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Natural Source Zone Depletion of Petroleum Hydrocarbons in Response to Water Table Fluctuations

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In hydrocarbon-contaminated soils, water table fluctuations (WTFs) affect the kinetics of petroleum hydrocarbons (PHC) biodegradation and the generation and transport of carbon dioxide (CO_2) and methane (CH_4). Thus, understanding the impacts of WTFs on natural source zone depletion (NSZD) of PHCs is critical for environmental risk assessment and the design of remediation strategies for contaminated soils and groundwater. In this study, a 300 day-long column experiment was conducted to simulate the effects of WTFs on the aerobic and anaerobic PHC biodegradation pathways. Eight columns were each filled with 45 cm of PHC contaminated soil cores. Four columns simulating WTFs were subjected to cycles of drainage and imbibition. The remaining columns remained fully saturated, simulating a static water table. Except for the controls, the columns received injections of ethanol or ethanol plus naphthalene after 111 days of pre-equilibration. Over the duration of the experiment, soil moisture, surface CO_2 and CH_4 effluxes, dissolved concentrations and $\delta^{13}C$ compositions of CO_2 and CH_4, dissolved naphthalene concentrations, and geochemical parameters were monitored. A reactive transport model representing 13 biogeochemical reaction pathways was verified against the acquired data. The experimental and modeling results confirmed that the prevailing pathway generating CH_4 shifted from hydrogen-based to acetate-based methanogenesis in the ethanol and ethanol plus naphthalene spiked columns, while CH_4 oxidation played a key role in controlling the CH_4 efflux during the drainage periods. The WTF columns exhibited significantly faster naphthalene attenuation while the cumulative CO_2 and CH_4 effluxes were about twice as high. These observations were attributed to the periodic incursion of air during the WTFs, which increased the porewater-air interface area for gas transfer while also accelerating the aerobic degradation of soil organic matter and naphthalene. Overall, this study advances the quantitative modeling of the biogeochemical reaction network for NSZD of PHCs in contaminated soils under WTFs, including the role of methanogenic pathways.