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Occurrence and Sources of Radium in Groundwater Associated with Oil Fields in the Southern San Joaquin Valley, California

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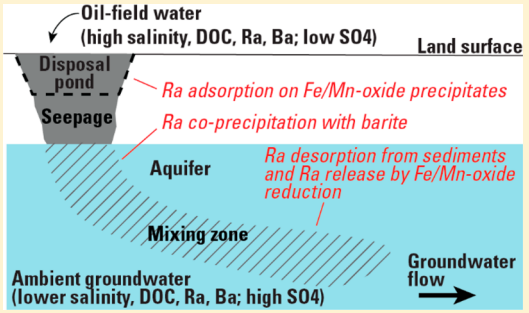
Supporting Information

ABSTRACT: Geochemical data from 40 water wells were used to examine the occurrence and sources of radium (Ra) in groundwater associated with three oil fields in California (Fruitvale, Lost Hills, South Belridge). $^{226}Ra+^{228}Ra$ activities (range = 0.010−0.51 Bq/L) exceeded the 0.185 Bq/L drinking-water standard in 18% of the wells (not drinking-water wells). Radium activities were correlated with TDS concentrations ($p < 0.001$, $\rho = 0.90$, range = 145−15,900 mg/L), Mn + Fe concentrations ($p < 0.001$, $\rho = 0.82$, range = <0.005−18.5 mg/L), and pH ($p < 0.001$, $\rho = -0.67$, range = 6.2−9.2), indicating Ra in groundwater was influenced by salinity, redox, and pH. Ra-rich groundwater was mixed with up to 45% oil-field water at some locations, primarily infiltrating through unlined disposal ponds, based on Cl, Li, noble-gas, and other data. Yet $^{228}Ra/^{226}Ra$ ratios in pond-impacted groundwater (median = 3.1) differed from those in oil-field water (median = 0.51). PHREEQC mixing calculations and spatial geochemical variations suggest that the Ra in the oil-field water was removed by coprecipitation with secondary barite and adsorption on Mn−Fe precipitates in the near-pond environment. The saline, organic-rich oil-field water subsequently mobilized Ra from downgradient aquifer sediments via Ra-desorption and Mn/Fe-reduction processes. This study demonstrates that infiltration of oil-field water may leach Ra into groundwater by changing salinity and redox conditions in the subsurface rather than by mixing with a high-Ra source.



INTRODUCTION

Consumption of water containing elevated radium (Ra) activities has been associated with various adverse human-health effects, including some forms of cancer.[1−3] The U.S. Environmental Protection Agency established a maximum contaminant level (MCL) for $^{226}Ra+^{228}Ra$ in drinking water of 0.185 Bq/L (5 pCi/L).[4] Previous studies have shown that water in some hydrocarbon reservoirs is enriched in Ra nuclides, which could be problematic if that water mixes with nearby groundwater. $^{226}Ra+^{228}Ra$ activities up to 666 and 64 Bq/L were reported in produced water from unconventional hydrocarbon reservoirs in the Marcellus Shale and Bakken Formation, respectively.[5,6] Produced water from oil reservoirs in the southern San Joaquin Valley (SJV), California, has reported $^{226}Ra+^{228}Ra$ activities up to ∼12 Bq/L.[7,8] Differences in Ra activities between water from the SJV and other reservoirs reflect, in part, differences in salinity between the reservoirs. At elevated salinities, exchangeable Ra on clay minerals can be mobilized due to exchange with other dissolved ions.[9−11] While concentrations of total dissolved solids (TDS) in water from SJV oil reservoirs are typically <40,000 mg/L,[8] TDS in water from the Bakken and Marcellus commonly exceed 100,000 mg/L.[5,6]

The focus of this study is shallow groundwater associated with the Fruitvale (FV), Lost Hills (LH), and South Belridge (SB) oil fields in the SJV (Figure 1),[12] where oil production has occurred for ∼100 years.[8] Disposal of oil-field water in unlined ponds has occurred in parts of the study area since the 1950s and is a direct pathway for oil-field water to enter the near-surface environment.[13] Several studies have reported the presence of Ra from oil-field water in near-surface environments, typically in aquatic sediment or soil associated with releases of Ra-rich produced water.[6,14−16] Those studies found most of the Ra was retained on solid phases relatively close to the release site due to Ra immobilization by processes like coprecipitation with barite ($BaSO_4$) and adsorption on solid phases.[6,14,15] In groundwater

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