Quantifying Radium-226 in Heart Tissue Using ICP-QQQ-MS: A Study of Radium Exposures

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Abstract

This study investigates the distribution of radium (^{226}Ra) in cardiac and bone samples of two exposed individuals. ^{226}Ra was preconcentrated from acid-digested tissues using cation exchange chromatography and measured with an Agilent 8900 ICP-QQQ-MS. An instrumental detection limit was 3.4 fg ^{226}Ra/g (0.12 mBq ^{226}Ra/g) and method limit of detection was 4.8 fg ^{226}Ra/g material (0.18 mBq ^{226}Ra/g material). ^{226}Ra concentrations in heart tissue samples from Case 01-0175 ranged from 0.047 pg ^{226}Ra/g tissue (1.9 mBq ^{226}Ra/g tissue) to 1.5 pg ^{226}Ra/g tissue (54 mBq ^{226}Ra/g tissue). For Case 03-666 bone samples, ^{226}Ra concentration of 2200 pg ^{226}Ra/g bone (81 Bq ^{226}Ra/g bone) was measured in the femur and 1700 pg ^{226}Ra/g bone (64 Bq ^{226}Ra/g bone) in the vertebra.

Introduction

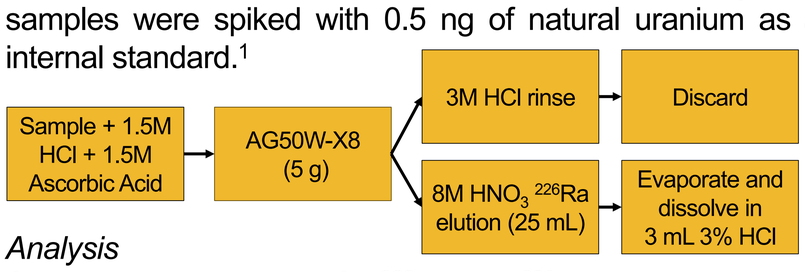
- The United States Transuranium and Uranium Registries (USTUR) follows up occupationally exposed workers by studying the biokinetics and tissue dosimetry of the actinides to refine dose assessment methods in support of reliable epidemiological studies, radiation risk assessment, and regulatory standards for radiological protection of workers and public.
- The radium dial workers cohort is a valuable resource for developing biokinetic models of radium in the human body.[1]
- Early studies found increased jaw infections and anemia.[5]

Case	Cohort	Tissue type	Estimated intake	
01-175	Radithor	Cardiac	1.01×10^{7} Bq	273 µg
03-666	Dial Painter	Bone	5.89×10^{8} Bq	1592 µg

Methods

Sample Preparation

Acid-digested samples were received from the USTUR.[6] From a 15 mL aliquot, ^{226}Ra was separated and concentrated using a cation exchange resin (AG 50W-X8). ^{226}Ra was eluted from the column with 8M HNO_3, evaporated to dryness, dissolved in 3 ml of 3% HCl acid, and submitted for ICP-MS analysis. All samples were spiked with 0.5 ng of natural uranium as an internal standard.[1]



Analysis

Samples were analyzed for ^{226}Ra and ^{238}U on an Agilent 8900 in standard mode, without collision gas. The 3% HCl sample was measured (r_T), then spiked with 0.5 pg ^{226}Ra standard to achieve 2–3× the signal (C), then analyzed again (r_{T+C}). Concentration in solution (T) was calculated using the method of standard editions and the following equation:[2]

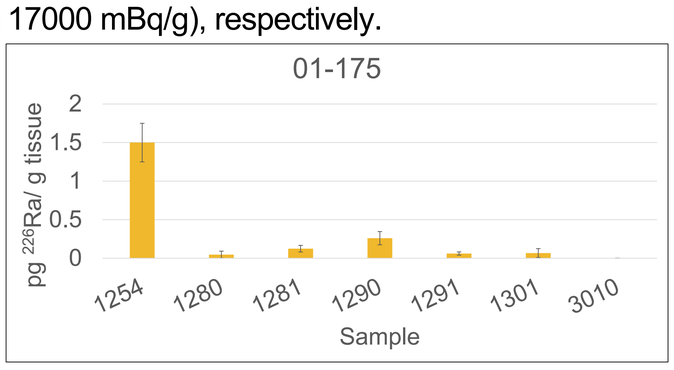
$$T = C\frac{r_T}{r_{T+C}-r_T}$$

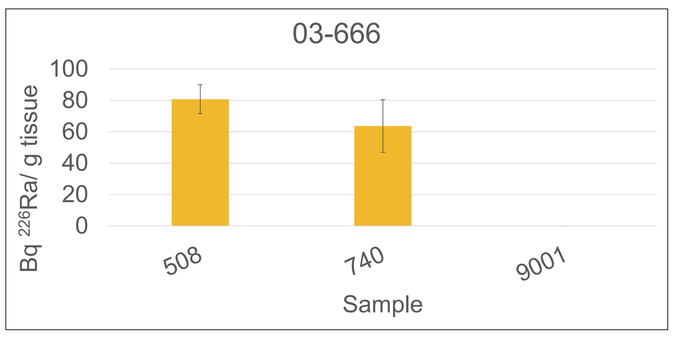
Results

A 1 pg/g ^{226}Ra solution yielded a signal of 1410 ± 45 cps. The instrument blank averaged 2 cps, confirming adequate inter-sample and standard washout. The instrumental LOD for ^{226}Ra was 3.4 fg ^{226}Ra/g (0.12 mBq ^{226}Ra/g) and the sample LOD was 4.8 fg ^{226}Ra/g material (0.18 mBq ^{226}Ra/g material). ^{226}Ra recovery through the chromatographic process was assessed using an acid-matched solution spiked with 1.5 pg ^{226}Ra and resulted in 97% ± 14% (n=7) recovery. Total propagated uncertainties were calculated within one standard deviation using the guidelines for uncertainty measurement (GUM) workbench.

^{226}Ra was measured in all cardiac tissues from case 01-175. The highest concentration was observed in the aorta (1.5 ± 0.3 pg ^{226}Ra/g), while the lowest was measured in the right atrium (0.047 ± 0.045 pg ^{226}Ra/g). ^{226}Ra was non-uniformly distributed among cardiac tissue samples.

For Case 03-666, similar ^{226}Ra concentrations were measured in the femur middle shaft and the body of the 7th thoracic vertebra: 2210 ± 250 pg ^{226}Ra/g (80800 ± 9200 mBq/g) and 1740 ± 460 pg ^{226}Ra/g (63700 ± 17000 mBq/g), respectively.





Sample number	Tissue sample	pg ^{226}Ra/g	mBq ^{226}Ra/g
01-175-1254	Aorta	1.5 ± 0.3	54 ± 10
01-175-1280	Atrium (R)	0.047 ± 0.045	1.9 ± 1.6
01-175-1281	Ventricle (R)	0.12 ± 0.04	4.5 ± 1.6
01-175-1290	Atrium (L)	0.26 ± 0.09	10 ± 3
01-175-1291	Ventricle (L)	0.060 ± 0.02	2.2 ± 0.8
01-175-1301	Adipose (heart)	0.067 ± 0.06	2.5 ± 2.1
03-666-508	Femur (L): Middle shaft	2210 ± 250	80800 ± 9200
03-666-740	Vertebra: Thoracic 7 body	1740 ± 460	63700 ± 17000

Discussion

- The LOD in this work was 3.4 fg/g in 1 minute, which is greater than alpha spectrometry at 0.027 fg/g with a count time of up to 7 days, and MC-ICP-MS at 0.55 fg/g with a count time of 10 minutes.[3,1]
- Acid matched blanks 3010 and 9010 revealed no ^{226}Ra content.
- Observed ^{226}Ra concentrations in bone samples were higher than those in heart tissue, consistent with the greater exposure for case 03-666.[5]
- ^{226}Ra concentrations in the aorta were greater than the ventricles and atria.
- No prominent trend in accumulation between right and left systems or atria and ventricles.
- Sample heterogeneity and proximity to LOD may have caused high error in the ventricle(R) and adipose tissue.

Conclusions

- For a single case, ^{226}Ra is not uniformly distributed in the heart.
- Cation exchange chromatography is effective for ^{226}Ra separation in soft human tissues.
- The Agilent 8900 combined with the method of standard additions is capable of low-level ^{226}Ra measurements.
- LOD could be improved by using a desolvating nebulizer sample introduction system or helium collision gas.

Acknowledgements

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