characterization of radium and radon isotopes in hydraulic fracturing flowback fluid and gas from the Marcellus Shale

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High volume hydraulic fracturing of unconventional deposits has expanded rapidly over the past decade in the US, with much attention focused on the Marcellus Shale gas reservoir in the northeastern US. We use naturally occurring radium isotopes and 222Rn to explore changes in formation characteristics as a result of hydraulic fracturing. Gas and produced waters were analyzed from time series samples collected soon after hydraulic fracturing at two Marcellus Shale well sites in the Appalachian Basin, USA. Analyses of $\delta^{18}O$, Cl^- , and 226Ra in flowback fluid were consistent with two end member mixing between injected slick water and formation brine. All three tracers indicate that the ratio of injection water to formation brine declined with time across both time series. There were substantial differences in Cl^- between the two wells samples with the highest concentrations reaching 2 M, while 226Ra activity was similar at both wells. On a plot of water isotopes, $\delta^{18}O$ in formation brine-dominated fluid was enriched by approximately 3 permille relative to the Global Meteoric Water Line, indicating oxygen exchange with shale. The ratio of 223Ra/226Ra and 228Ra/226Ra in produced waters was quite low relative to shale samples. This indicates that most of the 226Ra in the formation brine must be sourced from shale weathering or dissolution rather than emanation due to alpha recoil from the rock surface. During the first week of flowback, ratios of short lived isotopes 223Ra and 224Ra to longer lived radium isotopes changed modestly, suggesting rock surface area per unit of produced water volume did not change substantially. The 222Rn/methane ratio in produced gas declined with time and may represent a decrease in the brine to gas ratio in the reservoir over the course of six months after initial fracturing. Naturally occurring radium and radon isotopes show promise in elucidating sub-surface dynamics following hydraulic fracturing plays.