compounds are being transferred through the Lake Michigan food web from prey to predator fish. The well-known toxic compound PFOS was found in all fish species, which raises environmental and human health concerns. However, PFAS concentrations in Lake Michigan fish were lower than in other lakes, lower in muscle than in other tissues such as organs, and did not exceed current suggested guidelines for fish consumption. As in all fish consumption, the risks of contaminant ingestion must be balanced with the health benefits of eating fish protein. In a parallel study, we observed maternal offloading of PFAS by spawning salmonids to their eggs in Lake Michigan tributaries, which can represent a risk for wildlife when salmon migrations transfer PFAS from lakes to rivers. This finding has high physiological and ecological implications from the molecular scale for developing embryos to the ecosystem scale because salmonid migrations deliver high amounts of labile resources to streams annually including eggs that are consumed by other organisms such as crayfish and resident fishes. Hence, salmon eggs and carcasses provide important nutrient subsides to streams, but potentially also contaminants when they become a food source for a wide range of aquatic consumers.

PUBLICATIONS FROM THIS RESEARCH

Conard, W.M., Whitehead, H.D., Harris, K.J., Lamberti, G.A., Peaslee, G.F., Rand, A.A., (2022). Maternal Offloading of Per- and Polyfluoroalkyl Substances to Eggs by Lake Michigan Salmonids. Environ. Sci. Technol. Lett. 9, 937–942. https://doi.org/10.1021/acs.estlett.2c00627.

Miranda, D. A., Peaslee, G. F., Zachritz, A. M., Lamberti, G. A. (2022). A worldwide evaluation of trophic magnification of per- and polyfluoroalkyl substances in aquatic ecosystems. Integr. Environ. Assess. Manag, 00, 1–13. doi.org/10.1002/ieam.4579.

Miranda, D.A., Zachritz, A.M., Whitehead, H.D., Cressman, S.R., Peaslee, G.F., Lamberti, G.A., (2023). Occurrence and biomagnification of perfluoroalkyl substances (PFAS) in Lake Michigan fishes. Sci. Total Environ. 895, 164903, 1-9. doi.org/https://doi.org/10.1016/j.scitotenv.2023.164903.

TRAINING THE NEXT GENERATION

One of the missions of the Indiana Water Resources Research Center, and all Water Centers, is to train the next generation of water scientists. This project successfully funded research for five Ph.D. students and 11 undergraduate students within Drs. Lamberti and Peaslee's labs.

For more information about the Indiana Water Resources Research Center, visit iwwrc.org.

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