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The subsurface sulfur system following hydraulic stimulation of unconventional hydrocarbon reservoirs: Assessing anthropogenic influences on microbial sulfate reduction in the deep subsurface

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Hydraulic fracturing is a reservoir stimulation technique that involves the injection of high-pressure fluids to enhance recovery from unconventional hydrocarbon reservoirs. Often this involves the injection of surface waters (along with additives such as biocides) into formational fluids significantly different isotopic and geochemical compositions facilitating geochemical fingerprinting of these fluid sources. In some instances, the produced fluids experience an increase in hydrogen sulfide (H_2S) concentration over the course of production resulting in an increased risk to health and safety, the environment, and infrastructure due to the toxic and corrosive nature of H_2S. However, questions remain as to the origin and processes leading to H_2S formation following hydraulic fracturing. In this study, we analyzed a series of produced waters following hydraulic fracturing of a horizontal well completed in the Montney Formation, Western Canada to evaluate variations in geochemical and microbiological composition over time and characterize potential sulfur species involved in the production of H_2S. Initially, sulfur isotope ratios ($\delta^{34}S$, VCDT) of dissolved sulfate in produced water had a baseline value of 27‰ similar to the $\delta^{34}S$ value of 25‰ for solid anhydrite derived from core material. Subsequently, $\delta^{34}S$ values of sulfate in produced fluids sequentially increased to 35‰ coincident with the appearance of sulfides in produced waters with a $\delta^{34}S_{H2S}$ value of 18‰. Oxygen isotope values of dissolved sulfate exhibited a synchronous increase from 13.2‰ to 15.8‰ VSMOW suggesting sulfate reduction commenced in the subsurface following hydraulic fracturing. Formation temperatures are <100°C precluding thermochemical sulfate reduction as a potential mechanism for H_2S production. We suggest that microbial reduction of anhydrite-derived sulfate within the formation is likely responsible for the increase in H_2S within produced waters despite the use of biocides within the hydraulic fracturing fluids. Initial assessments of microbial communities indicate a shift in community diversity over time and interactions between *in situ* communities and those introduced during the hydraulic fracturing process. This study indicates that biocides may not be fully effective in inhibiting microbial sulfate reduction and highlights the role anthropogenic influences such as hydraulic fracturing can have on the generation of H_2S in the subsurface.