Determination of U, Pu, and Am in Biological Samples by SF-ICPMS for Biokinetic Studies of Actinides.

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Plutonium isotopes (^{239}Pu, ^{240}Pu, ^{241}Pu), uranium isotopes (^{234}U, ^{235}U, ^{236}U, and ^{238}U), and ^{241}Am were measured in digested bone and soft tissue samples with SF-ICPMS; these results were compared to results previously obtained by alpha spectrometry (AS). The samples were obtained from USTUR registrants (Cases 0269, 0425, 0720 and 1028) with known occupational exposure histories. Samples investigated in this study were obtained as un-spiked 6-8 M HCl solution aliquots from USTUR's National Human Radiobiology Tissue Repository (NHRTR). USTUR dissolution procedures consist of dry-ashing, followed by complete decomposition of the residue with mineral acids and dissolution of the residue in aqueous HCl. Prior to ICPMS measurements, chemical separations were conducted using extraction chromatography. Sample extracts were 5-15 grams and typically represented only 1-3% of the total sample digest. ^{242}Pu and ^{243}Am were used as spike isotopes for the determination of Pu isotopes and ^{241}Am, respectively. Pu was isolated using microcolumns of TEVA resin (EIChrom). Am was isolated using a three column sequence: TEVA-UTEVA-DGA. The TEVA and UTEVA were first used to remove Pu and U, respectively; thereafter, Am is retained and purified on a microcolumn comprised of DGA resin (EIChrom). Uranium activities were measured using two aliquots: an unspiked aliquot was used to prepare a U fraction with UTEVA resin; this unspiked aliquot was used to measure the ^{234}U-^{235}U-^{236}U-^{238}U isotopic composition. A second U aliquot was spiked with ^{236}U and was used to measure U activities by comparison to standards prepared to known ^{236}U/^{xxx}U ratios. Since some of the samples contained ^{236}U, two U runs per sample were required to subtract the indigenous ^{236}U content. In future work, ^{233}U of high isotopic purity will be used as a spike to measure all U isotope activities and ratios in a single sample aliquot.

The U, Pu, and Am results were compared to AS, which has long been used at USTUR for Pu and Am measurement after chemical separation of the analytes. Six bone and eight soft tissue samples were used and analyzed in the study. ^{239}Pu and ^{240}Pu activities, along with the ^{240}Pu:^{239}Pu atom ratio, were measured by ICPMS in 13 samples; $^{239+240}$Pu along with ^{238}Pu were measured in all samples by AS. The ^{240}Pu:^{239}Pu atom ratios were uniform among individual cases (0.0628 ± 0.0022, 1 SD), and are consistent with 1950's-1960's weapons grade material. In one of the samples the $^{239+240}$Pu activity was below the SF-ICPMS detection limit. ^{238}Pu measurements were not attempted by ICPMS due to isobaric interference from natural ^{238}U. However, beta-emitting ^{241}Pu was measured by SF-ICPMS in five samples of high $^{239+240}$Pu content. Positive correlation (r^2 = 0.9997) was observed between $^{239+240}$Pu values measured by SF-ICPMS and AS. With SF-ICPMS, ^{241}Am was measured in four samples out of five with high $^{239+240}$Pu. For a small number of data points (n =4) significant correlation (r^2=9985) was observed between ^{241}Am activities reported by SF-ICPMS and α-spectrometry. The comparison of SF-ICPMS with AS confirmed the suitability of the ICPMS method when applied to the analyses of $^{239+240}$Pu and ^{241}Am in the USTUR samples. U measurements confirm the presence of grossly elevated U activities in soft tissues and bones of occupationally exposed individuals; some samples exhibited non-natural ^{238}U:^{235}U atom ratios and the presence of ^{236}U, clearly indicating exposure to anthropogenic U.

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