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Clumped isotope approaches to evaluate the sources, potential migration and fate of methane and ethane in sedimentary basins

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Throughout the last few decades, chemical and isotopic ($\delta^{13}C$ and $\delta^{2}H$) compositions have been widely used to differentiate thermogenic sources of methane and ethane from unconventional and conventional hydrocarbon reservoirs from methane in dissolved or free gas of shallow aquifers that is frequently of microbial origin. Nevertheless, accurately determining the occurrence and effects of the occasional migration of gases from the hydrocarbon reservoirs or the intermediate zone towards shallow aquifers and the atmosphere along imperfectly sealed boreholes or natural pathways remains challenging. This is especially true in environments where the oxidation of methane obscures the isotopic fingerprint of the source signal. Therefore, improved separation of potential fugitive gases from hydrocarbon reservoirs from natural occurrence and migration of methane and ethane and subsequent alteration in shallow aquifers can benefit from an integrated understanding of the geological and hydrological evolution of hydrocarbons in the subsurface over time and additional tracer approaches. Clumped isotopologues of methane ($\Delta^{13}CH_3D$ and $\Delta^{12}CH_2D_2$) combined with chemical and traditional stable isotope compositions of gas hydrocarbon from Western Canada were used for improved characterization of methane and ethane associated with unconventional hydrocarbon reservoirs. Similar analyses in dissolved and free gases from numerous groundwater samples revealed no widespread evidence that methane and ethane from hydrocarbon reservoirs have impacted the geochemistry of shallow groundwater systems. Instead, it was found that dissolved and free gases in shallow aquifers are dominated by microbial methane that can be markedly affected by methane oxidation. Only in a few cases, gases with apparent thermogenic signatures were observed in shallow groundwater. Trace amounts of ethane were observed in many methane-rich groundwater samples and co-production of traces of ethane by methanogens is a plausible explanation. Alternately, ethane found in groundwater samples may also be associated with early mature thermogenic gas or with radiolytic gas from organic matter in uranium-rich shales. This study has provided new insights into the complicated interplay between multiple sources of methane and ethane and their fate in shallow aquifers above sedimentary basins containing hydrocarbon resources.