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Impact of substrate bioavailability on stable isotope fractionation during biodegradation in environmental systems

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The microbial degradation of organic substrates often exhibits a fractionation of stable isotopes which leads to an enrichment of the heavier isotope in the remaining substrate. The use of this effect to quantify the amount of biodegraded substrate in contaminated aquifers requires that the isotope fractionation factor is constant in time and space. In many natural and engineered systems the bioavailable concentration at the location of the enzymes differs from the average bulk concentration of the substrate. When enzymatically driven substrate degradation is coupled to a preceding transport step controlling the bioavailability of the substrate, the apparent isotope fractionation becomes a function of the bulk substrate concentration. The sensitivity of the apparent isotope fractionation factor towards such substrate concentration changes depends on the ratio of bulk substrate concentration and Michaelis-Menten constant, and on the ratio between the specific affinity of the microorganisms towards the substrate and the first order rate constant of the bioavailability limiting transport process. As a consequence, even in controlled systems changes in concentration and isotopic composition of a bioavailability limited substrate should not follow the relation predicted by the Rayleigh model but should show a decreasing apparent isotope fractionation factor with concentration decrease. A theoretical analysis of this effect is compared with results from laboratory experiments.