# Age-related trend in elevated $^{239}Pu$ measured by AMS in urine samples collected in 1998-2003 from Enewetak residents and Rongelap resettlement workers

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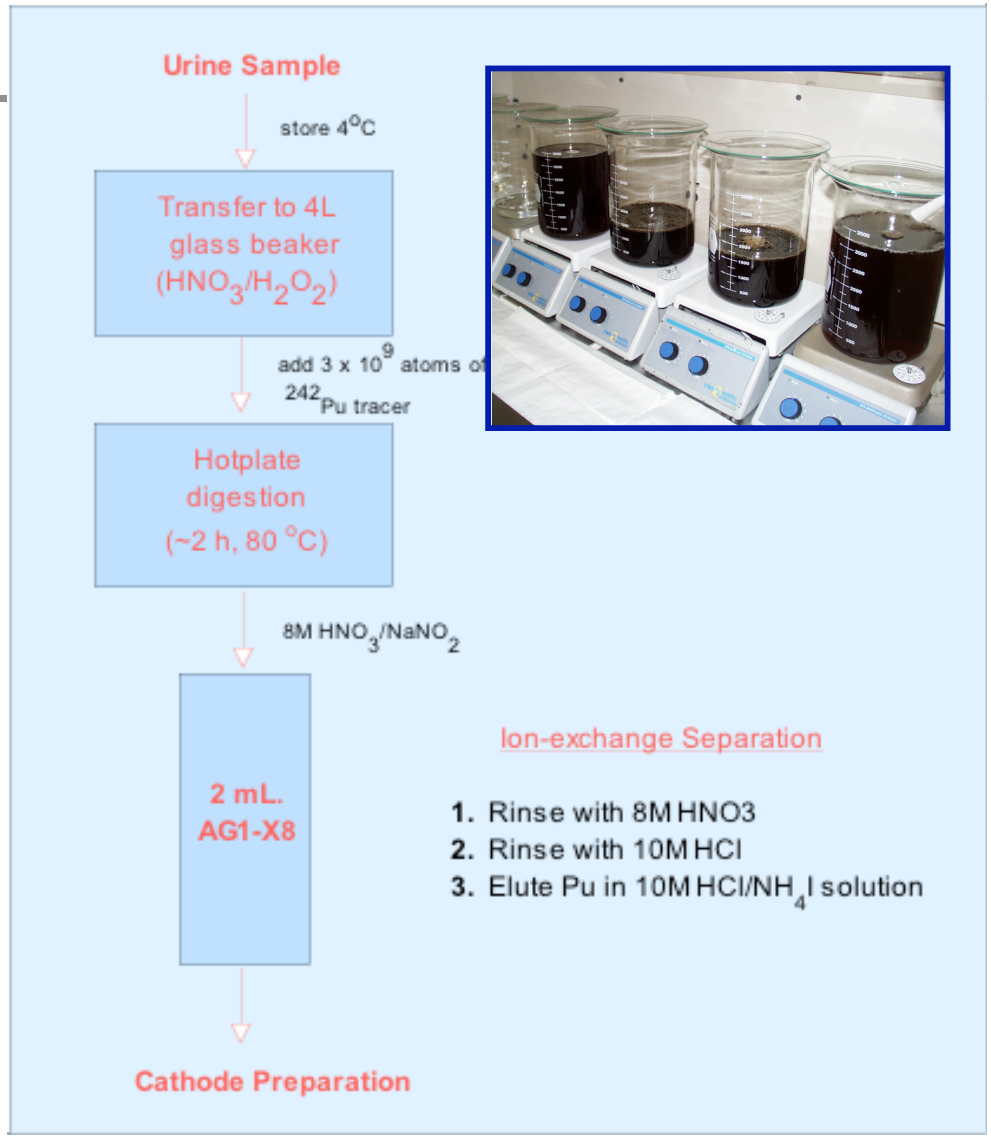
# Urine collection at LLNL's Enewetak Radiological Laboratory

**Constructed during 2000, this facility includes a whole body counting chair and clean living space for collecting 24-h void bioassay samples**

# Radiochemistry for $^{239}Pu$ analysis by AMS

**STANDARD METHODS**

1. Hotplate Digestion ($HNO_3/H_2O_2$)
2. Ca Phosphate drop
3. Wet Ash ($HNO_3/H_2O_2$)



# Cathode prep for $^{239}Pu$ analysis by AMS

**Pu fraction**
(50 mL 'C' tube)

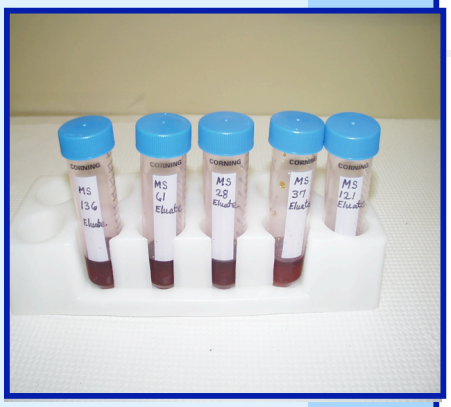
**Adjust pH with ammonia solution**

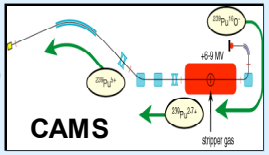
Precipitate Pu on $Fe(OH)_3$

**Centrifuge**

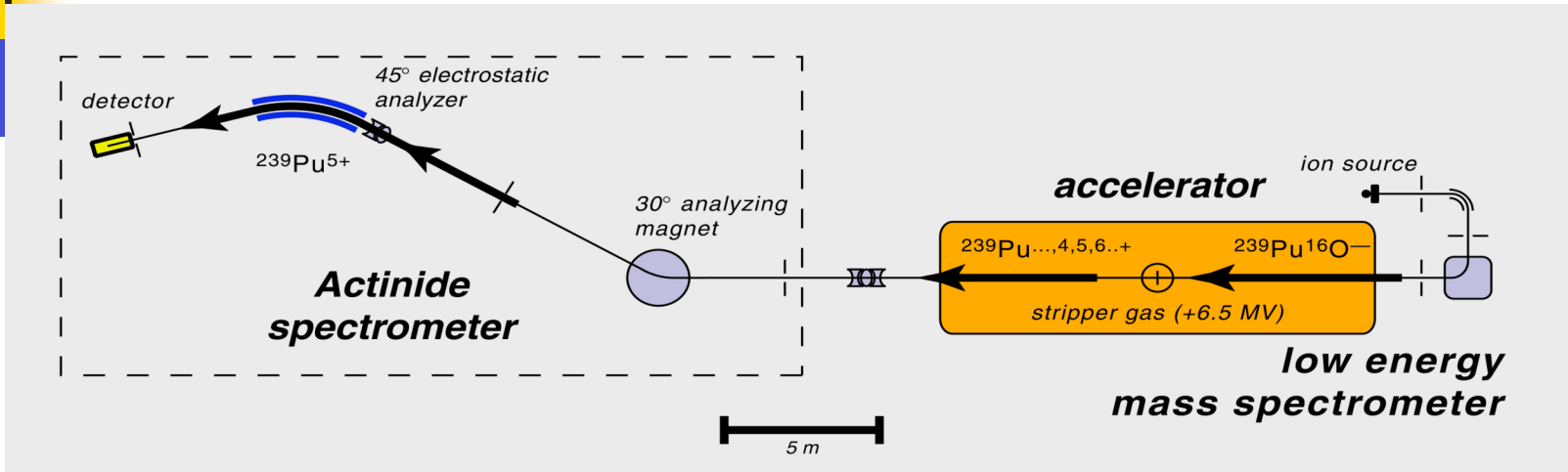
Dissolve precipitate in Nb oxide containing $HNO_3$ / $H_2O_2$

**Transfer to Si vial, dry under heat lamp, & bake at 800 °C**

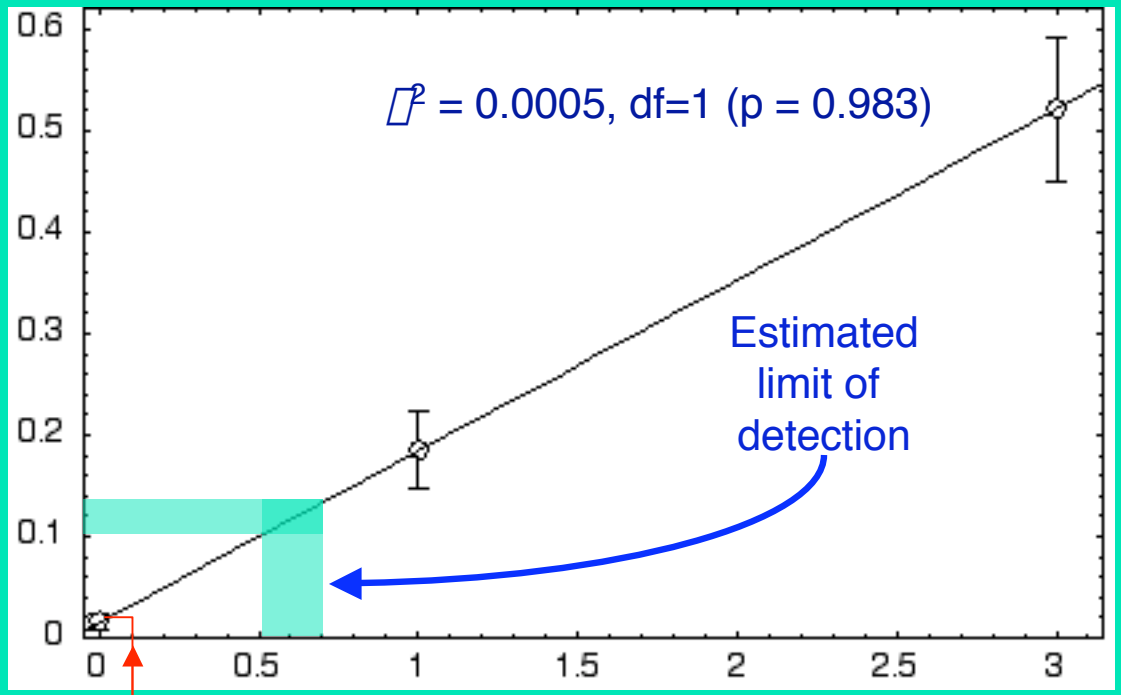




# AMS detection of $^{239}Pu$ at LLNL



# LLNL now detects $^{239}Pu$ above backgrounds equivalent to ~$10^5$ $^{239}Pu$ atoms (~0.1 μBq)



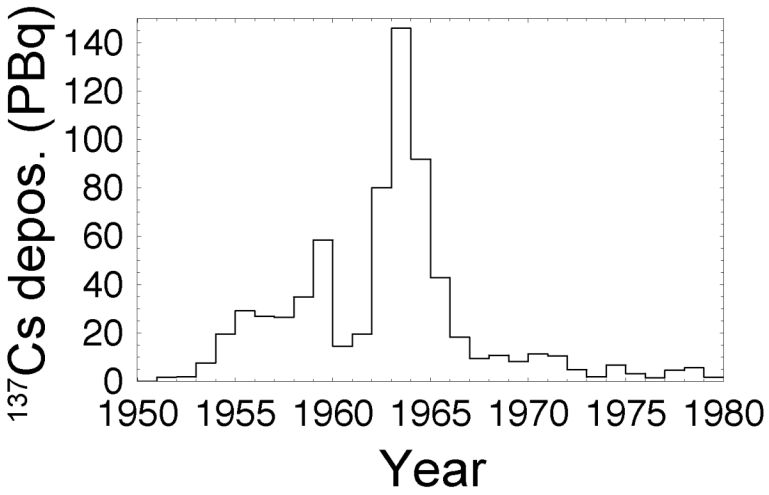
# Summary of plutonium bioassay samples prepared on Enewetak and Rongelap Atolls

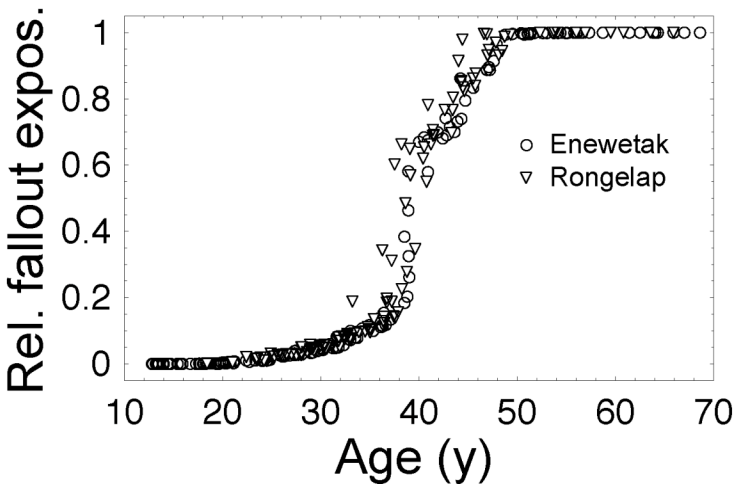
| Island | Source[a] | # Samples | # Individuals | Age (y)[a] | | |
|---|---|---|---|---|---|---|
| | | | | Median | Minimum | Maximum |
| Enewetak | FB | 26 | - | - | - | - |
| | F | 60 | 52 | 30 | 13 | 67 |
| | M | 150 | 118 | 32 | 13 | 88 |
| Rongelap | FB | 19 | | - | - | - |
| | F | - | - | - | - | - |
| | M | 167 | 115 | 36 | 18 | 66 |

[a] FB = field blanks (samples of deionized $H_2O$ rather than urine, but otherwise prepared and treated identically to urine samples), Male = male participant, F = female participant, "–" = not applicable. Ages rounded to nearest year.

# Estimated exposure to atmospheric-testing fallout

**Annual Northern hemisphere $^{137}$Cs deposition from atmospheric nuclear testing**

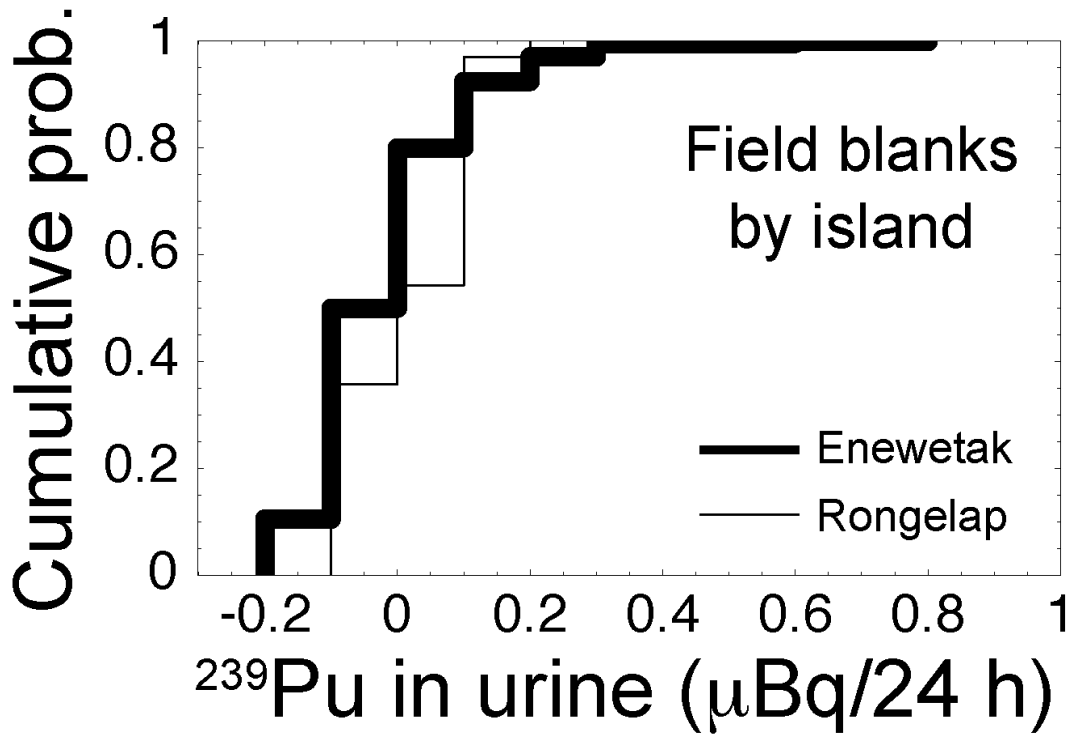



Source: UNSCEAR 2000

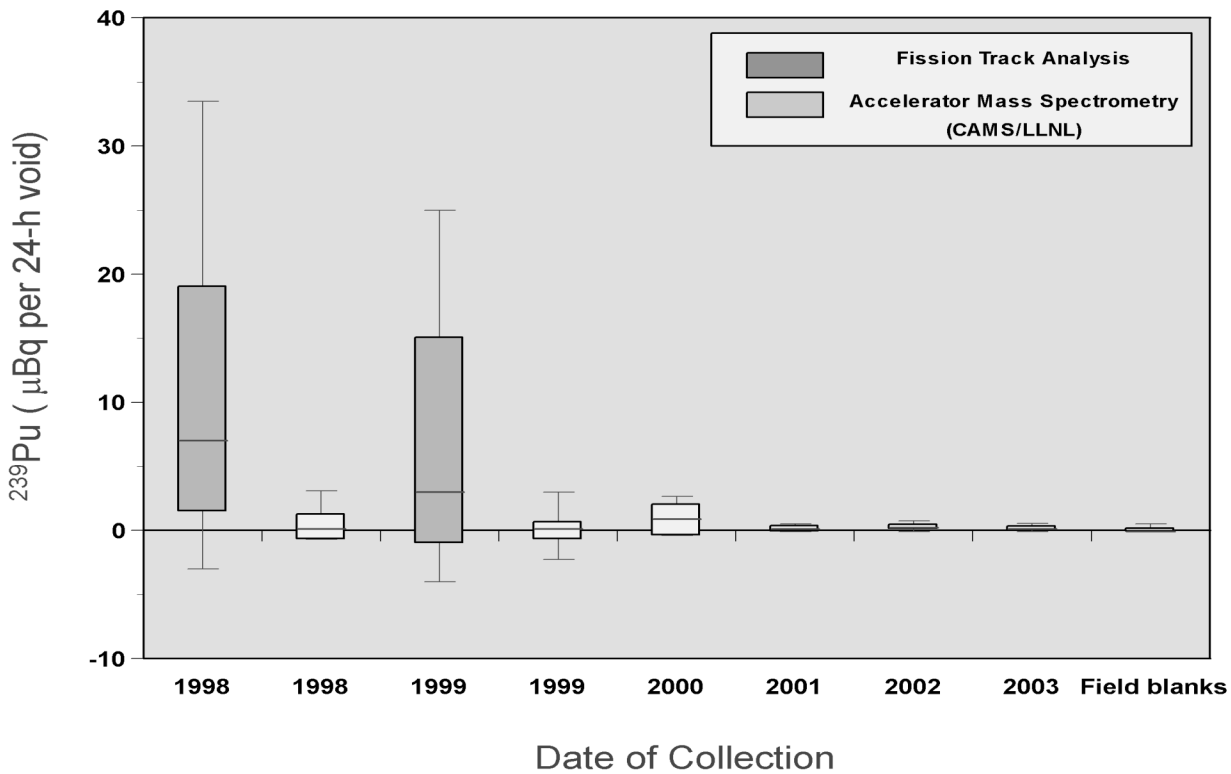
Assumes $^{239}$Pu:$^{137}$Cs ~ 1:1

# $^{239}$Pu measured in field blanks




Field blanks prepared on Enewetak ($n$ = 26) vs. on Rongelap ($n$ = 19). Each cmf is weighted by inverse squares of corresponding measurement errors. The data sets do not differ significantly by 2-tail compound-Monte-Carlo Mann-Whitney test (0.81).

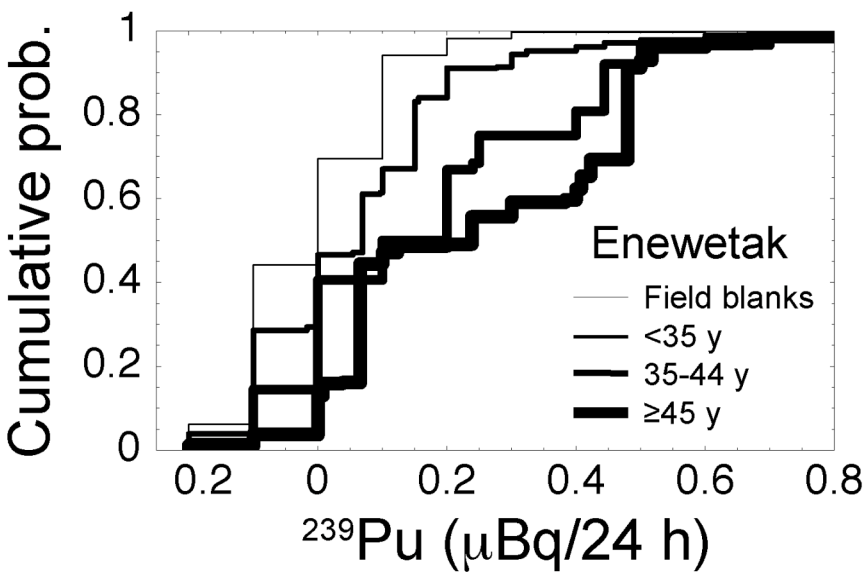
# AMS measures of MI-urinary $^{239}Pu$ are lower than previous measures made by FTA



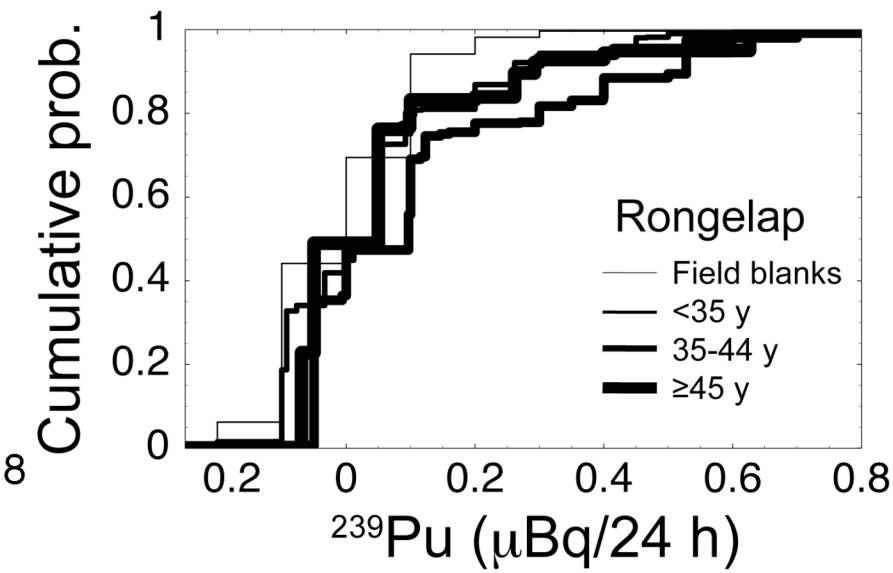

Explanation: The lowest, second lowest, middle, second highest and highest box points represent the 10th, 25th, median, 75th and 90th percentiles, repectively.

# $^{239}$Pu in urine by age, vs. field blanks

170 females + males on Enewetak




115 males on Enewetak




Each cmf is weighted by inverse squares of corresponding measurement errors. Urinary $^{239}$Pu measures are significantly elevated among participants ≥45 y old on each island ($p \leq 0.03$), and among participants 35 to 44 y old on Rongelap ($p = 0.0064$), by 2-tail compound-Monte-Carlo Mann-Whitney test (0.81).

# Contingency analysis results are also consistent with age-related trends

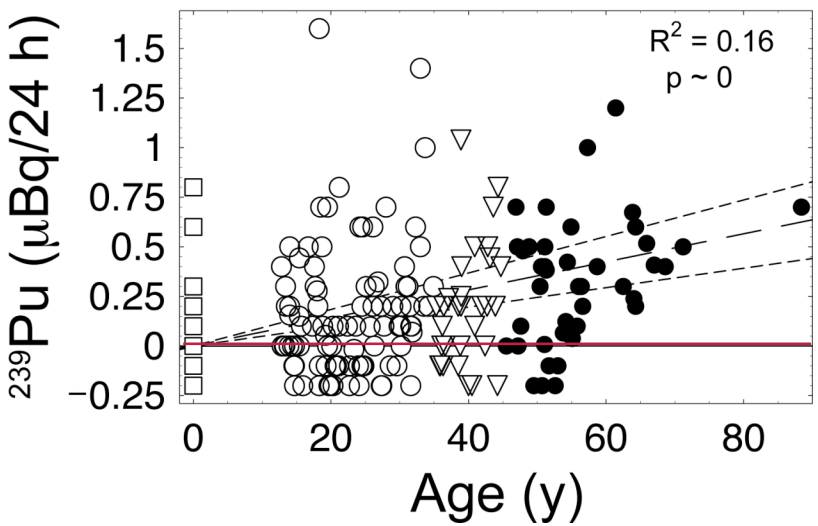
| Island | Comparison[a] | $N$ | # >0.35 μBq | p-values[b] Association | Homogeneity | Trend |
|---|---|---|---|---|---|---|
| Enewetak | FB | 45 | 2 | – | $3.5\times10^{-6}$ | $5.8\times10^{-7}$ |
| | <35 y | 97 | 20 | 0.018 | | |
| | 35-44 y | 30 | 8 | 0.016 | [0.0015] | [0.00027] |
| | ≥45 y | 43 | 22 | $9.2\times10^{-7}$ | | |
| Rongelap | FB | 45 | 2 | – | 0.0011 | 0.00051 |
| | <35 y | 51 | 11 | 0.027 | | |
| | 35-44 y | 42 | 15 | 0.00043 | [0.28] | [0.12] |
| | ≥45 y | 22 | 7 | 0.0086 | | |

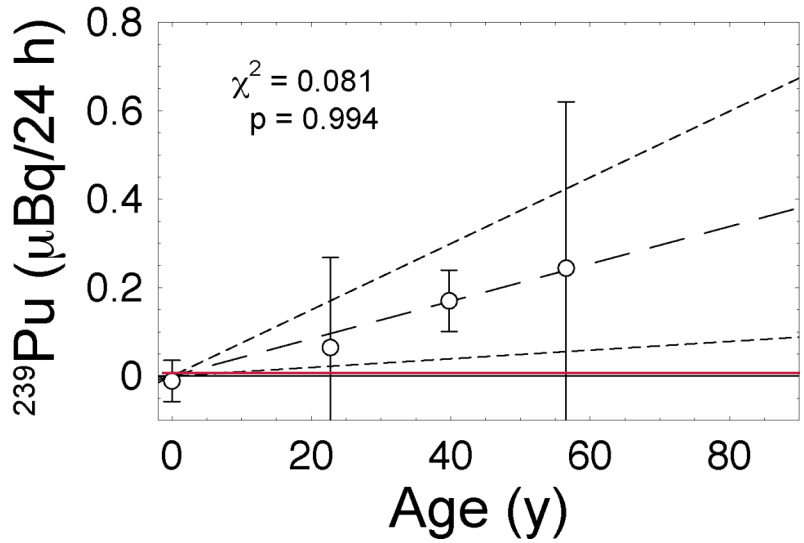
[a] FB = Combined field blanks, $N$ = total number of measures (for FB, or for all participants on each island for each age group listed). Measurement errors were ignored in these analyses. Multiple samples from any single individual were pooled as explained in Methods, so each sample value here corresponds to a single participant.

[b] Association with age (relative to FB data) was assessed by 2-tail Fisher exact test; homogeneity and linear trend were assessed using extended Fisher exact tests for independence, and by Bartholemew's test for trend, respectively, in the implied $2\times k$-fold contingency table with $k$=4 ordered as listed by age group; p-values in brackets correspond to $k$=3 excluding the FB data.

# Urinary $^{239}$Pu is positively associated with age for all (male + female) urine donors on Enewetak

***n* = 210, ——— = 0 μBq/24-h**




□ = field blank, ○ = <35 y, ▯ = 35-44 y, ● = ≥45 y. The weighted 0-intercept linear regression (ZLR) is shown with 95% CLs on slope for all data excluding 2 outliers >5 μBq/24-h (1 each among the older age groups). The 95% CLs on fitted intercept include 0.

ZLR and 95% CLs on slope are shown for all data pooled by age group, vs. corresponding average age, with error bars denoting ±1 SEM. The 95% CLs on the fitted intercept include 0. No significant trend was observed for all (male) donors on Rongelap (p = 0.14).