

Understanding methane reactivity in the critical zone using abandoned oil and gas wells

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Methane is a potent greenhouse gas, but the controls on methane fluxes to the atmosphere from the critical zone, including from groundwaters, are poorly constrained. Methane in groundwater has also attracted public and scientific concern as the most cited water quality impact of oil and gas extraction. While the potential sources of methane in shallow groundwater are relatively well constrained, the reactivity of methane is not. Through field and lab observations, we found evidence for anaerobic oxidation of methane (AOM) coupled to iron or sulfate reduction when methane leaked from abandoned oil and gas wells mixes with groundwater at shallow depths. These microbially-mediated reactions consume methane, but can also produce changes to aquifer redox chemistry that produce or mobilize hazardous species. However, the extent of methane consumption and secondary changes to aqueous geochemistry likely depend on the rates of iron vs. sulfate reduction and the reaction of their product species (ferrous iron and sulfide, respectively).

We used lab experiments and constructed reactive transport models to explore how the movement of methane and groundwater, along with pre-existing aquifer geochemistry, influence these redox reactions consuming methane within the critical zone. When the rate at which methane enters the system is limiting to AOM, Fe reduction (the more thermodynamically favorable reaction) predominates and we observe high concentrations of ferrous iron and low concentrations of reduced sulfur. If methane fluxes are non-limiting with respect to AOM, the equilibrium state of the aquifer is dictated by the availability of reducible iron vs. sulfate, which are influenced by pre-existing aquifer mineralogy and groundwater discharge, respectively. Understanding AOM in the critical zone is important for better constraining how methane leaks from modern or legacy oil and gas operations alter groundwater chemistry, as well as the reactivity of methane in shallow aquifers in general.