

An Investigation of Specific Problems with the Determination of Plutonium Isotopic Ratios in Environmental and Biological Samples

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Two projects were investigated in the course of this study. The first was to determine whether reported differences in the behavior of ^{238}Pu and $^{239+240}\text{Pu}$ could be observed in a laboratory situation and if so, to determine the mechanism(s). Soil samples were obtained from Rocky Flats Plant (RFP) and four islands in Bikini Atoll. The $^{238}\text{Pu}/^{239+240}\text{Pu}$ ratio was determined during column extractions, sequential batch extractions, and soil particle size studies to monitor the behavior of ^{238}Pu and $^{239+240}\text{Pu}$. The first conclusion was that the RFP soil indicated no differences in the behavior of ^{238}Pu and $^{239+240}\text{Pu}$. Even though previous reports had indicated differences in the isotopes at this site, this study suggested that these could be explained by analytical and statistical uncertainties in the data. For three of the soils from Bikini Atoll, ^{238}Pu was found to be more easily extracted from the soil than $^{239+240}\text{Pu}$. Furthermore, it was found that ^{238}Pu was enriched in the finer particles size fraction of the soil which explained its enhanced extraction. The second project involved the development of a combined alpha spectrometry and fission track method for the determination of the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio in human tissues. Dissolved tissues of occupationally exposed workers who were registrants of the United States Transuranium and Uranium Registries were obtained. In this study, Pu was purified and isolated from Am, U and Th, after the addition of ^{238}Pu as a radiotracer. After electrodeposition onto vanadium planchets, the $^{239+240}\text{Pu}$ activity was determined by alpha spectrometry. A fission track method was developed to determine ^{239}Pu in the presence of ^{238}Pu and ^{240}Pu , using the WSU Triga III reactor and LexanTM polycarbonate track detectors. Combining the two techniques allowed the determination of the $^{240}\text{Pu}/^{239}\text{Pu}$ ratios. This method provided an alternative to mass spectrometry, which is currently the routine method used for $^{240}\text{Pu}/^{239}\text{Pu}$ determinations.

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