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Influence of Freeze-Thaw Cycles on Methanogenic Hydrocarbon Degradation: Experiment and Numerical Simulation

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In hydrocarbon-*contaminated soils* in *cold regions*, freeze-thaw cycles (FTCs) modify the biogeochemical and physical processes controlling petroleum hydrocarbons (PHCs) biodegradation and the generation of associated by-products methane (CH_4) and carbon dioxide (CO_2). Thus, understanding the effects of FTCs on the soil biodegradation of PHCs is critical for the environmental risk assessment and the design of remediation strategies for contaminated soils in cold regions. In this study, we developed a diffusion-reaction model that accounts for the effects of FTCs on methanogenic toluene biodegradation. The model is verified against data generated in a 200 day-long batch experiment with soil collected from a PHC contaminated site in Canada. The fully saturated soil was exposed to successive 4-week FTCs under anoxic conditions with temperatures fluctuating between -10°C and +15°C. We measured the headspace concentrations and ^{13}C compositions of CH_4 and CO_2 and analyzed the porewater for acetate, sulfate, dissolved organic and inorganic carbon, and toluene concentrations. The numerical model represents solute diffusion, diffusion and volatilization, sorption, as well as a reaction network of 14 biogeochemical processes. The model successfully simulates the soil porewater and headspace concentration time series data by representing the temperature dependencies of microbial reaction and gas diffusion rates during FTCs. According to the model results, the observed increases in the headspace concentrations of CH_4 and CO_2 by 87% and 175%, respectively, following toluene addition are explained by toluene fermentation and subsequent methanogenesis reactions. The experimental results and numerical simulations both confirm that methanogenic degradation is the dominant attenuation process for toluene in anoxic, electron-acceptor limited soil. Also, the model-predicted contribution of acetate-based methanogenesis to total produced CH_4 agrees with that derived from the ^{13}C isotope data. The simulation results of a no FTC scenario indicate that, in the absence of FTCs, there is lower CO_2 and CH_4 production and that toluene is biodegraded faster. Given its ability to represent the dominant processes controlling CH_4 and CO_2 fluxes and porewater chemical changes, our modelling approach can be used to simulate the sensitivity of soil biodegradation processes to FTC frequency and duration driven by temperature fluctuations.