

TRACING SOURCES OF URANIUM TO THE HANFORD REACH OF THE COLUMBIA RIVER

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RESEARCH OBJECTIVES

As part of the World War II-era Manhattan Project, the Hanford Site was established along the Columbia River to take advantage of local hydroelectric power and ample water supply for cooling of a series of nuclear reactors. Decades of nuclear-related activities have left significant local contamination (e.g., nitrate, uranium (U), tritium, Cr^{6+} , ^{99}Tc) 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 strontium (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 and sources of contaminant U from the Hanford Site to the river.

APPROACH

We analyzed three sample traverses across the Columbia, one near the Vernita Bridge, upstream from Hanford Site contamination, a second just downriver of the 300 Area, and a third about 5 km downstream of the Hanford Site, adjacent to the town of Richland. Additional samples were collected up to 350 km downstream. Sampling was conducted in fall 2003, spring 2004, and fall 2004. Filtered (0.45 micron) water samples were analyzed for U isotopic composition (including ^{236}U , one marker of spent U fuel) and U concentration, as well as $^{87}\text{Sr}/^{86}\text{Sr}$ and Sr concentration. Uranium isotopes were measured by multiple-collector-inductively-coupled-plasma source mass spectrometer (MC-ICPMS), and Sr isotopes by thermal ionization mass spectrometry (TIMS).

ACCOMPLISHMENTS

The samples from the upstream traverse had no detectable ^{236}U ($^{236}\text{U}/^{238}\text{U} < 2 \times 10^{-8}$), natural $^{238}\text{U}/^{235}\text{U}$, uniform $^{234}\text{U}/^{238}\text{U}$, $^{87}\text{Sr}/^{86}\text{Sr}$, or U and Sr concentrations. In contrast, the downstream traverses showed variation in all of these parameters. As an example, $^{236}\text{U}/^{238}\text{U}$ data are shown in Figure 1a for the fall 2003 and spring 2004 Richland traverses.

The $^{236}\text{U}/^{238}\text{U}$ highlight a plume of U contamination coming from the Hanford Site. Correlation of $^{236}\text{U}/^{238}\text{U}$ with $^{238}\text{U}/^{235}\text{U}$ is consistent with a component of 2nd cycle enriched U fuels. For the river water sample with the highest $^{236}\text{U}/^{238}\text{U}$, about 30% of the total U comes from Hanford contamination. For the eastern portion of the traverse, no detectable ^{236}U was found (Figure 1a). However, $^{234}\text{U}/^{238}\text{U}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ (not shown) indicate significant contributions from agricultural return canals about 6 km upstream of the traverse. Measurable $^{236}\text{U}/^{238}\text{U}$, though small, was observed in all the downstream samples (Figure 1b), and together with $^{238}\text{U}/^{235}\text{U}$ was consistent with a Hanford Site source.

SIGNIFICANCE OF FINDINGS

The isotopic compositions of the U-contaminated river samples are consistent with a U-contaminated groundwater source in the 300 Area, where fuel elements (both 2nd cycle enriched and natural U) were fabricated. In fact, particular 300 Area groundwater and seep samples are matched as sources, pointing to particular points along the shore.

Both U concentrations and $^{236}\text{U}/^{238}\text{U}$ decrease downstream (Figure 1b), indicating

not only progressive dilution of the Hanford U signature, but also suggesting that at the same time U was being lost from solution (in this case, the <0.45 mm fraction) to particulates. Comparing fall 2003 and spring 2004 Richland traverses, we have found that the flux and apparent source of Hanford U changes with season, with higher relative flux in the fall.

RELATED PUBLICATION

Christensen, J.N., P.E. Dresel, M.E. Conrad, G.W. Patton, and D.J. DePaolo, Tracing and apportioning sources of uranium to the Hanford reach of the Columbia River using uranium isotopes. *Eos Trans. AGU*, 85(47), F858, 2004.

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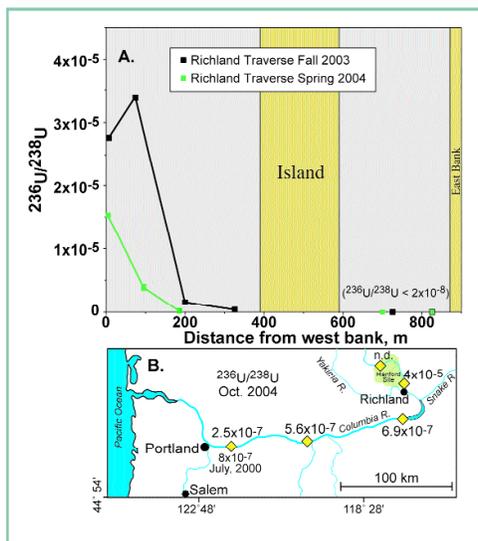


Figure 1. (a) River sampling traverse across the Columbia River at the Richland pump house, about 5 km downstream of the Hanford Site 300 Area. Shown are $^{236}\text{U}/^{238}\text{U}$ data for sampling in fall 2003 and spring 2004. The sampling traverse is divided by an island. (b) Map showing locations of the Hanford Site and downstream sampling sites along the Columbia River, with indicated $^{236}\text{U}/^{238}\text{U}$. N.D. = not detected.

