Elemental bio-imaging of thorium, uranium and plutonium in tissues from occupationally exposed former nuclear workers

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Internal exposure from naturally occurring radionuclides (including the inhaled long-lived actinides $^{232}$Th and $^{238}$U) is a component of the ubiquitous background radiation dose. It is of interest to compare the concentration distribution of these natural α-emitters in the lungs and respiratory lymph nodes with those resulting from occupational exposure, including exposure to anthropogenic plutonium, depleted and enriched uranium. This study examines the application of laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) to quantifying and visualizing the mass distribution of uranium and thorium isotopes from both occupational and natural background exposure in human respiratory tissues, and for the first time, extends this application to the direct imaging of plutonium isotopes. Sections of lymphatic and lung tissues taken from deceased former nuclear workers with known history of occupational exposure to specific actinide elements (uranium, plutonium or americium) were analyzed by LA-ICP-MS. Using a previously developed LA-ICP-MS protocol for elemental bio-imaging of trace elements in human tissue and a new software tool, we generated images of thorium ($^{232}$Th), uranium ($^{235}$U and $^{238}$U) and plutonium ($^{239}$Pu and $^{240}$Pu) mass distributions in sections of tissue. We used a laboratory-produced matrix-matched standard to quantify the $^{232}$Th, $^{235}$U and $^{238}$U concentrations. The plutonium isotopes $^{239}$Pu and $^{240}$Pu were detected by LA-ICP-MS in 65-µm-diameter localized regions of both a paratracheal lymph node and a sample of lung tissue from a person who was occupationally exposed to refractory plutonium (plutonium dioxide). The average (overall) $^{239}$Pu concentration in the lymph node was 39.2 ng/g, measured by High Purity Germanium (HPGe) γ-spectrometry. Localized mass concentrations of thorium ($^{232}$Th) and uranium ($^{238}$U) in lymph node tissue from a person not occupationally exposed to these elements (chronic natural background inhalation exposure) ranged up to 400 and 375 ng/g, respectively. In lung samples of occupationally non-exposed to thorium and uranium workers, $^{232}$Th and $^{238}$U concentrations ranged up to 200 and 170 ng/g, respectively. In a person occupationally exposed to air-oxidized uranium metal, the maximum $^{235}$U and $^{238}$U isotopic mass concentrations in a lymph node, measured at higher resolution (with a 30-µm laser spot diameter), were 70 and 8500 ng/g, respectively. The ratio of these simultaneously measured mass concentrations signifies natural uranium. The current technique was not sufficiently sensitive, even with a 65-µm laser spot diameter, to detect $^{241}$Am (at an overall tissue concentration of 0.024 ng/g, i.e., 3 Bq/g).

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