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Uranium and noble gas isotopes to decipher naturally-occuring radionuclide release into aquifers

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Previously unknown relationships between helium isotopes and 234U/238U activity ratios in granular and fractured aquifers of the St. Lawrence Lowlands, Canada might help to resolve the long-standing debate of internal vs. internal sources of natural radionuclides in aquifers. Indeed, radiogenic 4He found in groundwater more than what could be produced in situ during the residence time of water has often been related to basal fluxes entering the bottom of aquifers. Other views suggest that only small portions of accumulated radiogenic helium in aquifer rocks can be released at steady-state. Most of locally produced helium would be released episodically during thermo-tectonic events. Here we show that 4He is correlated with 234U/238U activity ratios in local aquifers of Ordovician age of the St. Lawrence Lowlands, Québec. Groundwater flow and aquifer geometry was largely modified during the last post-glacial episode as observed for most periglacial aquifers of North America. This relation suggests the occurrence of three water masses: 1) freshwater containing atmospheric helium and 234U/238U activity ratios close to the secular equilibrium, indicating local and very shallow recharge; 2) freshwater penetrating the ground, dissolving rocks, and releasing 234U/238U activity ratios slightly higher than the secular equilibrium and large amount of radiogenic helium, indicating that rocks still preserve a reservoir of helium; 3) post-glacial groundwater containing large amount of helium and 234U/238U out of the secular equilibrium, indicating a fossil water having modified its U and He content during water-rock interactions. The U-He relationship is not straightforward. We suggest that post-glacial rebound and increased fracturing has favored the opening of new cracks and increased the specific surface of aquifer rocks. Along these newly created surfaces, He is released by diffusion and 234U by alpha-recoil, increasing the content of these two radionuclides in the water phase. U and noble gases could be a new tool for understanding mechanism of release of radionuclides in aquifers.