COMPARATIVE DISTRIBUTION OF ^{238}U, ^{234}U, AND $^{239+240}Pu$ IN THE SAME SETS OF HUMAN TISSUES OF GENERAL POPULATION

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INTRODUCTION

The purpose of these studies is sufficient understanding of the metabolism of the different actinides in both man and experimental animals. Toward this end, we have determined the concentrations of "naturally" occurring uranium, thorium and plutonium in tissues of control beagle dogs from our research colony (1), and the concentrations of plutonium and thorium in human tissues from two states of the United States (2). Determining both plutonium and thorium in the same human tissue set allowed comparison of the distribution pattern of these two actinides, which have very similar chemical and biological properties. The present work describes the distribution of uranium and plutonium in human tissues. Because measurements of both elements are made on the same set of human tissues, effects of environmental, physiological, and dietary intake should be the same for both elements. The present work will also suggest the distribution patterns of the two actinides.

EXPERIMENTAL

Collection of Tissues

Ten sets of tissues were obtained at autopsy from persons who died suddenly from two states of the United States (Pennsylvania and Colorado). All ten tissue sets included lung, liver, kidney, vertebrae and ribs, but in some cases, we also obtained lymph nodes and spleen. For each subject, the tissue samples were placed in individual plastic bags and packaged in separate labeled containers. The specimens were frozen after autopsy and transported to our laboratory packed in dry ice.

The date of death, age at death, sex, residential history, smoking and drug history, past medical history and occupational data were obtained for each individual. The cause of death was determined by either gross inspection or toxicological examination. All the organs were examined for degeneration or impairment, and for evidence of tumors or carcinomas.

Radiochemical Determinations of Uranium and Plutonium

Uranium and plutonium were determined using the radiochemical procedures of Singh et al. (3,4). The weighed amounts of soft tissues were spiked with 1-2 dpm of ^{232}U and ^{242}Pu tracers. The tissues were wet ashed with HNO_3 followed by a mixture of HNO_3 and H_2SO_4, with occasional additions of a few drops of HNO_3 and H_2O_2. Uranium and plutonium were coprecipitated with iron as hydroxides, and extracted into a 20% tri-lauryl amine (TLA) solution in xylene from 10 M HCl. Plutonium was first back-extracted by reducing to the trivalent state with 0.05 M NH_4I solution in 8 M HCl. Uranium was then back-extracted by shaking with an equal volume of 0.1 M HCl. Uranium and plutonium were electrodeposited separately onto platinum discs and counted alpha-spectrometrically using a surface barrrier silicon diode and a multi-channel analyzer.

Weighed amounts of bone tissue were spiked with ^{232}U and ^{242}Pu tracers followed by slow heating on a hot plate. The bone was dry ashed in a muffle furnace at 550°C, followed by wet ashing with HNO_3, with occasional additions of HNO_3 and H_2O_2, until the evolution of brown fumes ceased. Uranium was reduced to the tetravalent state by addition of 200 mg $SnCl_2$ and 25 ml HI. Uranium and plutonium were coprecipitated as oxalate by 10% oxalic acid. The precipitate was heated in a muffle furnace at 550°C overnight and dissolved in 10 M HCl. Uranium and plutonium were extracted, back-extracted, electrodeposited and counted alpha-spectrometrically as described for soft tissues.

RESULTS

The mean concentrations of plutonium and uranium are given in Figure 1. Among soft tissues, the concentrations of $^{239,240}Pu$ were highest in liver, ranging from 0.14 to 2.41 pCi/kg (mean: 1.06 ± 0.26 pCi/kg wet weight), followed by lung with a range of 0.06 to 1.79 pCi/kg (mean: 0.43 ± 0.22 pCi/kg). Concentrations in kidney ranged from 0.01 to 0.09 pCi/kg (mean: 0.05 ± 0.01 pCi/kg). In lymph nodes, the concentrations ranged from 0.04 to 3.54 pCi/kg (mean: 1.02 ± 0.56 pCi/kg); however, these values are as high as the analytical errors associated with the measurement.

The concentrations of ^{238}Pu in all soft tissues were below the detection limit except in liver, where the concentration ranged from 0.02 to 0.10 pCi/kg (mean: 0.05 ± 0.01 pCi/kg wet weight).

The concentration of $^{239,240}Pu$ in vertebrae ranged from -0.12 to 0.34 pCi/kg (mean: 0.18 ± 0.04 pCi/kg). Concentrations ranged from 0.03 to 0.41 pCi/kg (mean: 0.21 ± 0.06 pCi/kg) in ribs, and from 0.002 to 1.46 pCi/kg (mean: 0.46 ± 0.26 pCi/kg) in sternum. The concentrations of ^{238}Pu were below the detection limit in these bones.

The concentrations of ^{238}U were highest in lymph nodes, ranging from 0.51 to 19.88 pCi/kg (mean: 5.2 ± 3.0 pCi/kg). In lung, concentrations ranged from 0.27 to 2.28 pCi/kg (mean: 1.18 ± 0.26 pCi/kg); in kidney they ranged from 0.12 to 3.40 pCi/kg (mean: 0.68 ± 0.32 pCi/kg). Liver contained the lowest concentrations, ranging from 0.02 to 0.76 pCi/kg (mean: 0.15 ± 0.08 pCi/kg).

The concentrations of ^{234}U followed the same pattern. Lymph nodes contained the highest concentration, ranging from 0.99 to 19.2 pCi/kg (mean: 7.6 ± 2.7 pCi/kg), followed by lung with a range of 0.19 to 3.16 pCi/kg (mean: 1.36 ± 0.39 pCi/kg). Concentrations in kidney ranged from 0.10 to 3.89 pCi/kg (mean: 0.91 ± 0.36 pCi/kg). Liver contained the lowest concentrations, ranging from 0.08 pCi/kg to 0.94 pCi/kg (mean: 0.24 ± 0.09 pCi/kg).

The concentrations of ^{238}U were similar in vertebrae and ribs, ranging from 0.09 to 4.66 pCi kg (mean: 0.85 ± 0.43 pCi/kg), and from 0.14 to 2.81 pCi/kg (mean: 0.87 ± 0.40 pCi/kg), respectively. The concentrations of ^{234}U were also similar in vertebrae and ribs with ranges of 0.17 to 5.75 pCi/kg (mean: 1.08 ± 0.54 pCi/kg), and of 0.26 to 4.47 pCi/kg (mean: 1.17 ± 0.58 pCi/kg), respectively. The sternum contained slightly higher concentrations of ^{238}U and ^{234}U with ranges of 0.25 to 3.91 pCi/kg (mean: 1.63 ± 0.62 pCi/kg) and 0.27 to 6.54 pCi/kg (mean: 2.34 ± 1.08 pCi/kg), respectively.

The concentrations of ^{235}U in all tissues were below the limit of detection (<0.01 pCi).

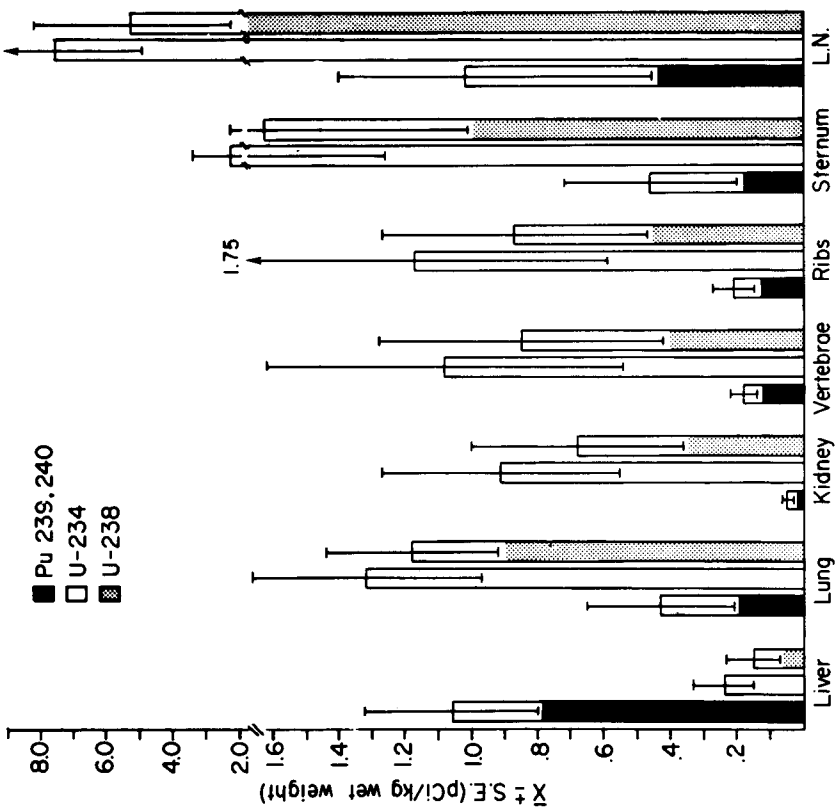

Figure 1. Comparisons of the concentrations (pCi/kg wet weight) of $^{239+240}Pu$, ^{234}U, and ^{238}U in the same sets of human tissues from the general population of the United States.

DISCUSSION

A comparison of uranium and plutonium concentrations are given in Figure 1. There are certain obvious differences between the concentrations of uranium and plutonium in different organs. The large differences between the concentrations of uranium isotopes (^{238}U and ^{234}U) and $^{239,240}Pu$ in lung may simply be due to the higher concentrations of uranium isotopes in the air as compared to plutonium isotopes.

The other obvious difference is between the concentrations of uranium and plutonium in the liver. The concentration of $^{239,240}Pu$ was highest in the liver, yet the concentration of uranium was lowest in the liver. This may be due to the fact that inhaled plutonium, when cleared from the lungs and circulated in the bloodstream, hydrolyzes to form colloidal particles which are taken up by reticuloendothelial (RE) cells in the liver. Uranium is believed to occur in the body as UO_2^{++}, which does not undergo hydrolysis; it accumulates mostly in bone and clears through the kidney. Hydrolysis occurs more readily in elements with higher valence and small ionic size; therefore, plutonium, which is mostly in the tetravalent state and has an ionic size of 0.93 Å, hydrolyzes more readily than uranium, which exists in the hexavalent state but behaves as divalent because it is present as UO_2^{++} and has a larger ionic size than Pu(IV).

The higher concentrations of ^{238}U and ^{234}U as compared to $^{239,240}Pu$ in bones may possibly be due to a larger intake of uranium through the food chain and inhalation. Only a very small amount of plutonium is inhaled, and a negligible amount is ingested through the food chain.

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