The source and migration of natural gas in shallow aquifers: Insights provided by the integration of noble gas and hydrocarbon isotopes

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Horizontal drilling and hydraulic fracturing have enhanced energy production but raised concerns over drinking-water contamination and other potential health risks. Specifically, the presence and environmental implications of elevated methane and aliphatic hydrocarbons (ethane, propane, etc.) in drinking-water remain highly controversial and require a distinction between naturally occurring and anthropogenic sources. Previous efforts to resolve these questions have generally focused on identification of the genetic fingerprint of natural gas using the molecular (e.g., C_2H_6/CH_4) and stable isotopic (e.g., $\delta^{13}C\text{-}CH_4$, $\delta^{2}H\text{-}CH_4$, or $\Delta^{13}C=(\delta^{13}C\text{-}CH_4 - \delta^{13}C\text{-}C_2H_6)$) compositions of hydrocarbon gases. In many cases, these techniques can resolve thermogenic and biogenic contributions of natural gas and further differentiate between multiple thermogenic sources (e.g., Marcellus production gases vs. intermediate Upper Devonian gas pockets). However, these parameters are subject to alteration by microbial activity and oxidation and may not always uniquely identify the source or mechanism of fluid migration. Moreover, they do not necessarily identify the transport mechanisms by which material would migrate into shallow aquifers. In contrast to hydrocarbon gases, noble gases provide a suite of elemental and isotopic tracers that are unaffected by chemical reactions or microbial activity. Here we develop an integrated noble gas and hydrocarbon isotope analysis to evaluate if elevated levels of natural gas in drinking-water aquifers near gas wells are derived from natural or anthropogenic sources and to determine the mechanism by which stray gas contamination occurs.