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UCRL-ABS-213684

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July 18, 2005

51st Annual Radiobioassay & Radiochemical Measurements Conference (RRMC)
Stateline, NV, United States
October 24, 2005 through October 28, 2005

AMS Analysis of ^{239}Pu in Archived Occupational Samples

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Under an IRB-approved human subjects protocol, accelerator mass spectrometry (AMS) was used to recover previously inaccessible information concerning short- and long-term excretion patterns of urinary Pu-239 for LLNL Pu workers who uniquely were both monitored periodically at LLNL via urine sampling over a period of decades and also had corresponding records archived at LLNL, in some cases for ≥20 years. Because the AMS methods used were substantially (50- to 300-fold) more sensitive than alpha spectrometry, excretion biokinetics were recovered from the archived LLNL alpha spectrometry discs to obtain measures pertaining to the biokinetics of human Pu-239 excretion with unprecedented sensitivity. AMS analysis was performed on residue from alpha spectrometry discs used historically to monitor Pu-239 excretion from a total of 11 current or former LLNL employees: 7 suspected of having ≥1 previous low-level occupational Pu-239 intake, and 4 thought to have had no such intake. AMS data obtained clearly confirmed occupational intakes for all workers suspected of previous exposure. AMS data for one worker time series provide evidence of an historical Pu-injection wound that most likely occurred 15-20 y prior to its recent discovery by body counting methods (Figure 1). AMS data obtained for this and other workers indicate that even extremely low-level occupational exposures to Pu-239 typically yield body burdens that remain readily detectable in urine analyzed by AMS, effectively marking any such worker for life—an observation that may have important occupational health and national/homeland security applications. [This work was performed under auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory (LLNL) under Contract W-7405-Eng-48.]

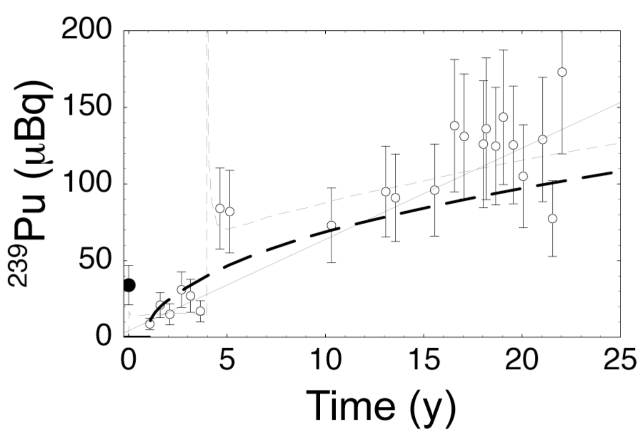

Figure 1. Urinary ^{239}Pu (points ±1 SE) detected by retrospective AMS analysis of archived sample plates prepared from urine samples obtained from a male worker by retrospective AMS analysis of archived bioassay plates, vs. relative time t, shown fit to three different SE^{-2}-weighted regression models. The linear fit shown (solid line) is 4.3 (±3.1) μBq + t 5.96 (±0.583) μBq y^{-1} (χ^2 = 18.7, df = 21, p = 0.36; this fit excludes outlying solid point, $F_{1,21}$ = 5.68, p = 0.027). A fit of the biokinetic model $A(t) = a\, U_{cIV}(t-t_1)$, where $U_{cIV}(t) = \int_0^t U_{IV}(s)\,dt$ and U_{IV} is the current ICRP biokinetic model for IV injection of Pu, is shown (bold long dashed curve) with estimates $t_1 = 1.06$ (±0.12) y and $a = 489$ (±37.4) (χ^2 = 32.8, df = 22, p = 0.065). The fit to a similar model assuming Pu injection at times $t_0 = -10/365$ y and $t_1 = 4$ y is also shown (short dashed curve; χ^2 = 14.4, df = 21, p = 0.85). All three models are statistically consistent with the AMS data.