Effect of corrinoid cofactor type on the reductive dehalogenation and protonation reaction

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During the last decades several bacterial strains were isolated capable of organohalide respiration. However, the actual enzymatic reaction mechanism and the role of the corrinoid cofactor is still a point of interest. The type of corrinoid-cofactor is suspected to have a major influence on the substrate spectrum of the enzyme, as well as its degradation rate. Therefore, we investigated and characterized the reductive dehalogenation and protonation reaction with respect to different corrinoid-cofactor using compound-specific stable isotope analysis (CSIA) of carbon, chlorine and hydrogen.

In order to characterize the reaction, stable isotope fractionation of chlorinated ethenes during dehalogenation and subsequential protonation was studied for different commercially available corrinoid cofactors, as well as native cofactors and compared to enzymatic reaction of several strains capable of reductive dehalogenation.

During dehalogenation the stable isotope fractionation of both carbon and chlorine for TCE was the same for all investigated systems, confirming the similarity in reaction. In contrast, dehalogenation of PCE was observed to be different for enzymes in comparison to all corrinoids, and surprisingly, corrinoid-cofactors could be differentiated according to their structure. In addition, all protonated products were measured to be highly depleted in deuterium isotopes, whereas abiotic reaction products were measured to be significantly more depleted in deuterium then enzymatic ones. Therefore, the structures of the corrinoid-cofactor, as well as the enzyme appear to affect the dehalogenation reaction mechanism to a certain extent.