

Depositional history of PFAS in the bay of Green Bay in Lake Michigan over the last 80 years

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The bay of Green Bay in Lake Michigan has been designated as an Area of Concern since the 1980s due to increased anthropogenic activities and accumulation of legacy pollutants. Despite decades of regulation and remediation efforts, it remains a hypereutrophic freshwater estuary with seasonal hypoxia. A sediment core was collected from central Green Bay in 2021 and analyzed for PFAS and total organic carbon (TOC) to explore the pollution history of PFAS and trends in different PFAS species over the past 80 years in the bay of Green Bay. Our results indicate that the predominant PFAS compounds in surface sediments with concentrations higher than 0.18-ng/g were N-EtFOSAA, N-MeFOSA, N-MeFOSE, PFOS, N-EtFOSE, PFUnA, PFOA, PFDoA, PFDA, FOSA, PFHxA, PFNA, and 6:2 FTSA. Among all detectable PFAS in sediment samples, EtFOSAA exhibited the highest concentrations. Vertically, the contents of N-MeFOSAA, PFOS, PFOA, 6:2 FTSA, and 8:2 PTSA generally increased from the 1940s to 2021. A similar increasing trend was observed for the PFOS/PFOA concentration ratio. In contrast, PFBS and PFHxA peaked in the 1960s, along with the N-EtFOSAA/N-MeFOSAA concentration ratio. After reaching its peak in 1960s, PFBS concentrations remained lower than 0.05 ng/g. In addition, N-EtFOSAA, N-EtFOSE, and FOSA demonstrated peak concentrations in the 1980s (~20 cm depth). PFOS also had a peak in 1980s but increased again after 2000. Overall, PFOS concentrations were significantly correlated with TOC content whereas weak or no significant correlations were observed for other PFAS species. The depositional history of PFAS in the bay of Green Bay appears to be species-specific, likely related to pollution sources, production and application history, and/or post depositional transformation and degradation within sediments. Further studies are needed to better reconstruct the pollution history of PFAS and their removal and transport processes in the bay of Green Bay.