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Reactive transport of micropollutants in laboratory aquifers: combining Compound-Specific Isotope Analysis (CSIA) and biomolecular approaches

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Groundwater quality is of increasing concern due to the ubiquitous release of micropollutants, often originating from surface water. Micropollutants comprise a wide range of substances such as pesticides, pharmaceuticals and personal care products (PCPs), and pose risks to groundwater contamination due to their high persistence and toxicity. Although biodegradation is a major process for the removal of organic contaminants in aquifers, the interplay of hydrogeochemical conditions, microbial diversity and micropollutant dissipation at low concentrations remains yet poorly understood. We developed here an integrative approach to understand and predict the factors affecting micropollutant dissipation within the surface-/ground-water transition zone. Compound-specific Isotope Analysis (CSIA) was used to evaluate micropollutant transformation based on changes in the ratio of stable isotopes (i.e., $^{13}\text{C}/^{12}\text{C}$ and $^{15}\text{N}/^{14}\text{N}$). The responses of aquifer microbes – the key players during contaminant transformation – to micropollutant exposures was examined through biomolecular approaches, proving advantageous in combination with CSIA.

We examined the dissipation of a micropollutant mixture in two lab-scale aquifer systems fed with river water collected from an agricultural area, thus representing the highly reactive surface-/ground-water transition zone. The micropollutant mixture included legacy and currently used pesticides such as atrazine, terbutryn, S-metolachlor and metalaxyl. Caffeine and metformin were also examined as anthropogenic compounds with physico-chemical properties close to currently used pesticides. Changes in bacterial diversity was examined in both aquifer systems during variations of micropollutant exposures under static hydrological conditions. It is hypothesized that such variations may induce bacterial changes and thus alter micropollutant transformation pathways. To this end, three periods of micropollutant injections during 140 d were induced as follow: (i) a first pulse (about 25 μM) to identify dissipation processes and bacterial adaptation to micropollutants, (ii) a constant injection (2 pore volumes) at 10 fold lower concentrations (chronic exposure phase), and (iii) a second pulse injection (25 μM) to examine whether transformation of micropollutants was enhanced. Concentration breakthrough curves (BTCs) of atrazine, terbutryn and metaxyl showed sorption as the main dissipation process for the three periods, whereas both sorption and degradation were observed for caffeine and S-

metolachlor. Carbon and nitrogen CSIA further supported the *in situ* transformation of caffeine and S-metolachlor ($\Delta\delta^{13}\text{C}$ of $\geq 4\text{‰}$ and $\geq 2\text{‰}$, respectively), while no significant enrichment of ^{13}C and ^{15}N were observed for atrazine, terbutryn and metalaxyl ($\Delta\delta^{13}\text{C} < 2\text{‰}$). In parallel, surface-water microcosm experiments showed half-life times of atrazine, terbutryn and metalaxyl of >200 days. Microbial diversity is currently examined in pore water and sand samples. A numerical model is under development to improve the interpretation of micropollutant dissipation in the highly reactive surface-/ground-water transition zone based on concentrations, CSIA and bacteria diversity data obtained in this study. Altogether, our results demonstrated degradation capacity within the laboratory systems, mainly for caffeine and S-metolachlor, and highlight the persistence and risk to long-term groundwater contamination of both legacy and currently used pesticides.