AMS Analysis of $^{239}$Pu in archived occupational samples

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Unique sample & records archive available for LLNL Pu-workers

- Since the inception at LLNL of bioassays for Pu, >500 workers have been monitored
- Workers with positive a spectrometry results for tested urine had follow-up health physics review and subsequent periodic monitoring
- Original electroplated a-spec. discs have been archived at LLNL for the past 20 years, and are linked to corresponding worker exposure records
  - Similarly archived records/materials not available at any other U.S. Pu facility
- Approximately 10 LLNL workers are known or suspected to have had Pu intake, based on a-spec. monitoring results
Radiochemistry for AMS analysis of $^{239}\text{Pu}$ in urine adapted for $\alpha$-spec plates ($<10^{13} \ 238\text{U}$ atoms/sample)

Sample Digestion → Column Separation → AMS Target Preparation → AMS Ion Source

Fe(OH)$_3$ Co-precipitation

Disposable Quartz Crucibles → Oxide Formation

$\alpha$-Spec. discs
AMS detection of $^{239}$Pu at LLNL
LLNL now detects $^{239}$Pu above backgrounds equivalent to $\sim 10^5$ $^{239}$Pu atoms ($\sim 0.1$ $\mu$Bq)

$^{239}$Pu/$^{242}$Pu $\times$ 100%

$\mathcal{P} = 0.0005$, df=1 ($p = 0.983$)

Estimated limit of detection

Estimated background

Equiv. $^{239}$Pu atoms (millions)
Above-background original and recounted \( \alpha \)-spec measures found to be similar
Above-background AMS and \( \alpha \)-spec measures found to be similar.
AMS measures of $^{239}\text{Pu}$ from archived §-spec disks for 4 workers with no record of previous potential occupational exposure to Pu

Mean ($\pm$ 1 SE) = 0.89 ($\pm$ 2.65) $\mu$Bq
Recent $\alpha$-spec counts for worker #1 led to confirmation of aux. lymph node Pu, after which these retrospective AMS data were obtained.

- $\chi^2 = 14.4$
- df = 21
- $p = 0.85$

- $\chi^2 = 32.8$
- df = 22
- $p = 0.065$

*Outlier excluded (bold)
An α-spec count and records for worker #2 are consistent with 1 or 2 respiratory Pu exposures.

\[ ICRP \text{ 2-inhalation}\]

\[ \chi^2 = 29.7 \]

\[ df = 16 \]

\[ p = 0.020 \]

*Outliers excluded (bold)
Records for worker #3 are consistent with possible respiratory exposure to Pu

\[ ICRP \text{ 3-inhalation}^* \]

\[ \chi^2 = 44.0 \]
\[ df = 25 \]
\[ p = 0.011 \]

*Outlier excluded (bold)