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## Reactive transport of dichloromethane in laboratory aquifers: insights from dual-element isotope analysis and biomolecular approaches

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Dichloromethane (DCM) is a toxic industrial solvent frequently detected in multi-contaminated aquifers. DCM often co-occurs with chlorinated ethenes resulting in complex mixtures posing challenges to predict its fate in groundwater. Changes in hydrochemistry and redox conditions in groundwater due to fluctuations in the water table may affect the extent and pathways of pollutant biodegradation. In this context, Compound-Specific Isotope Analysis (CSIA) is a useful tool to evaluate natural degradation of halogenated hydrocarbons. In this study, the impact of water table fluctuations on DCM biodegradation was examined in two laboratory aquifers using dual-element isotope analysis - the stable isotope fractionation of two elements (e.g., <sup>13</sup>C and <sup>37</sup>Cl), and high-throughput biomolecular approaches. The aquifers were supplied with contaminated groundwater from the former industrial site Thermeroil (France). High-resolution sampling and monitoring of pore water allowed examining, under steady and transient conditions, the aquifers response with respect to hydrochemistry and microbial composition. A dual C-Cl stable isotope approach ( $\Lambda^{C/Cl} = \Delta\delta^{13}C/\Delta\delta^{37}Cl$ ) was developed using GC-IRMS (C-DCM) and GC-MS (Cl-DCM) to estimate the extent of DCM degradation and to identify DCM degradation pathways. Under the experimental steady conditions, dissolved oxygen (<1.2 mg/L) and increasing Fe<sup>2+</sup> concentrations at lower depths of the aquifer models indicated iron-reducing prevailing conditions, while mass transfer of oxygen increased during water table fluctuations. Pronounced carbon isotope fractionation of DCM was associated with larger DCM mass removal under transient conditions (>90%) compared to steady conditions (mass removal of 35%). Under transient conditions, carbon enrichment factors ( $\epsilon_C$ ) became larger over time ranging from  $-18.9 \pm 3.4\text{‰}$  to  $-33 \pm 0.3\text{‰}$  whereas chlorine enrichment factors ( $\epsilon_{Cl}$ ) remained constant ( $-3.6 \pm 0.7\text{‰}$ ). In contrast, a similar  $\epsilon_C$  of  $-20 \pm 3.5\text{‰}$  (beginning of transient condition) but a larger  $\epsilon_{Cl}$  of  $-10.8 \pm 2\text{‰}$  were determined under steady conditions. As  $\Lambda^{C/Cl}$  values are independent of complicating masking effects, and thus reflect reaction mechanisms, dual C-Cl isotope plots suggested distinct DCM degradation pathways under steady and transient conditions with  $\Lambda^{C/Cl}$  values of  $1.68 \pm 0.26$  and  $3.41 \pm 0.50$ , respectively. Even though a contribution of different mechanisms may take place during transient conditions,  $\Lambda^{C/Cl}$  values fall in the range of S<sub>N</sub>1 pathways reported for *Cα*. Dichloromethanomonas elyunquensis

( $\Lambda^{C/Cl} = 3.40 \pm 0.03$ ). The distinct  $\Lambda^{C/Cl}$  values may imply mechanistically distinct C-Cl bond cleavage reactions subjected to microbial adaptations during dynamic hydrogeological conditions. Although bacterial communities did not significantly change over time, the occurrence of *Geobacter* under both steady and transient conditions supports DCM degradation under iron-reducing prevailing conditions. Altogether, our results highlight that water table fluctuations enhance DCM biodegradation and influence DCM degradation pathways compared to steady conditions. This integrative study provides new insights into *in situ* degradation of DCM in contaminated aquifers and accounts the effects of dynamic water tables on DCM degradation.