CHOOSING THE RIGHT TRACER FOR SPECIAL BIOASSAY ANALYSES

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INTRODUCTION

The In Vitro Analysis Group, of the Analytical Laboratories Department, at the Idaho National Engineering and Environmental Laboratory (INEEL), is responsible for analyzing bioassay samples from various facilities in order to determine possible uptake by INEEL personnel. At the INEEL principle radionuclides of interests are: Plutonium (Pu), Uranium (U), Americium (Am), Strontium (Sr), Cesium (Cs), and Tritium (H-3). Alpha and Gamma spectroscopy are used for detecting the alpha and gamma emitting radionuclides while liquid scintillation and gross Alpha/Beta gas proportional counting is used for detecting beta-emitting radionuclides. A majority of the time, bioassay samples are analyzed using standard approved analytical chemistry methods.

BACKGROUND

For nuclides such as Pu and U, which emit little or no gamma radiation, the preferred method of analysis is alphaspectroscopy. A suitable tracer nuclide of known activity is added for each isotope to monitor the chemical recovery factors through the extraction process. The actinides are gathered on a neodymium/iron hydroxide precipitate or an iron phosphate precipitate. The isotopes of the actinides are separated from each other using ion exchange resin. The purified radionuclides are gathered on a neodymium fluoride precipitate, filtered, and counted in an alphaspectrometer.

The In Vitro group has 24 EG&G OERTEC ULTRA series detectors with a thin, ion implanted contact, immediately under the silicon surface detectors. The measurement of alpha particle resolution is performed in a vacuum with a uniform, ultra thin source located at a source-to-detector distance at 1-1/2 times the detector diameter. A computer based alpha pulse height system is used for the analysis of samples containing alpha - particle - emitting radionuclides. The alpha detection and signal shaping/handling is accomplished using alphaspectrometry units (ASU), analog-to-digital converters (ADC), and multiplexers. The software includes features that support the concurrent operation of multiple detectors, photopeak location, background correction, automatic reference peak gain adjustment, and report generation.

Recently the In Vitro group was asked to analyze bioassay samples of the analysts working with the nuclear fuel containing 3714 microcurie of Uranium-233, 98 microcurie of Uranium-232, 9 microcurie of Thorium-232 and 3.8 microcurie of Th-229. At the

INEEL Uranium-232 is used as a tracer for routine bioassay analyses. For a special project, the In Vitro group deals with special source terms on a case-by-case basis and job specific bioassays are developed. Since this was a non-routine project, every aspect of it was reexamined and several options were considered to determine which tracer to use for the analysis of these bioassays.

OPTIONS

Option (1): Using U-232 as a tracer.

This option triggers the following factors to be considered.

1) The fact that subject waste has 2.6% U-232, may cause higher chemical recovery than added tracer amount. So, for the final report to quantify U-233, adjustments will have to be made for the counts contributed by U-232 in the waste.
2) In bioassay analyses, reported results are usually in very small amounts. While dealing with such a minute quantity, subtracting counts due to tracer becomes a technique rather than a scientific process and significant errors are more likely to occur. One has to be really careful in making those calculations for reporting possible intake of radioactivity.

Option (2): Using U-238 as a tracer.

It is almost impossible to get pure U-238 tracer. U-238 tracer obtained by the In Vitro Group was prepared from NBS SRM U-0002 @ 17.17479 d/s/g. This tracer contained 16.66105 d/s/g of U-238, 0.49516 d/s/g of U-234 and 0.01858d/s/g of U-235 and < 0.00001 wt.% U-236 (activity not accounted for in calculations). Using U-238 as a tracer generates two factors to be considered.

1) The fact that the tracer has @3% U-234 creates a final result calculations error because of only a 50 keV difference in U-233 (@4824 keV and U-234 @4775 keV resolutions. In the final analysis report U-233 is quantified along with U-234. So adjustments will have to be made for the 3% of U-234 counts contributed due to tracer.

2) The sensitivity of In Vitro sampling as a Uranium bioassay tool is limited by the presence of environmental levels of Uranium, which is subject to some uncertainty in interpretation. In ICRP 30 (1979) the average daily ingestion intake of natural uranium in food and water is estimated to be 1.9 ug. Based on studies performed at one facility at INEEL, the concentration of Uranium found in potable water is approximately 3 ug per liter of water. This background quantity of Uranium is found in subsurface water that is consumed as drinking water by workers. All individuals are exposed to natural Uranium found in foods and in the soil/rocks found in their representative communities. These exposures constitute background exposures. An analysis of aquifer water was performed by the subject facility for Uranium isotopes which identified the following concentrations:

U-234 1.94E-6 uCi/L or 3.11E-4ug/L
U-235 7.00E-8 uCi/L or 3.24E-2 ug/L
U-238 9.54E-7 uCi/L or 2.84 ug/L

This gives a U-234/U238 activity ratio of 2.0.

Therefore, the contribution of "background" Uranium intake from a non-nuclear environment is very common in bioassay samples and should be subtracted from the total Uranium isotopic intake in order to establish an occupation intake.

The INEEL established a technique for which Uranium bioassay results are evaluated as something other than the detection of non-occupational Uranium uptakes. The basis of U-234/U238 ratio of 2.0 is taken into consideration before declaring any positive uptake. In order to rule out the possibility of natural/background Uranium in these non-routine bioassay samples, in case of a suspect uptake, it is necessary to know amount of U-234 and U-238 present in the sample. Using U-238 as a tracer may add errors in the final result calculations.

CONCLUSION

After considering the above options carefully, it was determined by the technical leader of the In Vitro group to use U-232 as a tracer. Most bioassay samples do not show any positive identification of radioactivity. In case of an uptake, it is easier to subtract U-232 counts contributed by the subject waste from the total counts vs. using U-238 as a tracer.

REFERENCES

1) DOE Standard Internal Dosimetry, DOE-STD-1121-98
2) INEEL Radiation Protection Program, PLN-260, Rev.2, July 6, 2000
3) Engineering Design File No. INEEL-2001-025