Beyond α-Spectrometry for Actinide Determination in Human Tissues?

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We present an overview of the analytical methods for Pu, Am and U isotopic determination in human tissue samples currently used at U.S. Transuranium and Uranium Registries (USTUR), including exploration of inductively coupled mass spectrometry (ICP-MS) techniques. $^{239+240}$Pu, ^{241}Am and ^{234,235,238}U determinations by both Sector Field (SF) ICP-MS and α-spectrometry (AS) have been intercompared. For $^{239+240}$Pu, activity values derived from ICP-MS measurements were 3% higher than the AS results (for 12 samples). The results of investigating this bias in the ICP-MS measurements using ^{240}Pu/^{242}Pu standards will be presented. With SF-ICP-MS, ^{241}Pu (β-emitter, T1/2 =14.1 y) was measured in 4 samples (with "high" Pu content). However ^{238}Pu was not measurable due to isobaric interference from background ^{238}U. For ^{241}Am, the ICP-MS values were 8% lower than those from AS based on 4 samples analyses. More samples are being analyzed to investigate this ^{241}Am measurement bias. Uranium measurements highlighted significant problems in using AS for low-activity (natural) isotopic determinations. However, for higher activity samples (occupationally-exposed workers), ICP-MS and AS results were in a good agreement. The SF-ICP-MS detection limit for ^{238}U is an order of magnitude lower than that by AS, and SF-ICP-MS accurately measures ^{236}U from anthropogenic uranium. For all isotopes, ICP-MS has the great advantage of rapid analysis (10 min per sample *vs*. 41 hr for AS).

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