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Transformation of the urban triazine type biocide terbutryn: insights from Compound-Specific Isotope Analysis

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Biocides are added in building materials like renders or paints on façades as protection against algae and fungi growth. With wind driven rainfall, biocides can leach from buildings and eventually contaminate urban groundwater. Studies on transfer, degradation kinetics, degradation mechanisms and persistence in the environment of urban biocides are rare. Traditional methods based on concentrations often reflect both dilution, due to non-destructive processes, and degradation involving bond-breaking of biocide molecules. Established since two decades for legacy point-source industrial contaminants, Compound-Specific Isotope Analysis (CSIA) is an emerging approach to evaluate magnitudes and mechanisms of non-point source micropollutant degradation in the environment, although it has not been applied yet to urban biocides. To use CSIA in field-based approaches, reference laboratory degradation experiments have to be conducted.

Here we carried out reference abiotic hydrolysis, photodegradation and biodegradation experiments for the urban biocide terbutryn to compare kinetics and evaluate the spectrum of stable isotope fractionation to interpret transformation pathways. Experimental setups for hydrolysis include pH = 1, pH = 13 and pH = 7. Photodegradation experiments were conducted under direct UV irradiation (λ = 254 nm) and under simulated sunlight. Simulated sunlight assays involved both direct and indirect photodegradation experiments. Biodegradation experiments were conducted in activated sewage sludge, soil and for the sediment-water interface of artificial wetland systems to evaluate various environmental compartments. We estimated degradation rates, followed-up the isotopic signatures based on carbon, nitrogen and sulphur stable isotopes and quantified the formation of transformation product using LC-MS. Reference degradation experiments for terbutryn showed that CSIA can be used as concentration-independent tool to identify the dominant degradation processes in the environment by combining (i) the isotopic enrichment of stable isotopes by dual isotope plots and (ii) the pattern of formed transformation products. For carbon, isotope fractionation values range from ε_C= -3.4 ± 0.3 ‰ for abiotic hydrolysis at pH=1 to an inverse isotope effect of ε_C= 0.8 ± 0.4 ‰ in direct photodegradation experiment under UV irradiation, which underscore the potential of terbutryn CSIA to differentiate degradation mechanisms. Biodegradation rates in soil and the sediment-water interface are rather low ($t_{1/2}$ > 200 days), indicating that terbutryn may not be easily biodegraded. Altogether, our study underscores that lab scale experiments are necessary to retrieve reference kinetics and

mechanistic values to follow micropollutant degradation based on CSIA in the environment. It also emphasizes the applicability of CSIA for the urban biocide terbutryn. Reference isotope fractionation values can be used in the future to monitor transport and transformation of terbutryn at urban sites while supporting predictive model development for urban biocide export.