



Measurement of radium isotopes with the ANU AMS facility

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SUMMARY. In contaminated environments the spatial distribution of thorium should be far more uniform than that for uranium. Accordingly, measurements of the $^{228}\text{Ra}/^{226}\text{Ra}$ ratio may provide a probe with which to assess variations in the amount of uranium-process derived ^{226}Ra . Furthermore, for contaminated or rehabilitated areas where the $^{226}\text{Ra}/^{228}\text{Ra}$ ratio is anomalous, measurements of the transport of material away from the site via the ratio could provide information on the local erosion rate.

Accelerator Mass Spectrometry (AMS) adds a tandem ion accelerator and additional analysis stages to a conventional mass spectrometry arrangement, in order to facilitate ultra-trace level abundance measurements of selected isotopes. In doing so, it also makes use of the detection and analysis techniques of traditional nuclear physics. For the $^{226,228}\text{Ra}$ isotopes AMS offers a number of advantages over the more traditional techniques of α - and γ - spectroscopy. AMS requires less sample mass, and because of its very high selectivity provides excellent discrimination against potential interferences. The smaller sample size ($\sim 1\text{g}$) also allows a considerable simplification of the radio-chemical processing compared with α -spectroscopy. Two major advantages are the ability to measure both isotopes with the one technique without the necessity of waiting for ^{228}Th to grow in and, that once prepared, the $^{228}\text{Ra}/^{226}\text{Ra}$ ratio for ~ 30 samples can be determined in about a day. This paper will describe the AMS technique, and highlight recent developments in the measurement of $^{226,228}\text{Ra}$ with the ANU system.

1. Introduction

The dispersion of radionuclides into the environment through surface water systems is an important pathway via which uranium mines and ore processing facilities can impact on the surrounding environment. The consumption of food obtained from the impacted area and/or direct γ -irradiation from material deposited on the surface within the area, are the dominant means by which a radiation dose can be received. Radium-226 is invariably the dominant contributor to the ingested component of the dose, and hence knowledge of the extent of the affected area and of the rate at which it is dispersing into the surrounding environment is important. The $^{228}\text{Ra}/^{226}\text{Ra}$ ratio may provide a probe with which to assess both these quantities. Radium-228 arises in the ^{232}Th decay series, while ^{226}Ra arises in the ^{238}U decay series. Because the spatial distribution of thorium should be far more uniform than that for uranium, the impacted area should show a change in the $^{228}\text{Ra}/^{226}\text{Ra}$ ratio relative to the surrounding region.

AMS combines the high efficiency of conventional mass spectrometry with excellent discrimination against isobaric and molecular interferences. This is achieved by injecting mass-selected negative ions into an accelerator and, after a first stage of acceleration, dissociating any molecular ions and stripping atomic ions to positive charge states. A second stage of acceleration is followed by magnetic and electrostatic analyses, and finally an ion detector is used to identify individual ions. Essential features of the AMS system currently used for radium analysis at the ANU are depicted in figure 1.

The AMS facility at the ANU has, in recent years, seen the development of techniques for the measurement of ^{236}U , ^{237}Np and $^{239,240,242,244}\text{Pu}$ isotopes (Fifield *et al.* (1996), Fifield *et al.* (1997)). All three elements produce good beams of negative monoxide ions for injection into the accelerator. This is not the case for radium isotopes. Negative

radium di-carbide ions have, however, proven to be a suitable alternative (Tims *et al.* in press), and their use is part of the current development for measuring radium taking place at the ANU.

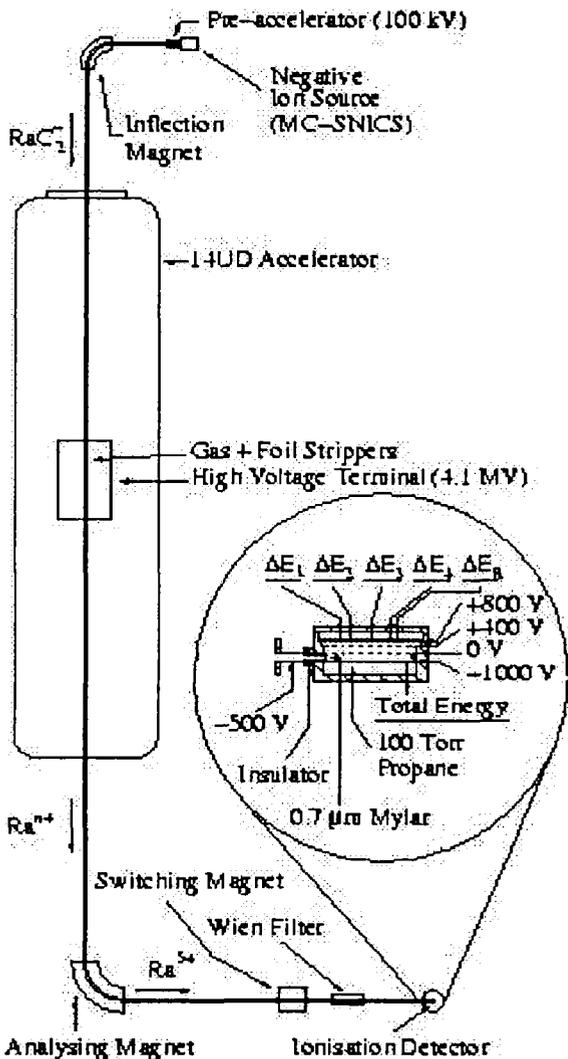


Figure 1. Schematic of the AMS system at the ANU.

2. Experimental details.

AMS measurements were carried out using the 14UD pelletron accelerator at the Australian National University and a methodology little changed from that described in (Fifield *et al.* (1996), Fifield *et al.* (1997)). Ions of charge state 5^+ and energy ~ 24 MeV were identified using only the total energy signal from a multi-anode gas ionisation detector (Fifield *et al.* (1987), Ophel *et al.* (1988)). The choice of the 5^+ state is a consequence of ongoing development of the ion source and beam transport system at the ANU, which has significantly improved the transmission of the beam through the accelerator. The accelerator potential and inflection magnet are cycled back and

forth between the appropriate values for each radium isotope so that each is injected into the analyzing magnet with the same magnetic rigidity. The cycle is typically 1 minute for ^{226}Ra and 3 minutes for ^{228}Ra , with a total measurement time for each sample ~ 15 minutes.

Samples are prepared for radium analysis as follows. The material is dissolved in acid and the radium extracted with an ion exchange column to separate ^{228}Th and ^{228}Ac , that arise as part of the ^{232}Th decay chain, and which would cause isobaric interference with the ^{228}Ra . The resultant solution is mixed with iron nitrate and graphite powder before drying at 90°C . Samples are then baked for 8 hours at 400°C , to remove any residual moisture and reduce the iron nitrate to iron oxide, thus yielding a carbon/iron-oxide matrix, pressed into holders and mounted in the ion source. The above procedure, which is still under development, aims to optimise the intensity and stability of the radium di-carbide ions extracted from the sample.

A number of test samples have also been measured to assess the detection limit of AMS for radium. To this end samples with varying $^{228}\text{Ra}/^{226}\text{Ra}$ ratios have been prepared from calibrated ^{228}Ra and ^{226}Ra solutions and the yield of each isotope recorded with the ionisation detector.

To measure the $^{228}\text{Ra}/^{226}\text{Ra}$ ratio only requires a single sample. If the radium concentration is also of interest then a second sample is required, with an added, known amount of either isotope as a tracer. The concentration can then be deduced from the measured ratio from both samples and the known amount of added tracer. Since the naturally occurring ^{228}Ra concentration is typically much lower than that for ^{226}Ra , a ^{228}Ra tracer is preferable. The tracer need not be isotopically pure, provided the $^{228}\text{Ra}/^{226}\text{Ra}$ ratio is known, but materials with a low ^{226}Ra content are clearly preferred. Suitable tracer materials are presently being evaluated using AMS measurements of the $^{228}\text{Ra}/^{226}\text{Ra}$ ratio in samples prepared with and without the addition of a known amount of ^{226}Ra .

3. Results

Injecting RaC_2^- from samples prepared with the technique described above leads to Ra^{5+} beams that are a factor ~ 50 times greater than those obtained by injecting RaO^- from oxide samples. The count rates measured in the detector are now typically ~ 1200 counts. min^{-1} per (picogram of radium / mg of matrix). Recent tests using silver nitrate in place of iron nitrate suggest that this rate could be further increased by a factor ~ 2 . The natural ^{228}Ra concentration in soil is typically ~ 5 fg per gram, and hence the above rates imply that, for a 5 g

sample and a matrix mass of 1 mg, ~1000 counts would be obtained in 15 minutes. The corresponding calculation for ^{226}Ra yields over 10^4 counts per minute. Furthermore, the stability of the radium beams has improved noticeably.

Measurements to determine the detection limit using the calibrated solutions indicate a sensitivity $\sim 10^7$ atoms which is comparable with that afforded by α -spectroscopy (Hancock and Martin (1991)), and approaches that provided by the isotope dilution TIMS technique (Volpe *et al.* (1991)), which have ^{226}Ra detection limits $\sim 10^7$ and $\sim 3 \times 10^6$ atoms, respectively. However, the calibrated ^{228}Ra solution presently in use has a significant ^{226}Ra component, which complicates the analysis, and is also driving the search for a better ^{228}Ra tracer material. Several candidates have already been identified, and preliminary measurements of the $^{228}\text{Ra}/^{226}\text{Ra}$ ratios are encouraging. Furthermore, the above estimate is based on early data, collected before the optimal sample preparation technique had been established, and an improvement in the detection limit can therefore be expected.

4. Conclusion

The present work indicates that AMS has the required sensitivity to measure the $^{228}\text{Ra}/^{226}\text{Ra}$ ratio in environmental samples. The recent improvement

in the intensity and stability of the radium beams is encouraging, and make the AMS measurement of $^{226,228}\text{Ra}$ in environmental samples possible over reasonable count periods. A more precise determination of the detection limit is needed, and tests of suitable calibration materials, which could also be used to determine the radium concentration, are presently underway.

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INDEX OF AUTHORS

Author	Page No.	Author	Page No.
Abbate, P.	11	Kibedl, T.	187
Acworth, I.	141	Kim, J.	144
Airey, P. L.	109	La Robina, M.	134
Akber, R.	164	Lane, G. J.	187
Aly, Z.	126	Lobanov, N. R.	193, 195
Begg, B. D.	124	Loy, J.	56
Blackburne, I.	1	Luca, V.	127
Blackford, M. G.	126	Matsumoto, K.	144
Blagojevic, N.	135	McGlinn, P. J.	124
Boldeman, J.	22	Moreton, B.	152
Bull, M. J.	183	Moricca, S.	124, 133, 134
Byrne, A. P.	173	Norman, P.	179
Cameron, R.	60	Oh, S-N.	144
Carlson, J.	72	Orchison, K.	91
Carter, M. L.	124	Ordoñez, J. P.	11
Chambers, S.	139, 144, 154	Peck, G. A.	152
Chaplin, D. H.	173	Perera, D. S.	124, 126
Child, D.	78	Perkins, C.	42
Cleary, R.	36	Poon, S.	144
Coster, H.	6	Ripley, M. I.	117
Craufurd-Hill, J.	135	Robotham, R.	32
Day, R. A.	124	Rosenfeld, A.	135
Dickson, C. L.	126	Ryan, T.	91
Doering, C.	164	Scales, N.	127
Dowling, K.	168	Seaborne, G.	21
Dracoulis, G. D.	183	Shrestha, S. K.	173
Dussol, R. J.	94	Sisoutham, O.	139, 154
Eddowes, R. C.	127	Smith, K. L.	124
Eddowes, T.	133	Stewart, M. W. A.	124
Edge, A. V. J.	173	Stone, D.	141
Ferris, J.	148	Szymczak, R.	152
Fifield, L. K.	197	Thomas, B.	124
Finlay, M. R.	117	Thomson, P.	82
Garea, V.	66	Timmers, H.	173
Garnett, D.	168	Tims, S. G.	197
Garrett, R.	21	Trautman, R. L.	126
Goossens, D. J.	183	Tuniz, C.	77, 109
Griffith, C. S.	127	Uematsu, M.	144
Hardy, C. J.	47, 101	Vance, E. R.	124, 126
Harries, J.	148	Waldron, H.	168
Harrison, J.	148	Walls, P. A.	124
Harvey, G.	168	Wang, T.	144
Heerdegen, A. P.	183	Webb, N.	133
Hobbs, R.	21	Weisser, D. C.	193, 195
Hoffmann, E.	148	Welberry, T. R.	183
Hotchkis, M.	78	Werczynski, S.	139, 144, 154
Hughes, C.	141	West, G.	32
Hutchison, W. D.	173	Whitbourn, G.	18
Jackson, A.	21	Willers, A.	66
Jeffree, R. A.	152	Williams, M.	78
Kang, C. H.	144	Zahorowski, W.	139, 144, 154
Keegan, E. A.	127	Zaplatin, E.	195
Kelly, J.	86	Zhang, Y.	124
Kemeny, L. G.	111, 158		