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Tracing and Apportioning Sources of Uranium to the Hanford Reach of the Columbia River Using Uranium Isotopes

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The U.S. Department of Energy's Hanford Site is situated along an unimpounded portion of the Columbia River, the highest discharge volume river west of the continental divide. Decades of nuclear related activities have left significant local contamination (e.g. nitrate, U, tritium, Cr⁶⁺, ⁹⁹Tc) in the vadose zone and groundwater within the site. Some of this contamination has reached the Columbia River, and there remains the potential for further contaminant migration to the river. We collected and analyzed samples of Columbia River water for U and Sr isotopes in coordination with the ongoing sampling and monitoring of the river. The U and Sr isotopic data allow us to evaluate sources of U (e.g. natural background, Hanford related, agricultural runoff) and their relative contributions to the river's U budget. The data also provide constraints on the flux of contaminant U from the Hanford Site to the river.

We analyzed two sample traverses across the Columbia, one near the Vernita Bridge, upstream from Hanford Site contamination, and a second about 5 km downstream of the Hanford Site. An island divides the downstream traverse into western (main channel) and eastern portions. Filtered (0.45 micron) water samples were analyzed for U isotopic composition (including ²³⁶U, one marker of spent U fuel) and U concentration, as well as ⁸⁷Sr/⁸⁶Sr and Sr concentration. U isotopes were measured by MC-ICPMS, and Sr isotopes by TIMS. The samples from the upstream traverse had no detectable ²³⁶U ($^{236}\text{U}/^{238}\text{U} < 2 \times 10^{-8}$), natural ²³⁸U/²³⁵U, uniform ²³⁴U/²³⁸U, ⁸⁷Sr/⁸⁶Sr, U and Sr concentrations. In contrast, the downstream traverse showed variation in all of these parameters. Concentrations of U are 0.5 ppb to 1.2 ppb and are all well below the EPA MCL of 30 ppb for drinking water. In the western channel, measured ²³⁶U/²³⁸U is from 3.4×10^{-5} to $< 2 \times 10^{-8}$, with a co-variation in ²³⁸U/²³⁵U toward enriched ratios. This correlation is consistent with the U isotopic compositions of a groundwater sample from a former fuel fabrication complex and samples of groundwater seeps at the adjacent river shore. These compositions indicate a component of 2nd cycle enriched U fuels. For the river water sample with the highest ²³⁶U/²³⁸U, about 30% of the total U comes from Hanford contamination. For the eastern portion of the traverse, no detectable ²³⁶U was found, however ²³⁴U/²³⁸U and ⁸⁷Sr/⁸⁶Sr are significantly different from the Vernita Bridge traverse, likely reflecting contributions from agricultural runoff.