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Pesticide degradation and export losses at the catchment scale: insights from compound-specific isotope analysis (CSIA) and distributed catchment models

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Pesticides in the environment is a worldwide issue of increasing concern. Determining pesticide fate in the field remains a challenge that may preclude adapted risk assessment and water resource management. Application of compound-specific isotope analysis (CSIA) to evaluate degradation of diffuse and low-concentration pesticides in surface hydrological contexts is mostly recent. Compound-specific isotope analysis (CSIA) can provide evidence of contaminant degradation extent, as it is generally independent of non-destructive dissipation (e.g., dilution, sorption, volatilization) regulating environmental concentrations. This study demonstrates the applicability of CSIA to track pesticide degradation and export at catchment scale and identify pesticide source areas contributing to changes in stable isotope signature in stream discharge under dynamic hydrological contexts. Based on maximum change in carbon stable isotope signatures ($\Delta\delta$13C = 4.6 ± 0.5 ‰ of S-metolachlor (S-met), a widely used herbicide, we estimate maximum degradation to have reached 96 ± 3% two months after first application. Maximum shifts in nitrogen isotope signatures were small and inverse ($\Delta\delta$15N = -1.3 ± 0.6 ‰ indicating potential secondary isotope effects during degradation. In combination with a mass balance approach including S-met main degradation products, total catchment non-destructive dissipation was estimated to have reached 8 ± 7% of the applied product. Our results show that CSIA can be applied to evaluate natural attenuation of pesticides at catchment scale. By providing a more detailed account of pesticide dissipation and persistence under field conditions we anticipate the contribution of pesticide CSIA to the improvement of regulatory and monitoring strategies.

However, field data distinguishing degradative from non-degradative pesticide dissipation processes for model calibration are scarce. Here control of degradation half-life (DT50) by topsoil water content and temperature was introduced in the pesticide degradation and transport component of a distributed model, and further tested in a 47-ha agricultural catchment. Controlled DT50 values improved predictions of S-MET degradation across the catchment and the agricultural season, and decreased prediction uncertainties (i.e. 95% confidence intervals) of top 12 soil pesticide leaching by a factor of 2. In contrast, constant DT50 values, independent of hydro-climatic conditions, increased prediction uncertainties. CSIA data constrained DT50 values and allowed to validate model predictions of S-MET degradation in soil and off-site export from the catchment. S-MET stable isotope data and concentrations reduced model equifinality by teasing apart degradative and non-degradative S-MET dissipation processes. The range of DT50 values was reduced by a factor 2 when including S-MET stable isotope data into the calibration process. Pesticide CSIA data from topsoil collected weekly across the catchment helped to constrain S-MET degradation in the PiBEACH model. We anticipate our results as a preliminary step to develop daily long-term continuous models that include hydro-climatic control of pesticide degradation. More reliable predictions of pesticide degradation dynamics at catchment scale may eventually guide pesticide risk assessment and management practices.