Enormous excess ^{223}Ra in fluids from the Lost City hydrothermal field

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We have discovered exceptionally high activities (10-100 dpm/L) of excess ^{223}Ra (half-life = 11 days) in fluids from the Lost City hydrothermal field, a serpentinite-hosted system on the mid-Atlantic Ridge. These activities surpass any published measurements of ^{223}Ra in seawater or other hydrothermal fluids by factors of 50 to 500. In sharp contrast to fluids from basalt-hosted settings, no other radium isotopes have unusually high activities in these fluids. In fact, ^{228}Ra and ^{224}Ra are below detection and ^{226}Ra is ~1 dpm/L. In basalt-hosted systems, high (10-70 dpm/L) ^{226}Ra and ^{228}Ra activities are thought to arise from leaching of the basalt by hydrothermal fluids. For ^{224}Ra and ^{223}Ra, alpha recoil is also important. Basalt-hosted systems have ^{223}Ra/^{226}Ra activity ratios (ARs) <0.05; at Lost City this AR is 10-100, indicating the radium signal is not due to rock alteration. We postulate that the parent of ^{223}Ra, ^{231}Pa (half-life = 34,000 years), is scavenged when seawater circulates through the rocky subsurface, similar to the explanation advanced for high ^{223}Ra activities measured over the Puna Ridge off Hawaii, although the activities measured at Lost City are orders of magnitude higher. The surface-bound ^{231}Pa generates ^{223}Ra, which can be leached by circulating fluids. We measured the emanation of the ^{223}Ra daughter, ^{219}Rn (half-life = 4 sec.), from the surfaces of carbonate chimneys and host rocks and determined that the emanation is supported by 0.04-0.10 dpm/gm of ^{231}Pa in equilibrium with ^{223}Ra. Most of the ^{223}Ra on the surfaces is leached by a simulated hydrothermal fluid. These measurements highlight the fact that serpentine-hosted systems follow a unique chemistry.