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Implications of Seasonality for NSZD Rate Measurements

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Background/Objectives. Despite significant attention to developing new approaches, monitoring and managing hydrocarbon impacted sites remains challenging. This is particularly pronounced at both remote sites and sites where engineered remediation is reaching ‘end of life’ performance. In these cases, natural source zone depletion (NSZD) may be an acceptable management option. However, in order to obtain regulatory acceptance, NSZD processes and mass removal rates need to be documented. Recently, the use of CO_2 flux measurements for estimating NSZD rates has become increasingly accepted; however, questions remain as to how seasonal processes affect these measurements, and guidance is needed for determining when sampling should be conducted. To address these issues, a two-year study was conducted at the National Crude Oil Spill Fate and Natural Attenuation Research Site near Bemidji, MN.

Approach/Activities. We conducted a two-year field study to investigate seasonal variations in the distribution and rate estimates of NSZD processes at a subsurface crude oil spill site. We evaluated NSZD rates using surficial CO_2 efflux measurements, complemented with the collection of shallow soil gas samples, analyzed for $^{14}CO_2$. To understand how seasonal processes affected the surficial CO_2 efflux measurements and time-integrated estimates of NSZD mass loss rates, we assessed seasonal variations in soil temperature, soil volumetric water content, and vadose zone gas composition. Our specific objectives included: (i) evaluating the magnitude and representativeness of monthly soil CO_2 efflux measurements for estimating NSZD rates; (ii) comparing mean annual NSZD rates determined from monthly CO_2 efflux measurements with NSZD rates determined from summer-only measurements; and (iii) evaluating the seasonality and importance of environmental processes (rainfall and temperature) in controlling CO_2 effluxes measured at the ground surface.

Results/Lessons Learned. Results showed strong seasonal variations in surficial CO_2 effluxes, and to a lesser extent, NSZD rate estimates. Largest CO_2 fluxes were measured in the late spring and summer, while values declined in the fall and were lowest in the winter. Both surficial CO_2 effluxes and radiocarbon data indicated that the fractional contributions of natural soil respiration and NSZD to the total CO_2 efflux varied seasonally. Subsurface gas concentrations also varied seasonally, with the highest values of CO_2 and CH_4 occurring in the fall and winter. Comparison of vadose zone temperatures and subsurface CO_2 concentrations revealed a correlation between NSZD rate estimates and temperature. In contrast, a time lag of 5 to 7 months was observed between peak subsurface CO_2 concentrations and peak surface CO_2 efflux, consistent with travel-time estimates for gas migration to the surface. Results also revealed that periods of frozen soils coincided with depressed surface CO_2 effluxes and elevated subsurface CO_2 concentrations, pointing to the temporary presence of an ice layer that inhibited gas transport. Quantitative reactive transport simulations were used to support the conceptual model developed from field measurements. Overall, results indicated that NSZD rate estimates and gas transport processes varied seasonally. Additionally, the average annual NSZD rate estimated from surface efflux measurements was 60% lower than the rate estimated from summer measurements only.