

Elemental Bio-imaging of Actinides and Beryllium in Lymph Nodes of Former Nuclear Workers



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Introduction

The U.S. Transuranium & Uranium Registries (USTUR) studies the uptake, translocation and biokinetics of actinides in humans. This study explored the application of laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) to elemental bio-imaging (EBI) of actinides [thorium (Th), uranium (U), plutonium (Pu), americium (Am)] and beryllium (Be) in samples of human tissue. The tissue samples were from occupationally exposed 'nuclear weapons site' workers, who had voluntarily donated their tissues to the USTUR. For this exploratory study, single sections of thoracic (respiratory tract) lymph nodes were mapped for actinide isotopes and ⁹Be.

The aim of this work was to develop LA-ICP-MS protocols for elemental bio-imaging (elemental distribution) of actinides and beryllium isotopes in soft tissue samples from occupationally exposed individuals.

Material and Methods

• USTUR Samples

Five formalin-fixed thoracic lymph nodes from USTUR Cases 0303, 0407, 0695 and 1028 were selected for the analysis at Elemental Bio-Imagine (EBI) Facility, University of Technology (UTS).

USTUR Case #	LN type	Type of exposure	Exposure nuclide		Comments
			Primary	Be	
0303	Tracheobronchial	Puncture Wound	²³⁹ Pu	no	Chelated
0407	Paratracheal	Acute Inhalation	²³⁹ Pu	no	Chelated
0695	Parabronchial	Chronic Inhalation	²³⁹ Pu	yes	24 yr (Be); Berylliosis
0846	Pulmonary	Chronic Inhalation	²⁴¹ Am	yes	Chelated, 20 yr (Be)
1060	Parabronchial	Acute Inhalation	²³⁸ U	no	317 µg d⁻¹ urinary

• Laser Ablation ICP-MS at Elemental Bio-Imaging Facility, University of Technology Sydney



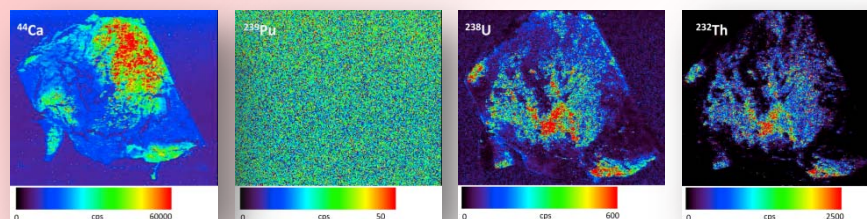
Agilent 7500ce ICP-MS		New Wave UP213 Laser Ablation	
RF Power	1250W	Wavelength	213 nm
Cooling gas flow rate	15 L min ⁻¹	Repetition frequency	20 Hz
Carrier gas flow rate	1.1 L min ⁻¹	Laser energy density	0.3 J cm ⁻²
Sample depth	4.0 mm	Spot size	12-30 µm
QP Bias	-5 V	Scan rate	12-30 µm s ⁻¹
OctP Bias	-8 V	Line spacing	12-30 µm
Scan mode	Peak hopping	Carrier gas	Ar
Dwell time	0.1 s per m/z		
Extracts 1, 2	6.8, -126 V		

The LA-ICP-MS system was tuned for maximum sensitivity prior to each experiment using NIST 612 Trace Elements in Glass. Laser scan speed was equal to the laser spot diameter. Thus, the image resolution was equal to the laser spot diameter.

Matrix matched tissue standards were used to quantify elemental concentrations. Tissue standards were prepared by taking known masses of chicken muscle tissue and spiking them with 1000 mg/L standard solutions in 2% HNO₃. The spiked tissues were then homogenised.

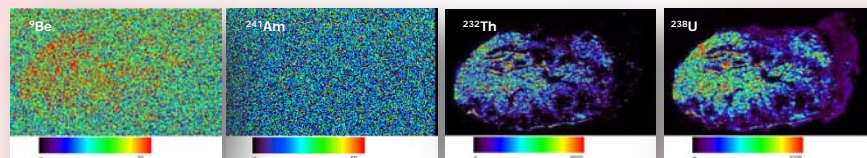
Results

• USTUR Case 0303 : Hanford - 1968 Puncture Wound - Soluble ²³⁹Pu - Chelated - Ca-DTPA



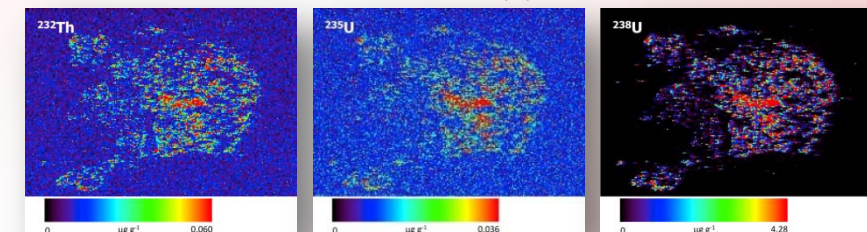
Elemental distribution maps for ⁴⁴Ca, ²³⁹Pu (not detected), ²³²Th, and ²³⁸U. **Non-quantitative analysis.**

• USTUR Case 0846: U.S. Radium Corp. - 1964-7 Chronic Inhalation - ²⁴¹AmO₂ - Chelated - Ca-DTPA



Elemental distribution maps for ⁹Be, ²⁴¹Am (not detected), ²³²Th, and ²³⁸U. **Non-quantitative analysis.**

• USTUR Case 1060: Hanford - 1948 Acute Inhalation - U₃O₈ (likely fume)



Concentration distribution maps for ²³²Th, ²³⁵U and ²³⁸U. **Quantitative analysis.**

Conclusions

- LA-ICP-MS protocols for elemental bio-imaging of actinide elements and beryllium were developed and successfully applied to the analyses of lymph nodes of former nuclear workers.
- ²³²Th and ²³⁸U were quantitatively measured in all samples and ²³⁵U was detected in a U-exposed case.
- The distribution of ⁹Be was measurable in one of two cases with 'self-reported' exposure to Be.
- Neither ²³⁹Pu nor ²⁴¹Am was detectable. LA-ICP-MS measurement conditions need to be optimized to decrease the limit of detection for ²³⁹Pu and ²⁴¹Am.