

Detection of fission-altered isotope ratios in groundwater at the US Department of Energy's Hanford Site using inductively coupled plasma – mass spectrometry

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Over 40 years of plutonium production at the Hanford Site, located in south-central Washington State left a legacy of environmental contamination. Currently the most significant radiological contaminants in the groundwater are tritium, technetium-99, iodine-129, and uranium because of their high mobility. Strontium-90 is also a constituent of concern because a significant amount partitions into the aqueous phase in spite of considerable sorption to the aquifer and vadose zone sediments. A number of contaminants such as ruthenium-106 and antimony-125 have decayed to levels where they are no longer detected in groundwater samples. Several other contaminants remain of concern in spite of their lower mobility because significant inventories remain in waste sites and in the vadose zone sediments. These include cesium-137 and plutonium.

Historical measurements of radiological contaminants at Hanford have relied almost exclusively on counting methods. However, these have a number of problems including the difficulty of distinguishing uranium-236 from uranium-235 and plutonium-240 from plutonium-239. Past use of mass-based methods, including thermal ionization mass-spectrometry, and accelerator mass spectrometry, has been limited by high cost and low sample throughput.

Inductively coupled plasma – mass spectrometry (ICP-MS) provides significantly enhanced ability to quantify many radionuclides of concern. In addition, use of ICP-MS has expanded our analytical capabilities to include a number of long-lived and stable fission products. These fission isotopes provide additional information for interpretation of transport processes and aid in distinguishing between contaminant sources.

Results to date show that isotopic abundances for several elements in the subsurface have been impacted by site operations. Altered ratios of stable ruthenium isotopes in groundwater show the presence of fission-ruthenium. Although fission molybdenum isotopes have been identified in vadose zone samples, in the groundwater natural molybdenum dominates. Measured cesium-135:cesium-137 ratios in vadose contamination show that the proportion has been altered from the fission yield due to escape of the xenon-135 precursor prior to its decay to cesium-135.

The success of the ICP-MS measurements is largely due to recent improvements in instrumentation and simple chemical separations to remove mass-interferences. The practicality of using ICP-MS vs. counting methods shifts to shorter-lived and lighter isotopes as the ICP-MS sensitivity increases. The expanded suite of measurable isotopes in many instances provides the capability to distinguish nuclear production contributions from natural background and from fallout. Local sources can be distinguished through consideration of source characteristics, or where the source is uncertain, through phenomenological differences between subpopulations of samples.