

Isotopic and Geochemical Evidence for the Freshening of Shale-Gas Produced Waters through Time Due to Water Vapor Condensation, Northeastern Pennsylvania, USA

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Abstract

The chemistry of waters co-produced with Marcellus shale gas have been reported to reflect variable mixtures of the original injection fluids, formation waters within the Marcellus (or other adjacent formations), and potentially, mobilized chemical constituents from the shale formation itself. In addition, time-series data reported in the scientific literature have commonly demonstrated an increase in the salinity of produced waters over time. Increasing produced water salinity has often been interpreted to reflect the dissipation of stimulation fluid returned to surface in the months to years post-stimulation and a concomitant increase in the relative proportion of formation fluids. However, at a subset of Middle Devonian Marcellus shale gas wells that initially produce waters with higher salinity than the injection fluids, produced water chemistry becomes less saline over time. Specifically, these wells exhibit decreasing concentrations of total dissolved solids, chloride, strontium, and barium. We hypothesize that this decrease in salinity over time is a function of water condensing out of the gas phase as it travels to the surface. Condensed water may dominate produced water composition once the daily volume of produced formation fluids and/or injection fluids has fallen below a certain threshold relative to the volume of gas. For the first time, we present time-series $\delta^{18}\text{O}$ and δD of produced waters from Marcellus shale gas wells in the northeastern Appalachian Basin which exhibit patterns that explain the role of water condensation. Specifically, water isotope data of increasingly fresh

produced water samples show trends inconsistent with mixing between formation fluids and injection fluids (where the latter plots on the local meteoric water line). Instead, the $\delta^{18}\text{O}$ and δD values of these produced waters are best explained by the condensation of water vapors in the gas phase near the surface.