Radium isotope quartet in groundwater as a proxy for identification of aquifer rocks and mechanisms of water-rock interactions: examples from the Negev, Israel

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Many aquifer systems are composed of multiple rock types. Previous attempts to evaluate the specific aquifer rocks that control the groundwater chemistry and possible flow paths within these multiple lithological systems have used major ion chemistry and isotopic tracers (e.g., strontium isotopes). Here we propose an additional isotopic proxy that is based on the distribution of radium isotopes in groundwater. Radium has four radioactive isotopes that are part of the decay chains of uranium-238, thorium-232, and uranium-235. The abundance of radium isotope quartet (^{226}Ra-half life 1600 y; ^{228}Ra-5.6 y; ^{224}Ra-3.6 d; ^{223}Ra-11.4 d) in groundwater reflects the Th/U ratios in the rocks. Investigation of groundwater from the Negev, Israel, enabled us to discriminate between groundwaters flowing in the Lower Cretaceous Nubian Sandstone and the Upper Cretaceous Judea Group carbonate aquifers. Groundwater flowing in the sandstone aquifer has distinguishably high ^{228}Ra/^{226}Ra and ^{224}Ra/^{223}Ra ratios due to the high Th/U ratio in sandstone. In contrast, the predominance of uranium in carbonate rocks results in low ^{228}Ra/^{226}Ra and ^{224}Ra/^{223}Ra ratios in the associated groundwater. We show that the radium activity in groundwater in the two aquifer systems is correlated with temperature, dissolved oxygen, and salinity. The increase of radium activity is also associated with changes in the isotopic ratios; ^{228}Ra/^{226}Ra ratios increase and decrease in the sandstone and carbonate aquifers, respectively. Given that the dissolution of radium isotopes depends on their decay constants, the use of the four radium isotopes with different decay constants enabled us to distinguish between dissolution (higher abundance of the long-lived isotopes) and recoil (predominance of the short-lived isotopes) processes. In spite of these isotopic fractionations, the radium isotopic discrimination between carbonate and sandstone aquifers is significant.