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Potential of diclofenac C+N isotope analysis: source signatures and degradation pathway dependent fractionation

M. Maier (1), S. De Corte (2), N. Boon (2), and M. Elsner (1)

(1) Helmholtz-Centre Munich, Institute of Groundwater Ecology, Neuherberg, Germany (michael.maier@helmholtz-muenchen.de), (2) Laboratory of Microbial Ecology and Technology (LabMET), Ghent University, Belgium

The widespread application of the anti-inflammatory and analgesic diclofenac comes along with two problems. First, numerous studies have shown its appearance in aquatic systems while its degradability in the environment is still unclear. Second, there is a growing market for faked pharmaceuticals and the increasing quality of fakes makes it difficult for producers to prove counterfeiting. To address the first problem it is rather challenging to determine the degradability of diclofenac in the environment solely from concentration measurements, since to this end also hydrological data is needed. In contrast, isotope ratios are independent of the hydrology (e.g. dilution effects) and can even deliver insights into reaction mechanisms. However, currently there is no method to determine compound specific isotope ratios of diclofenac at natural abundance. If such a method was available, it would also have the potential to verify different diclofenac products by analyzing isotope values, which are an intrinsic and hardly forgeable attribute of every compound.

In this study we developed a GC-IRMS (gas-chromatography coupled to isotope-ratio-mass-spectrometry) method to analyze carbon and nitrogen isotope values of diclofenac and tested its applicability to identify producers and to investigate degradation processes. The method was validated by measuring different diclofenac products, whose extracts were analyzed by EA-IRMS in parallel (elemental-analysis coupled to isotope-ratio-mass-spectrometry). Although the eight tested products were distinguishable in a two-dimensional isotope plot (carbon vs. nitrogen isotopes), the absolute values were quite close. Even if our survey is not complete it indicates low source variability. This makes it easier to attribute isotope values measured in the environment to degradation.

Isotope fractionation was monitored in two degradation experiments. In an aerobic water-sediment-test-system nitrogen isotopes showed an enrichment factor of ε_N = -6.4‰ while no carbon isotope fractionation occurred during degradation. This suggests that a transformation pathway occurred where the initial reaction takes place at the N-atom. In a second experiment diclofenac was dechlorinated with Pd-/Au coated microorganisms (S. *oneidensis*). The measured bulk carbon isotope enrichment for the whole molecule was ε_C = -2‰This shows that even in a molecule containing 14 C-atoms carbon isotope fractionation can be determined.

In conclusion the two-dimensional isotope analysis of diclofenac can be used to investigate its degradation in-situ. Additionally the two tested transformation pathways could be clearly distinguished by analyzing carbon and nitrogen isotopes values.