Elemental Bio-imaging of Uranium and Plutonium in Tissues from Occupationally Exposed Former Nuclear Workers

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The mission of the United States Transuranium & Uranium Registries (USTUR) is to (1) evaluate health outcomes, causes of death and life expectancy of former nuclear workers (volunteer registrants) who had documented accidental intakes of (trans)uranium elements (2) obtain, preserve, and make available for future research, samples of tissues at autopsy (3) conduct radiochemical analyses, as necessary, to validate and develop new state-of-the-art methods for quantifying tissue doses and their associated uncertainties (4) apply USTUR case study data to refine dose assessment methods for these internal emitters as the bases for reliable epidemiological studies, risk projection, and credible standards for radiological protection (5) assess adequacy of historical and current U.S. regulatory controls and practices in limiting tissue doses to workers having the greatest health risk from intakes of uranium and the transuranium elements [1].

This study examines the application of synchrotron radiation based micro-XRF (SR micro-XRF) to quantifying and visualizing the elemental distribution of uranium and plutonium isotopes from occupational exposure in human respiratory and lymph tissues. Sections of lymphatic and lung tissues were taken from deceased former nuclear workers with a known history of occupational exposure to specific actinide elements (uranium, plutonium, or americium). The first examined tissue sample (lymph node tissue) originates from USTUR registrant 1060, who worked at Hanford for 40 years. Urine data (1948-1950) indicates chronic exposure to uranium (U_3O_8), while working at the Melt Plant, a spike (317 µg/day) in urine data (1948) even indicates an acute uranium uptake in addition to chronic exposure. The second examined tissue (lung tissue) is from USTUR registrant 0407 who worked at Rocky Flats Plant for 17 years. In 1965 the registrant was involved in an extensive Pu fire, studies of the fire aerosol showed a highly refractory material which retains in the lungs, which was not responsive to intravenous chelation treatment with Ca-DTPA. A study of the surface contamination indicated Pu particles of 0.12 µm count median diameter [2].

The samples, which were originally frozen at -30 °C (the standard protocol for long-term storage of NHRTR tissues), were defrosted and fixed in 10% buffered formalin for preservation during shipment. After dehydration in graded water/acetone series and embedding in paraffin, thin section of 16 µm were obtained by a microtome, which were finally mounted free-standing on a dedicated sample holder for micro-XRF analysis. A multilayer monochromator delivered a high-flux monochromatic beam of ~19.5 keV optimally exciting the U-L3 and Pu-L3 edges. A slightly defocused polycapillary optic (XOS) was used to focus the incoming X-ray beam to a microbeam with moderate resolution (~80 µm FWHM) for obtaining overview scans of the tissues. When optimally aligned, elemental distributions of regions of interest could be obtained with the conventional microbeam (~17.5 µm FWHM). A dual detector configuration using two silicon drift detectors (SiII, Vortex) was used to collect fluorescence with a larger solid angle [3].

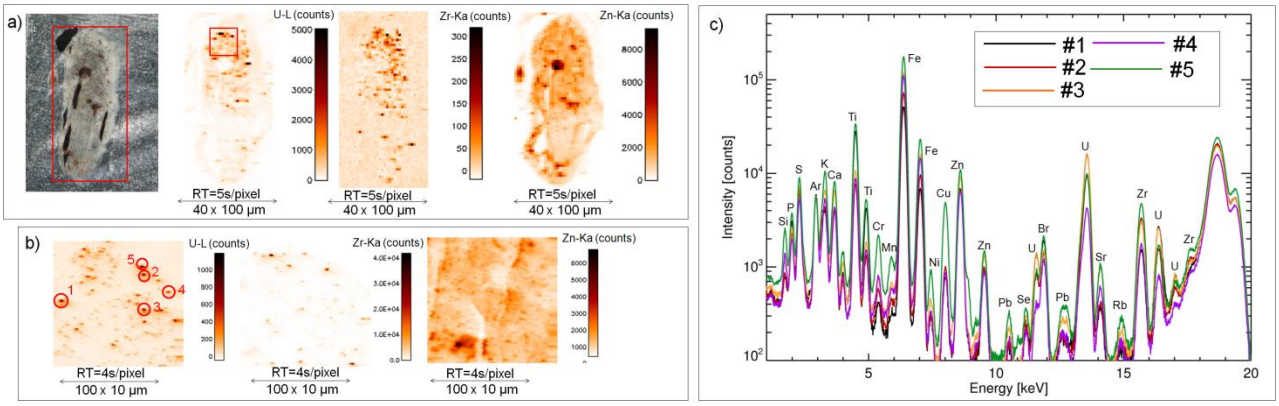

Figure 1: (a) microscope image and elemental distributions of U, Zr and Zn obtained with 80 µm FWHM beam in USTUR case 1060 (b) region of interest element maps obtained with 17.5 µm FWHM beam (c) XRF spectra from point measurements on 'hot spots' in region of interest element maps.

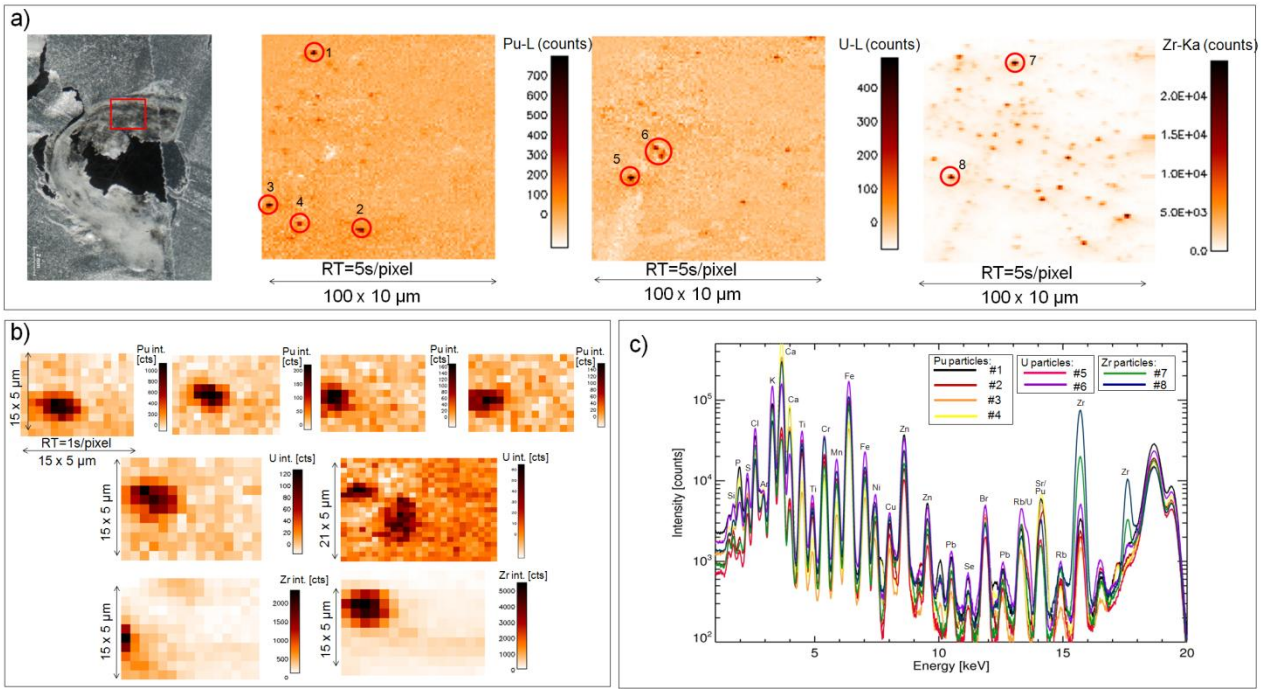

Figure 2: (a) microscope image and elemental distributions of Pu, U and Zr obtained with 17.5 µm FWHM beam in USTUR case 0407 (b) region of interest element maps with 17.5 µm FWHM beam (c) XRF spectra from point measurements on 'hot spots' in region of interest element maps.

Fig. 1 shows the elemental distributions obtained from USTUR case 1060 using the 80 µm beam (a) and 17.5 µm beam (b). On both resolution scales, U and Zr 'hot spots' were found which are not coinciding. Collection of point measurements on the U hot spots and spectral fitting with the AXIL software package revealed clear presence of U-L fluorescence peaks. Fig. 2 shows the presence of Pu and U non-coinciding 'hot spots' present in USTUR case 0407 (a). After rescanning the hot spots in smaller maps (b), XRF point measurements were performed clearly showing the presence of U-L and Pu-L fluorescence peaks.

From these measurements it can be concluded that synchrotron radiation based X-ray fluorescence is a powerful tool for trace level imaging and potential quantification of (trans)uranium elements within biological tissues. The results could contribute to a further refinement of dose assessment methods for these internal emitters as the bases for reliable epidemiological studies, risk projection, and credible standards for radiological protection.

References

[1] http://www.ustur.wsu.edu/

[2] D. Hare, S. Tolmachev, A. James, D. Bishop, C. Austin, F. Fryer, P. Doble, Anal. Chem., **82**, 3176-3182 (2010).

[3] B. De Samber et al., Dual Detector micro-XRF Cryotomography and Mapping, Hasylab Annual Report (2009)