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# **The Usefulness of Accelerator Mass Spectrometry in Retrospective Dosimetry Studies**

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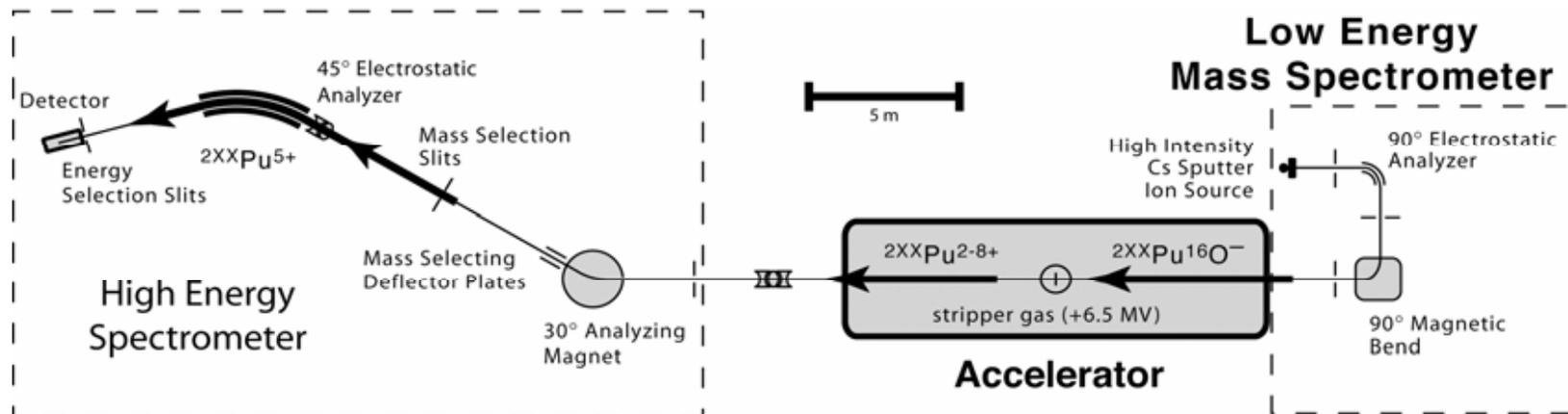
# Objectives

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- **Evaluate the usefulness of Accelerator Mass spectrometry in evaluating past exposures to Pu-239.**
- **Establish investigative techniques for retrospective determination of internal dose from intakes of Pu-239.**

# LLNL Accelerator Mass spectrometry



The LLNL AMS has a heavy-element beam line incorporating an isotope switching system unique to AMS that allows simultaneous measurement of several isotopes during each sample analysis, as well as simple selection of particular isotopes to be measured



# Detection Levels and Dose Implications

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- **Detection level using AMS is less than 1 uBq**
- **Current detection level by conventional alpha spectrometry is 117 uBq**
- **Dose represented by urine sample with 1 uBq, 1 year post inhalation, Type S Pu-239 is 50 uSv**
- **Dose represented by urine sample with 1 uBq, 20 years post inhalation, Type S Pu-239 is 90 uSv**

# Sample Selection

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- **Historical samples from 2 workers with known Pu-239 exposure measurable by alpha spectrometry**
- **Historical samples from 2 workers with suspected Pu-239 exposure based on work place indicators, known incidents, positive in vivo measurement results and/or sporadic alpha spectrometry results**
- **Historical samples from 4 workers with no known or suspected Pu exposure**



# Historical Samples

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- **1 cm diameter Pu-electroplated stainless steel disks**
- **Uniquely identified by etched sample number on back of disk (older disks had indelible marker identifications)**
- **Archived in jewel box containers**
- **Over 20 years of archives**
- **Recounted before analysis by AMS**



# Original Urine Sample Plutonium Chemistry - Quick Summary

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- Pu-242 tracer solution added to raw sample (most samples)
- Wet ash with HCl & nitric acid
- Co-precipitated, dissolved back into HCl
- Separated via anion column exchange
- Eluted from column and electroplated

**Note: Some samples were processed for both Am-241 and Pu-239 via a sequential separation procedure. Pu-242 and Am-243 tracer solutions were added to these samples prior to wet ashing.**

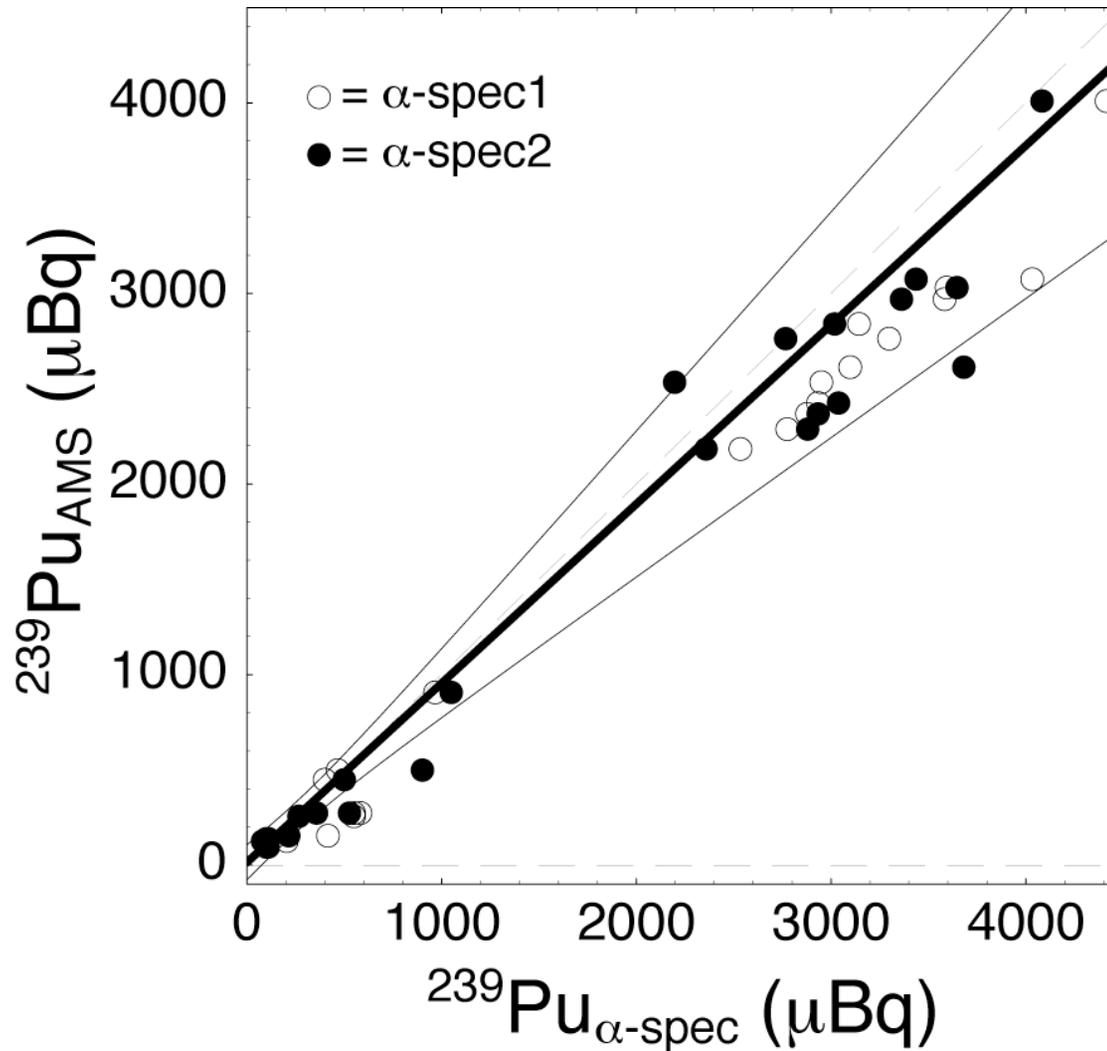
# AMS Sample Preparation

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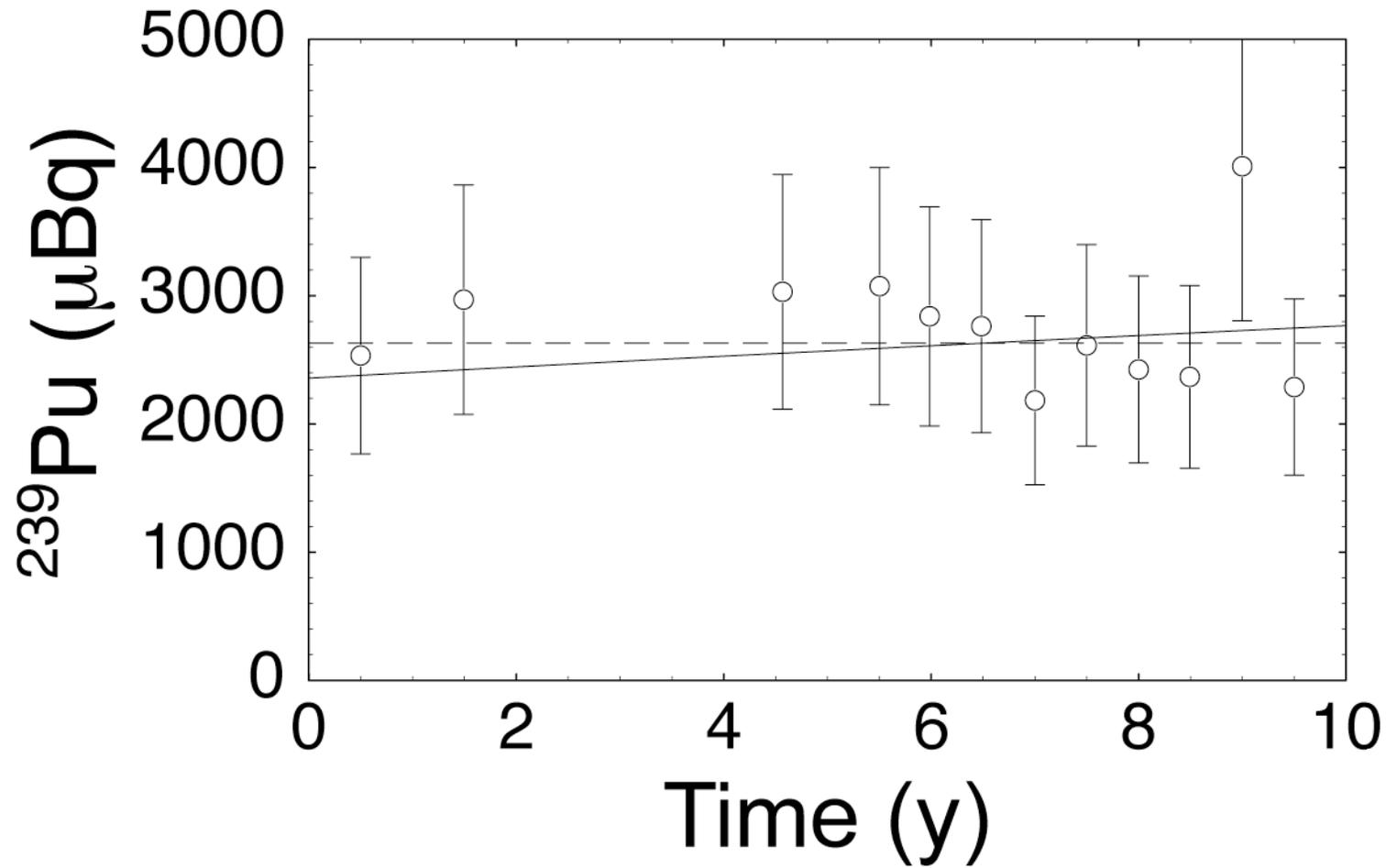
- **Sample plates counted prior to and after AMS preparation**
- **Pu (including the Pu-242 tracer solutions) etched from stainless steel disk using concentrated HF pickling followed by 8M nitric rinse**
- **Pu co-precipitated and separated on anion exchange column**
- **Eluted from column and precipitated with Fe-Nb and dried**
- **Powder complex packed into AMS cathode**

# AMS vs. Alpha spectrometry

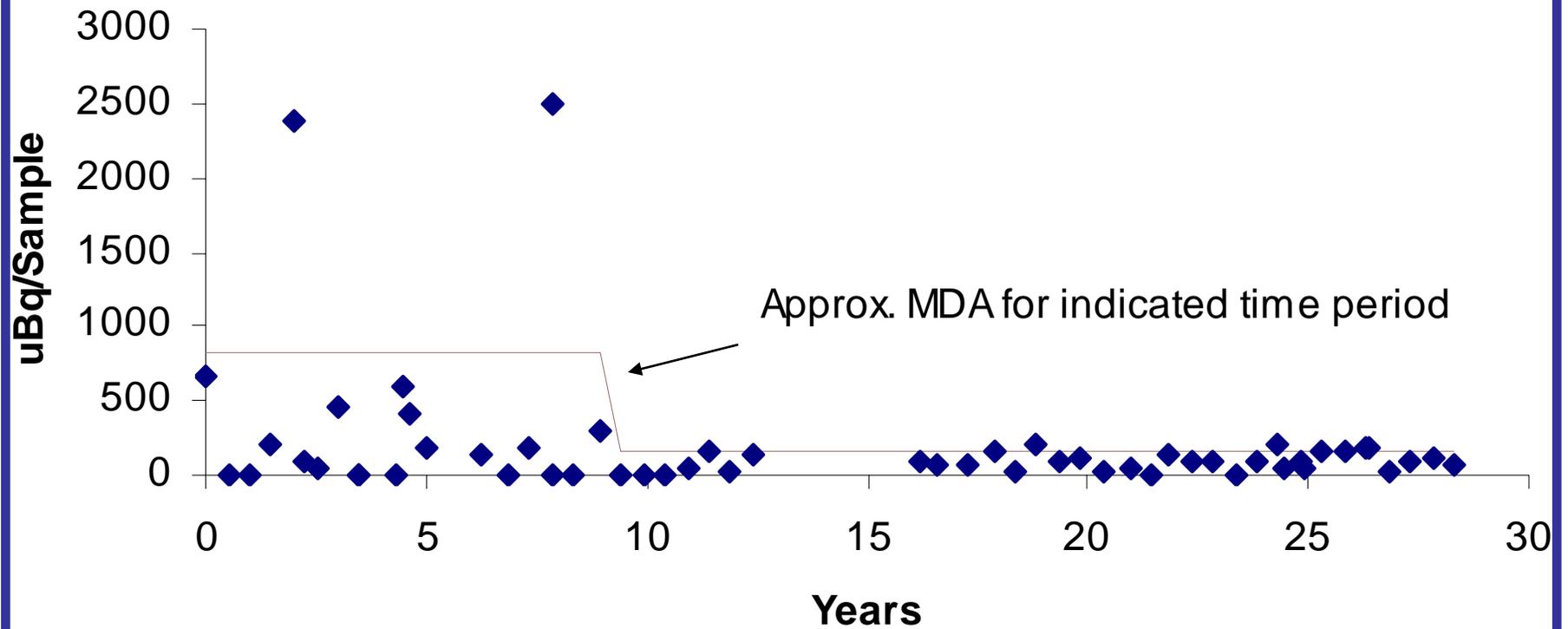


Linear relationship between positive alpha spectrometry count and AMS measures

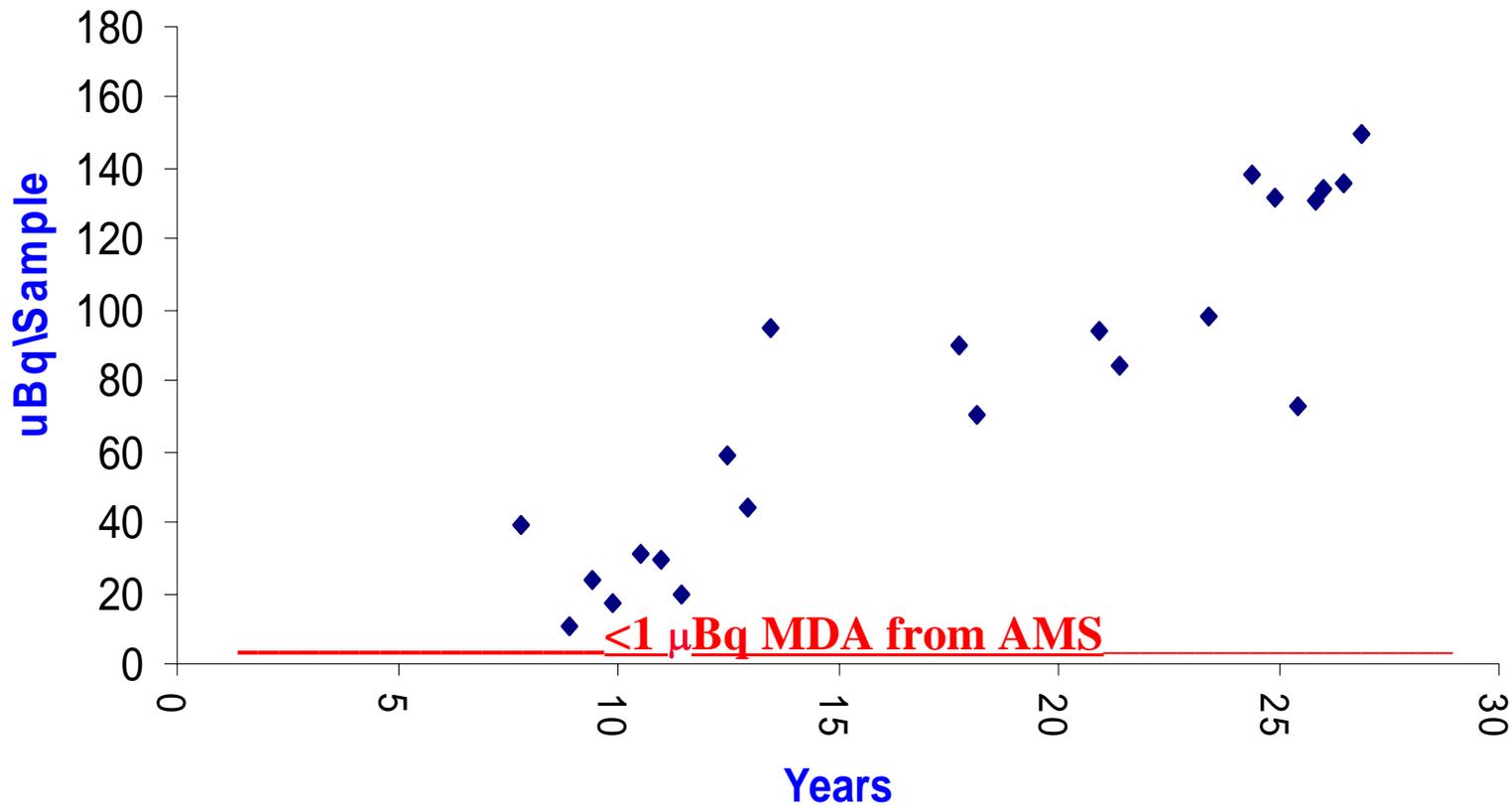
# AMS results for a known intake case



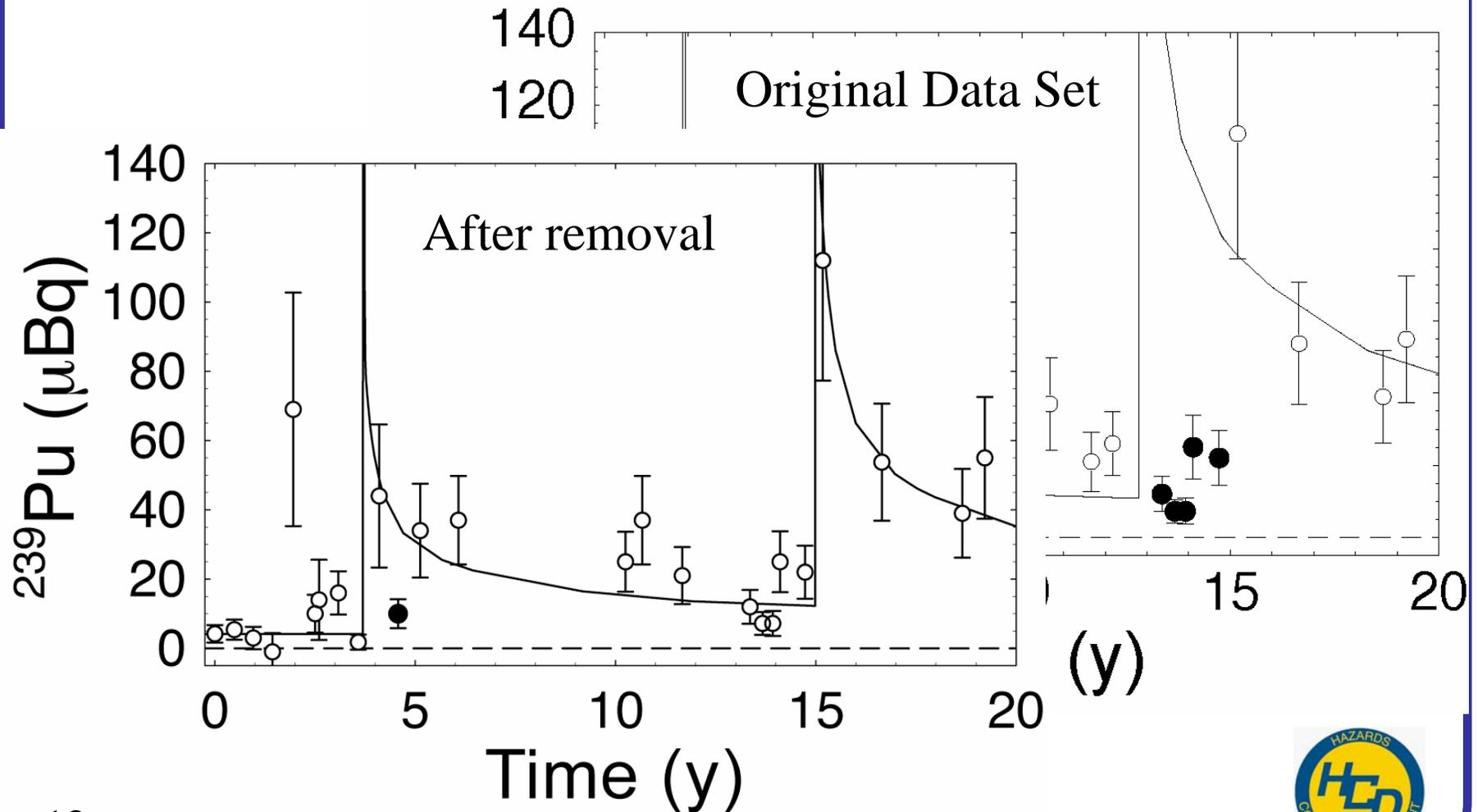
# Alpha spectrometry results for a suspected Pu-239 intake case



# AMS results for the same suspected Pu-239 intake case



# Effect of small amounts of Pu-239 contained in Am-243 tracer



# Discussion

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- **Linear response of AMS relative to alpha spectrometry for several orders of magnitude**
- **AMS is more sensitive than alpha spectrometry**
- **Alpha spectrometry tracers need to be well characterized if alpha spectrometry samples are reanalyzed with AMS.**

# Conclusions

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- **Analysis for Pu-239 twenty years post exposure is readily possible with AMS**
- **AMS reveals exposures at much lower levels than previously possible (<100 uSv @ 20 yrs)**
- **Determination of low level exposure to Pu-239 is possible for purposes of historical compensation (EEOIPCA) and litigation.**
- **Enhanced ability to study human excretion patterns of Pu**
- **No technology shortfall exists for monitoring current as well as past exposures to Pu-239 at regulatory levels when using AMS**

