The U.S. Transuranium and Uranium Registries: Forty Years' Experience and New Directions in the Analysis of Actinides in Human Tissues

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September 2008 was the 40th anniversary of the U.S. Atomic Energy Commission's vision in establishing the National Plutonium Registry. Its successors, the U.S. Transuranium and Uranium Registries (USTUR), continue to follow individuals with documented accidental exposures to actinide elements, to study their uptake, translocation and retention (biokinetics), and tissue dosimetry. To date, 330 past-worker volunteers have donated their tissues for scientific research, including 36 whole body donors. This talk outlines the history, mission and research goals of the Registries, and the current status of the USTUR's Radiochemistry Program. We present an overview of the analytical methods for plutonium (Pu), americium (Am) and uranium (U) isotopic determination in human tissues currently applied; including inductively coupled mass spectrometry (ICP-MS) based techniques. Recent developments in rapid radiochemical separation of the actinides from soft tissue and bone samples using vacuum-assisted extraction chromatography are also outlined. The results of inter-comparing $^{239+240}$Pu, ^{241}Am and ^{234,235,238}U determinations by ICP-MS, α-spectrometry (AS) and kinetic phosphorescence analysis (KPA, for total U) are discussed. ICP-MS is a major advance over AS and KPA in enabling the measurement of the ^{240}Pu/^{239}Pu atom ratio, the short-lived β-emitter ^{241}Pu, and the anthropogenic ^{236}U. For the first time ^{241}Am and ^{241}Pu were measured in human tissues using ICP-MS. The measured ^{240}Pu/^{239}Pu, ^{235}U/^{238}U and ^{236}U/^{238}U atom ratios clearly identify the origins of these actinides in human tissues. A new avenue of research with samples of USTUR human tissue is the application of laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) to elemental bio-imaging (EBI) of the actinides (of both anthropogenic and natural origin). We present elemental distribution maps for ^{235,238}U and ^{232}Th in lymph nodes measured for both non-exposed (chronic intake) and occupationally exposed (inhalation) subjects. The implications of these findings for dose assessment are discussed.

USTUR-0276A-09 (Presentation)