Age-related trend in elevated plutonium-239 measured by AMS in urine samples collected in 1998-2003 from Enewetak residents and Rongelap resettlement workers

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Under an IRB-approved human subjects protocol, a total of 377 urine samples were collected from 167 (male + female) residents of Enewetak Atoll during 2001-2003, and from 115 (male) Rongelap Atoll resettlement workers, together with field blanks prepared by identical methods substituting double-distilled H$_2$O for collected urine. Extensive precautions were taken to avoid sample contamination, including physical isolation in an indoor facility during a uniform 24-h sample collection period. Measures of $^{239}$Pu subsequently made at the LLNL Center for Accelerator Mass Spectrometry (AMS) included estimates of combined measurement error (ME) due to chemical recovery and AMS isotope sampling. Measures from each multiply sampled individual were pooled, and participants on each atoll were grouped by age at sampling time into three ranges (<35, 35-44, ≥45 y, rounded down to nearest y), corresponding to low, medium and high levels of relative historical exposure to fallout due to atmospheric nuclear testing estimated using published UNSCEAR data. The $^{239}$Pu measures obtained from older age groups were significantly elevated compared to pooled blanks, as assessed by 2-tail Monte Carlo compound Mann-Whitney (CMW) tests that account for all ME (Figure 1). Across age groups (with blanks taken as age 0), the group-specific fractions of measures >0.35 mBq were found to be positively associated with age, ignoring ME ($p < 0.001$, by atoll-specific chi-square tests for linear trend). Accounting for all ME, the positive age-related trend for Enewetak is statistically significant (Figure 2). Observed trends in relation to age were stronger that those in relation to relative exposure to cumulative fallout. [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, with support from the LLNL Marshall Islands Program.]

![Figure 1](image1.png)

**Figure 1.** Empirical probability mass functions (cmfs) for $^{239}$Pu in pooled field blanks vs. urine samples from all 285 study participants (each cmf is weighted by corresponding inverse-square ME sets). $^{239}$Pu is significantly elevated among all participants ≥35 y ($p < 0.035$, by CMW tests).

![Figure 2](image2.png)

**Figure 2.** Urinary $^{239}$Pu is significantly positively associated with age for all Enewetak ($n = 170$) participants, as assessed by 0-intercept weighted linear regression of all data pooled by age group, vs. corresponding average age, using ME$^{-2}$ as weights (solid horizontal line = 0 mBq/24-h, short-dashed lines = 95% confidence limits on slope). Error bars denote ±1 SEM; 95% confidence limits on the fitted intercept include 0.