Emerging Contaminants: Fate, Transport, and Management Across Diverse Watersheds

Abstract

The growing concern over environmental pollution has driven extensive research into a range of emerging contaminants (ECs) that threaten ecosystem integrity and human health. These include pharmaceuticals, personal care products, endocrine disruptors, nanomaterials, microplastics, and per- and polyfluoroalkyl substances (PFASs). This dissertation addresses these issues through a combination of bibliometric analysis, critical reviews, mechanistic studies, and watershed-scale modeling.

A bibliometric analysis of 62,670 publications (2000–2024) revealed exponential growth in EC research, with annual outputs rising from fewer than 1,000 before 2006 to nearly 7,000 by 2022. China and the United States emerged as leading contributors, while *Environmental Science & Technology* was identified as the most influential journal. Core themes included contaminant degradation, removal, and ecological impacts, underscoring the urgent need for remediation strategies and regulatory frameworks.

Building on this, two comprehensive reviews focused on PFAS, which exemplify the persistence, bioaccumulation, and global dispersal of ECs. A synthesis of global regulatory frameworks revealed highly fragmented standards, with persistent gaps in monitoring and enforcement. Critical pathways include industrial discharges, consumer products, and atmospheric transport, with geographic disparities in exposure patterns. A mechanistic review of PFAS fate and transport demonstrated how their amphiphilic chemistry governs mobility across soils, surface waters, sediments, vegetation, and the atmosphere. Longer-chain PFASs exhibited greater sorption, while shorter-chain variants showed higher mobility and deposition potential. Interactions with co-contaminants and microplastics further complicate transport. Emerging models, such as the Leverett Thermodynamic Model and Surface Roughness Multipliers, show promise in predicting PFAS retention and transport dynamics but require refinement to capture complex physicochemical and biological interactions.

Finally, nutrient enrichment dynamics were evaluated through a case study in the St. Johns River Basin (Florida) using Weighted Regressions on Time, Discharge, and Season (WRTDS) and WRTDS-tidal models. Analysis of 25 years of data (1999–2024) showed strong spatial contrasts.

At Rockledge (headwaters), total Kjeldahl nitrogen (TKN) increased by 40% and total nitrogen (TN) by 23%, while total phosphorus (TP) declined by 73%. At Sanford (mid-basin), TN rose by 8.9%, but TP rebounded by +14% between 2019–2024 despite earlier declines. Cow Creek (agricultural sub-basin) exhibited episodic surges, with TKN rising 86% from 2014–2019, though long-term TN and TP decreased by 26–70%. At Jacksonville (estuarine outlet), nutrient loads consistently decreased (TKN –45%, TN –25%, TP –30%), reflecting both upstream interventions and tidal dilution. WRTDS tidal analysis confirmed a strong negative relationship between salinity and TKN, highlighting freshwater-driven nutrient loading upstream and estuarine mixing downstream. Land cover changes (1999–2024) showed urban expansion driving headwater and mid-basin nutrient pressures, while agricultural practices dominated Cow Creek export. Correlation analysis revealed that variations in TKN, TN, and TP concentrations were associated with land cover changes, where increased urban and agricultural expansion corresponded to higher nutrient loads, particularly in the upstream HUC12 watersheds, while forested and wetland areas exhibited nutrient attenuation effects.

Taken together, this dissertation provides a multi-scale perspective on ECs from global bibliometric patterns to molecular-level PFAS transport, and from watershed nutrient fluxes to estuarine dynamics. The findings highlight the interconnected challenges of ECs, PFAS, and nutrient enrichment, while identifying critical research gaps in regulation, modeling, and management. Collectively, this work highlights the necessity of integrative, interdisciplinary approaches to safeguard water resources, inform regulatory frameworks, and sustain ecosystem and human health in an era of accelerating environmental change.

Keywords: Emerging Contaminants (ECs); Per- and Polyfluoroalkyl Substances (PFAS); Fate and Transport; Regulatory Frameworks; Bibliometric Analysis; Estuarine Dynamics; Land Use Change; Salinity Gradient; St. Johns River Basin.