

Neptunium-237 in Human Tissue Samples

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Introduction

NEPTUNIUM-237 is present in the environment in concentrations similar to those of ^{239}Pu because the $(n, 2n)$ to (n, γ) reaction ratio for ^{238}U , which is the principle production mode for both species in global fallout, is 0.7 ± 0.2 (Ef84). Approximately 3t of ^{237}Np was released into the biosphere by atmospheric testing. This study was designed to determine how much of this Np is still present in humans.

Materials and Methods

Tissue samples were collected at autopsy from individuals who had no known occupational exposure to any of the actinide elements. They were dried, ashed, dissolved, and stored as acid solutions. Separate aliquots were analyzed for Pu and Np. The aliquots for Pu were spiked and equilibrated with ^{242}Pu , and the concentrations were determined by the isotope dilution technique using both α pulse-height analyzers and thermal ionization mass spectrometers. The aliquots analyzed for Np were spiked and equilibrated with ^{236}Np . The Np was isolated by a 2-thenoyltrifluoroacetone extraction (Mo57). Final purification was ac-

complished on anion columns. The neptunium was electroplated on Re filaments for mass spectrometric analysis.

Results and Discussion

The results of the analyses are summarized in Table I where the total number of atoms of ^{237}Np and ^{239}Pu are reported for each sample. The standard deviations are estimates of the 95% confidence interval as determined by repetitive scans of single filament loadings. For the Pu analyses, these precision errors are the major source of the overall uncertainty because the accuracy of the isotope dilution technique using the mass spectrometers has been shown to be better than 99.9% by measurements of certified National Bureau of Standards Pu standards. The ^{236}Np tracer used for these analyses was calibrated against a Np standard having a 0.25% uncertainty associated with its concentration. A series of blanks were analyzed with the tissue samples. No detectable Pu or Np was observed. The limit of detection for ^{239}Pu was 5×10^5 atoms; the limit of detection for ^{237}Np was 2×10^5 atoms.

The average $^{237}\text{Np}/^{239}\text{Pu}$ atom ratio was 0.04. If one assumes that the subject individuals were exposed to global fallout that had a $^{237}\text{Np}/^{239}\text{Pu}$ atom ratio of 0.7 ± 0.2 , the ratios measured in these tissues suggest that Np has been lost preferentially to Pu in the lung tissues. These data also suggest that the Np being lost from the lungs is not being concentrated in the liver.

Table 1. Neptunium and plutonium concentrations in liver and lung tissue

Tissue Type	Tissue Mass (g)	^{237}Np Atoms	^{239}Pu Atoms	$^{237}\text{Np}/^{239}\text{Pu}$
lung	736	$7.66 \times 10^7 \pm 30\%$	$1.27 \times 10^9 \pm 11\%$	0.06
lung	581	$1.91 \times 10^8 \pm 15\%$	$1.88 \times 10^{10} \pm 9\%$	0.01
lung	381	$1.28 \times 10^9 \pm 14\%$	$3.23 \times 10^{10} \pm 6\%$	0.04
lung	500	$2.33 \times 10^9 \pm 10\%$	$2.91 \times 10^{10} \pm 9\%$	0.08
liver	804	$8.52 \times 10^8 \pm 14\%$	$2.12 \times 10^{10} \pm 6\%$	0.04
liver	1425	$3.22 \times 10^8 \pm 16\%$	$6.43 \times 10^9 \pm 10\%$	0.05
liver	550	$9.13 \times 10^8 \pm 11\%$	$3.93 \times 10^{10} \pm 5\%$	0.02
liver	482	$1.73 \times 10^9 \pm 12\%$	$3.38 \times 10^{10} \pm 5\%$	0.05
liver	241	$8.63 \times 10^8 \pm 13\%$	$1.69 \times 10^{10} \pm 7\%$	0.05

This preliminary study demonstrates the presence of Np in human lung and liver tissue at levels that can be detected by mass spectrometry. It appears that the concentration of ^{237}Np in the samples warrants sample sizes of approximately 100 g or more to ensure a level of 100 times minimum detection limits.

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