Australian Government

Ansto

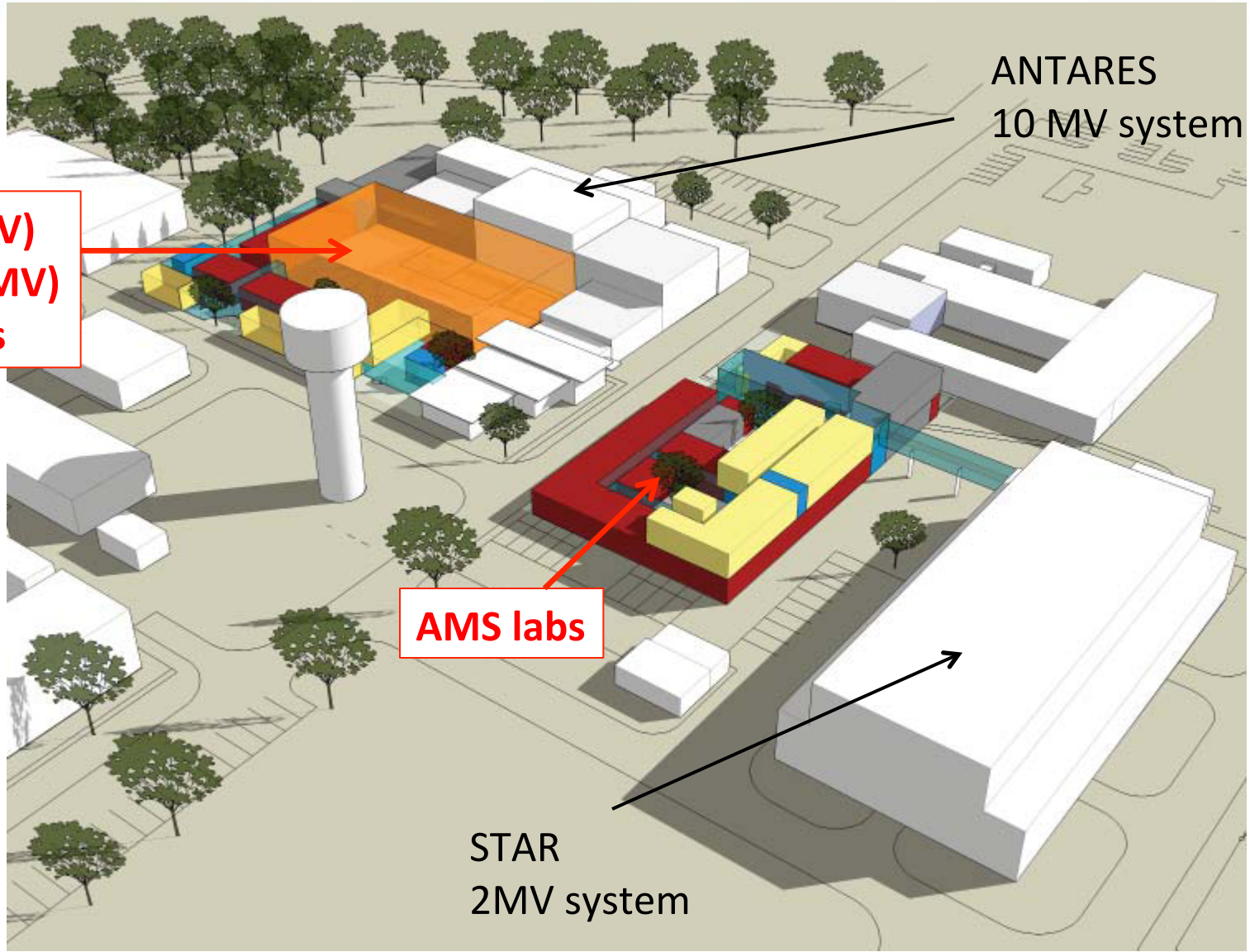
Use of AMS in research on fallout plutonium and uranium from nuclear tests

M.A.C. Hotchkis and D.P. Child
ANSTO, Sydney, NSW, Australia

ANSTO's Lucas Heights site in Sydney

→ home of the OPAL Research Reactor, the Centre for Accelerator Science, and a range of other nuclear science facilities

Centre for Accelerator Science



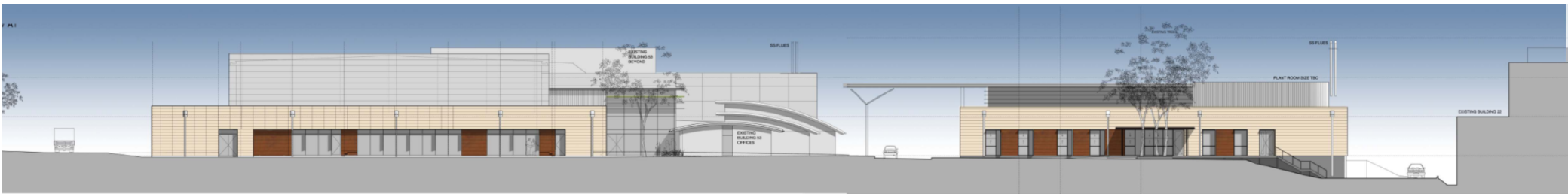
CAS AMS sample prep labs
and mass spec clean labs

VEGA accelerator

Talk outline

- AMS and performance of new VEGA AMS system
- Global fallout from nuclear weapons tests
- Applications of actinides in the environment
- Fallout in Australia

ANSTO's Centre for Accelerator Science, Lucas Heights, Sydney

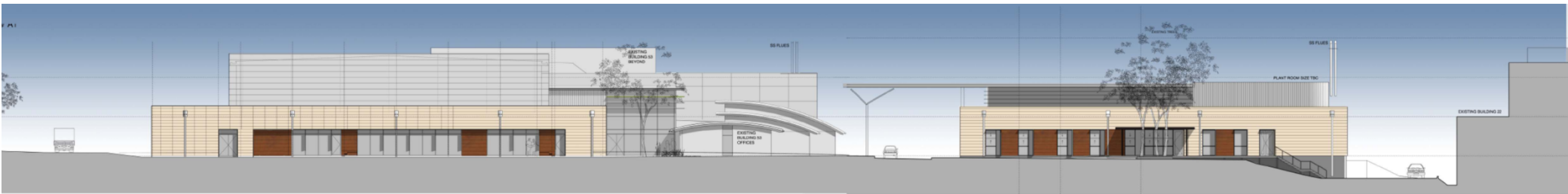


Accelerator Mass Spectrometry

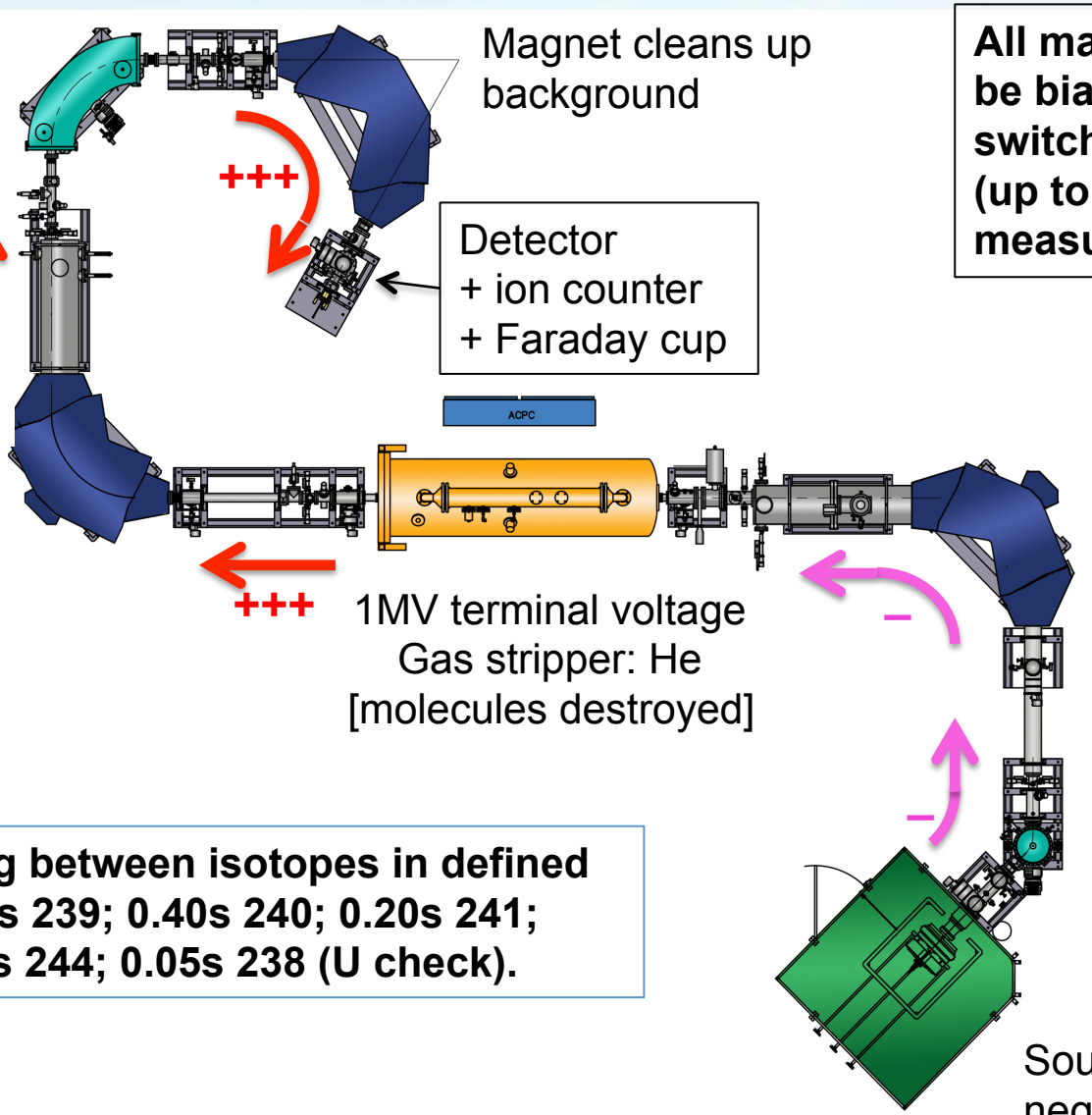
Ultra-sensitive detection of long-lived radioisotopes

- useful for ^{14}C, ^{10}Be, ^{26}Al, ^{36}Cl, ^{129}I, U, Pu, etc
- cosmogenic and anthropogenic radioisotopes
- various environmental science applications
- U and Pu isotopic analysis for IAEA nuclear safeguards (monitoring compliance with the Non-Proliferation Treaty), for contamination monitoring, and for radioecology studies

ANSTO's Centre for Accelerator Science, Lucas Heights, Sydney



VEGA 1MV AMS system: actinides example

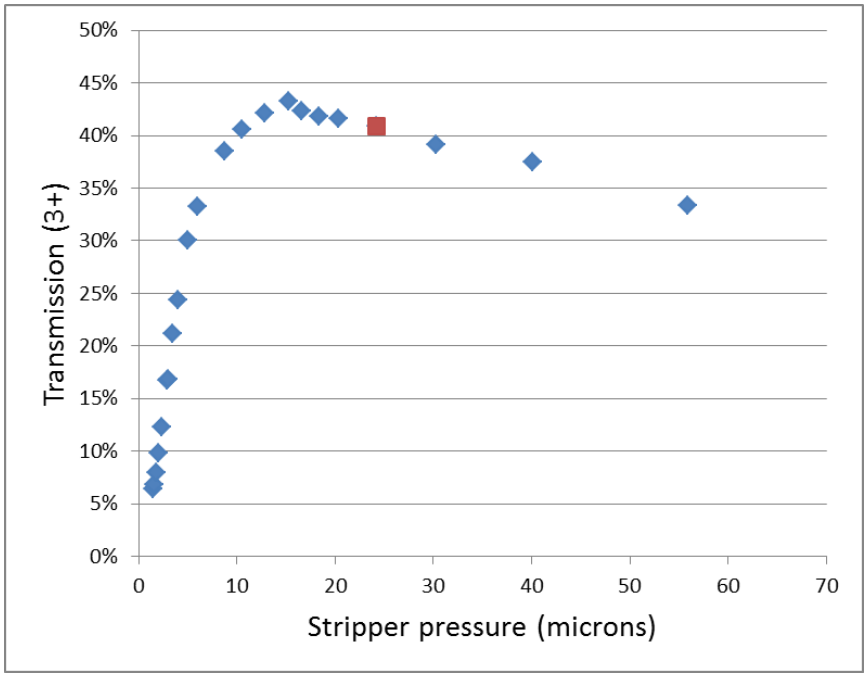


All magnet chambers can be biased allowing fast switching between isotopes (up to 8) during same measurement sequence.

Rapid switching between isotopes in defined sequence: 0.10s 239; 0.40s 240; 0.20s 241; 0.10s 242; 0.10s 244; 0.05s 238 (U check).

Actinides AMS – results for yields (thorium beam)

Injected ion	Voltage	Charge state	Gas stripper	Transmission
ThO^-	1.0 MV	3+	Argon	17 %
ThO^-	1.0 MV	3+	Helium	39 %
ThO^-	0.68 MV	3+	Helium	43 %
ThO^-	0.68 MV	3+	Helium	41 % *



Transmission as a function of stripper pressure at 0.68MV for Thorium (ThO^- injected ion).

- *indicates the pressure required to ensure destruction of molecular interferences that affect ^{236}U and ^{240}Pu detection.*

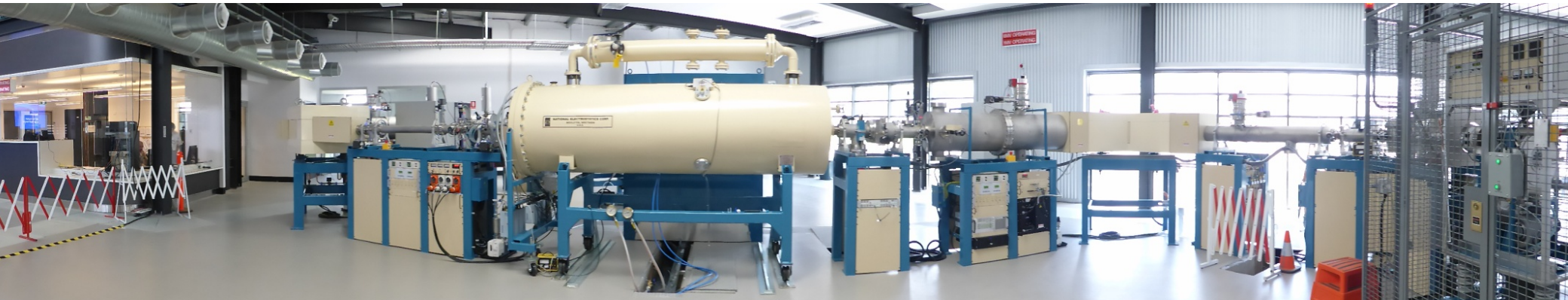
Actinides AMS – abundance sensitivity

Abundance sensitivity and isotope ratio sensitivity limits so far established for the VEGA AMS system.

Note: to avoid ion source memory effects, VEGA ion source used for samples with U masses <10µg U.

Isotope or species ratio	Sensitivity limit
$^{233}U/^{232}Th$	2.7×10^{-9}
$^{236}U/^{235}U$	1.8×10^{-9}
$^{239}Pu/^{238}U$	1.3×10^{-9}
$^{240}Pu/^{238}U$	5.0×10^{-10}
$^{233}U/^{238}U$	1.4×10^{-12}
$^{236}U/^{238}U$	1.3×10^{-11}

VEGA - the 1MV AMS system at ANSTO



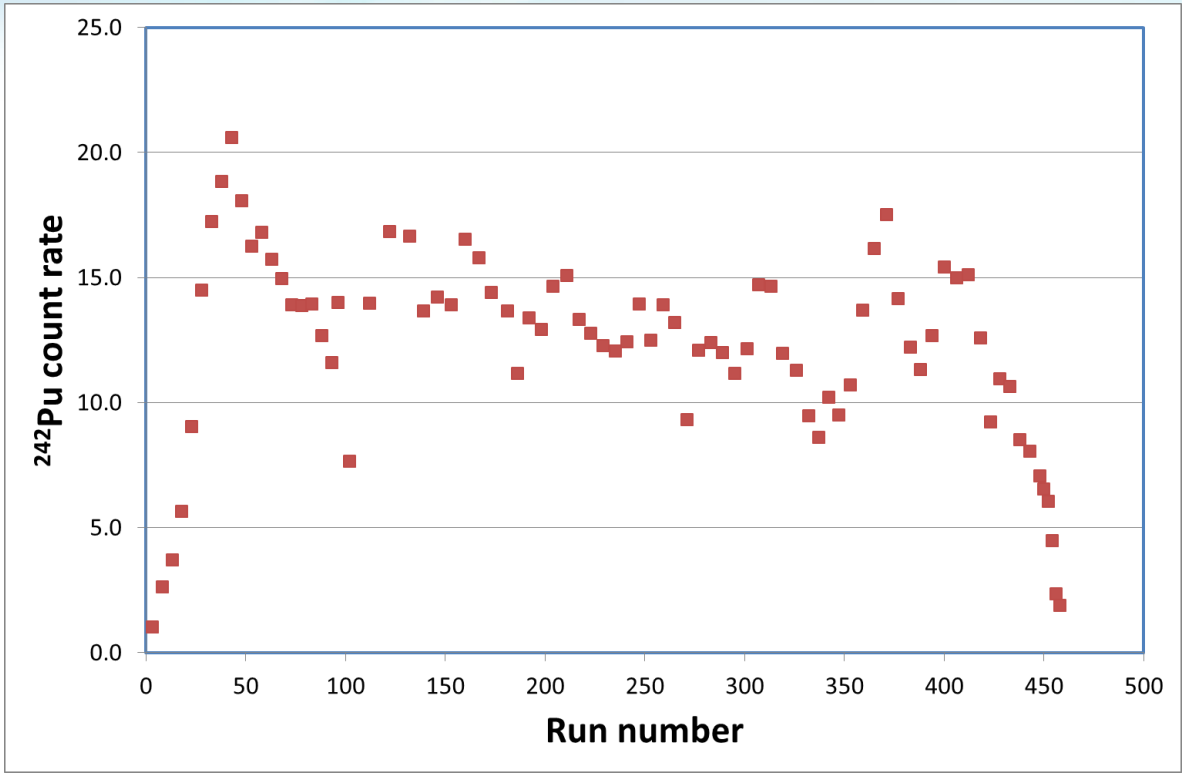
Actinides AMS – absolute sensitivity

Absolute sensitivity

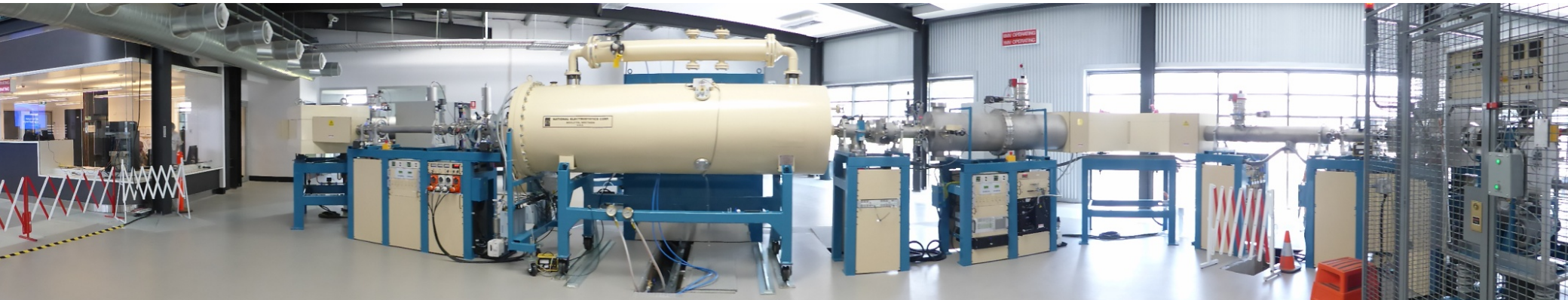
^{242}Pu sample run to exhaustion (16fg ^{242}Pu in 7mg Fe_2O_3)

Preliminary results:

- *overall efficiency > 0.5%*
- *ion source efficiency > 1%*
- *^{244}Pu sensitivity <0.1ag (<1x10^{-19}g)*

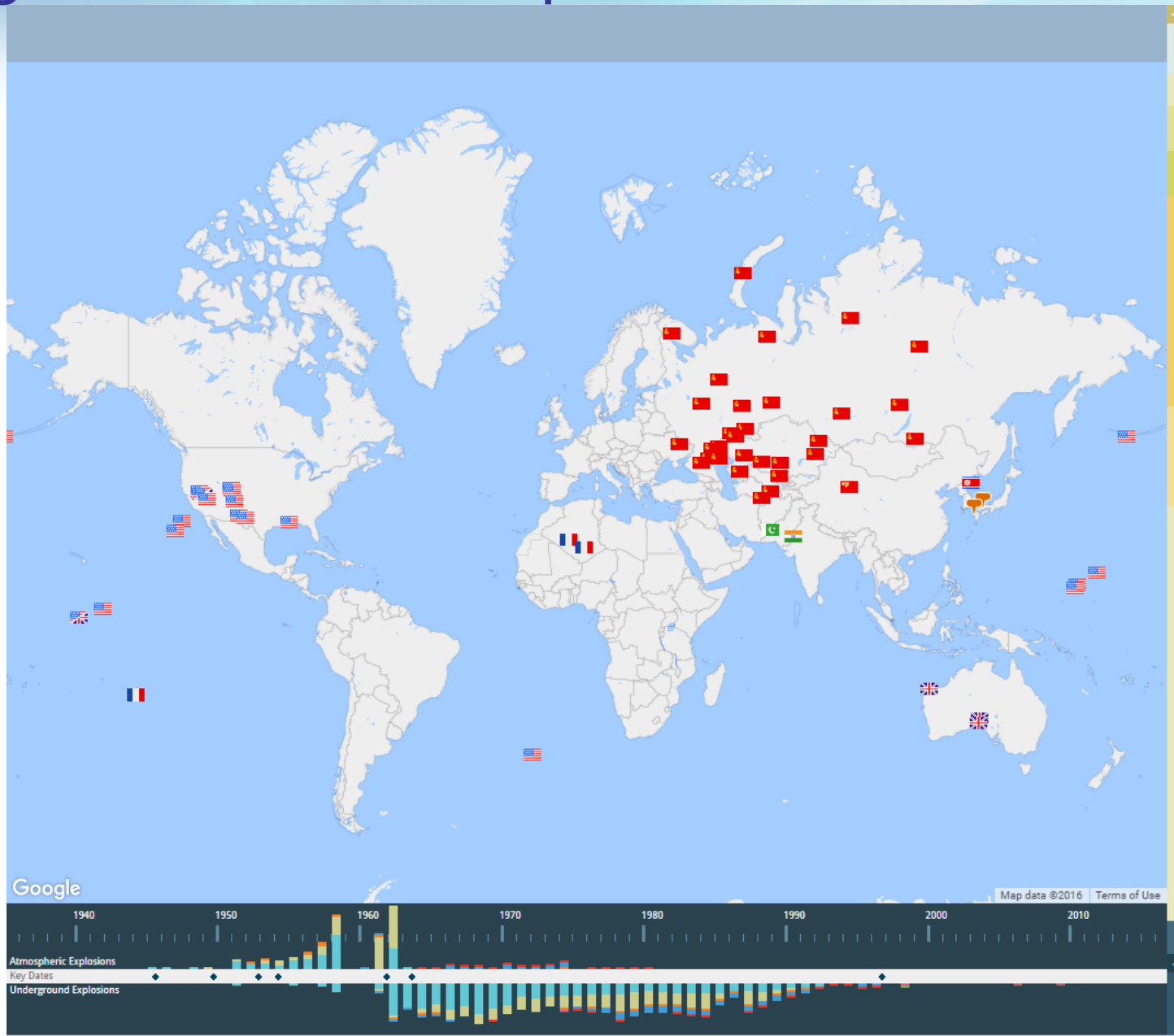


VEGA - the 1MV AMS system at ANSTO



Above-ground nuclear explosions

530 nuclear explosions above ground



Source: www.ctbto.org

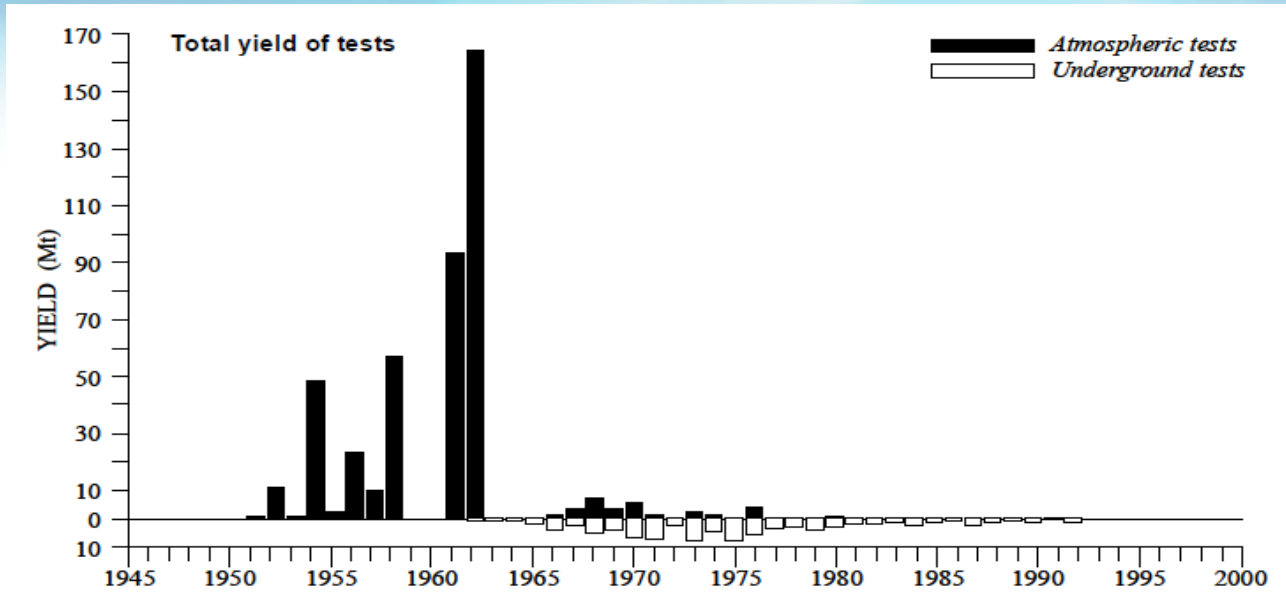
Nuclear tests

Globally:

530 above-ground tests

440 Mt explosive

~6t of Pu released



UNSCEAR 2000 Annexe C

~1pg/cm^2 across Earth's surface

Recent data indicates ~1t of ^{236}U released

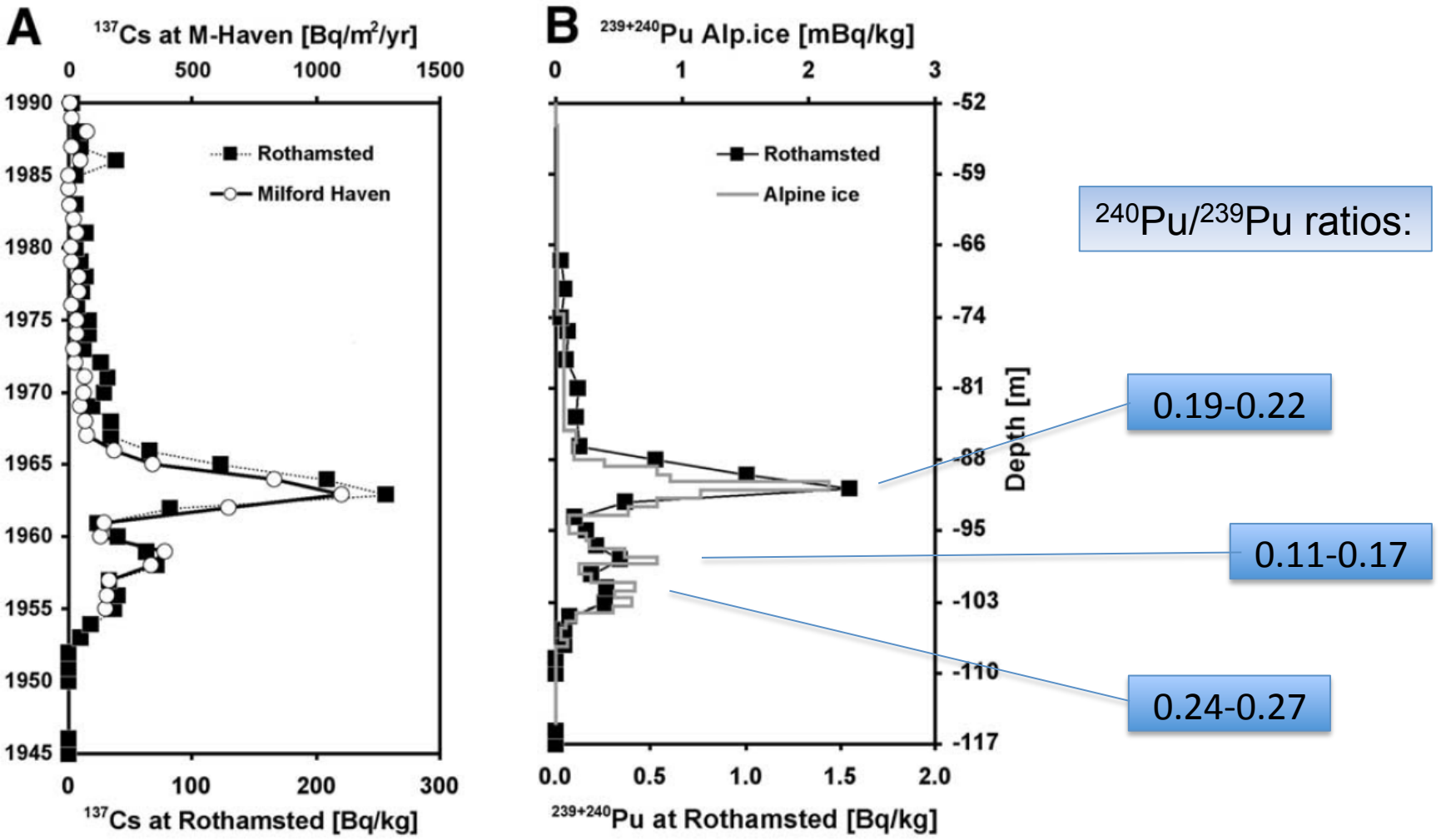
The majority of the 'global' fallout was produced in a relatively short period of time prior to the Limited Test Ban Treaty (1963)

^{240}Pu/^{239}Pu ratio: mean global value 0.18 (Kelley et al., 1999)

Plutonium in fallout

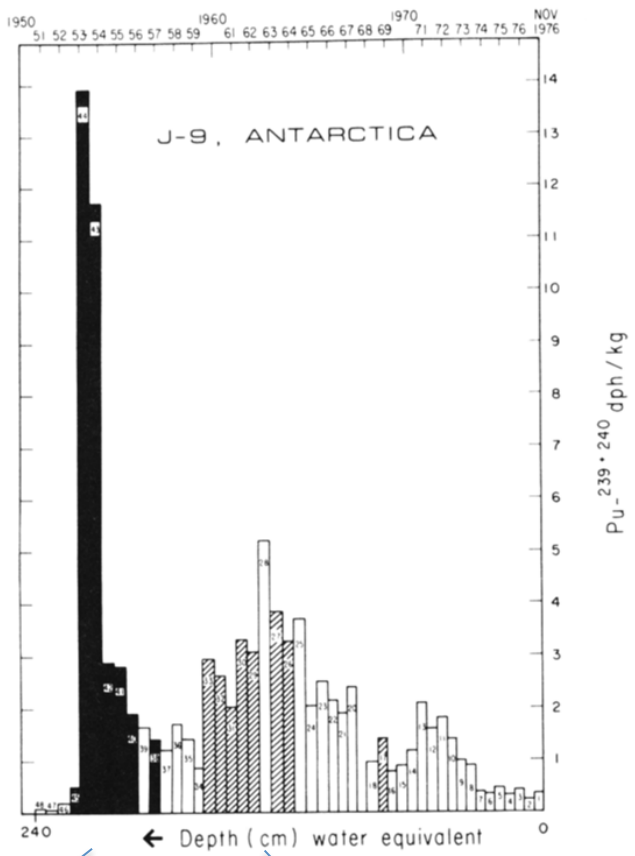
Pu fallout records from archived herbage (UK) and Alpine ice

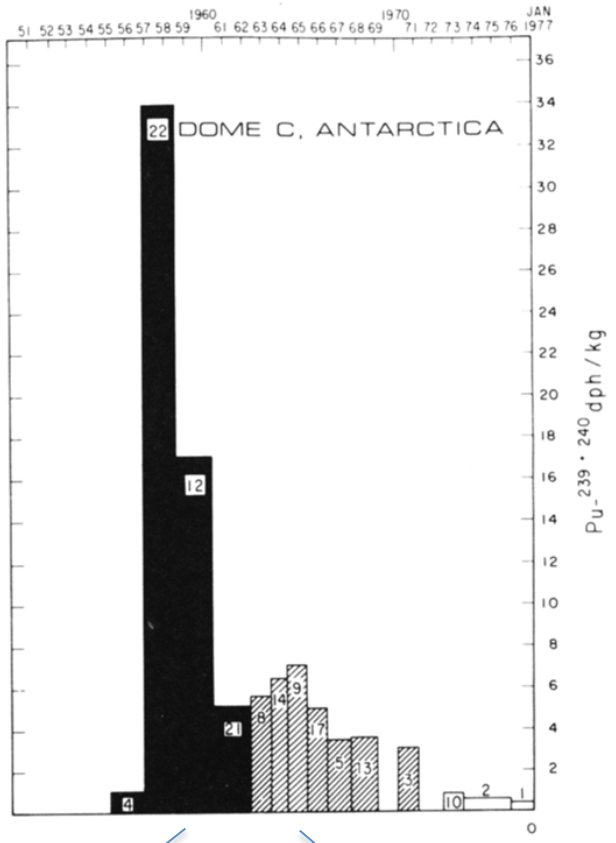
T. Warneke et al. / Earth and Planetary Science Letters 203 (2002) 1047–1057



Plutonium in fallout

Pu fallout records from ice cores (Antarctica)





$^{240}Pu/^{239}Pu$ ratios:

0.33-0.34

0.16-0.19

0.23-0.34

0.09-0.18

Koide et al., 1985.

Tests by UK in Australia

3 tests at Monte Bello Islands, Western Australia, early 1950s

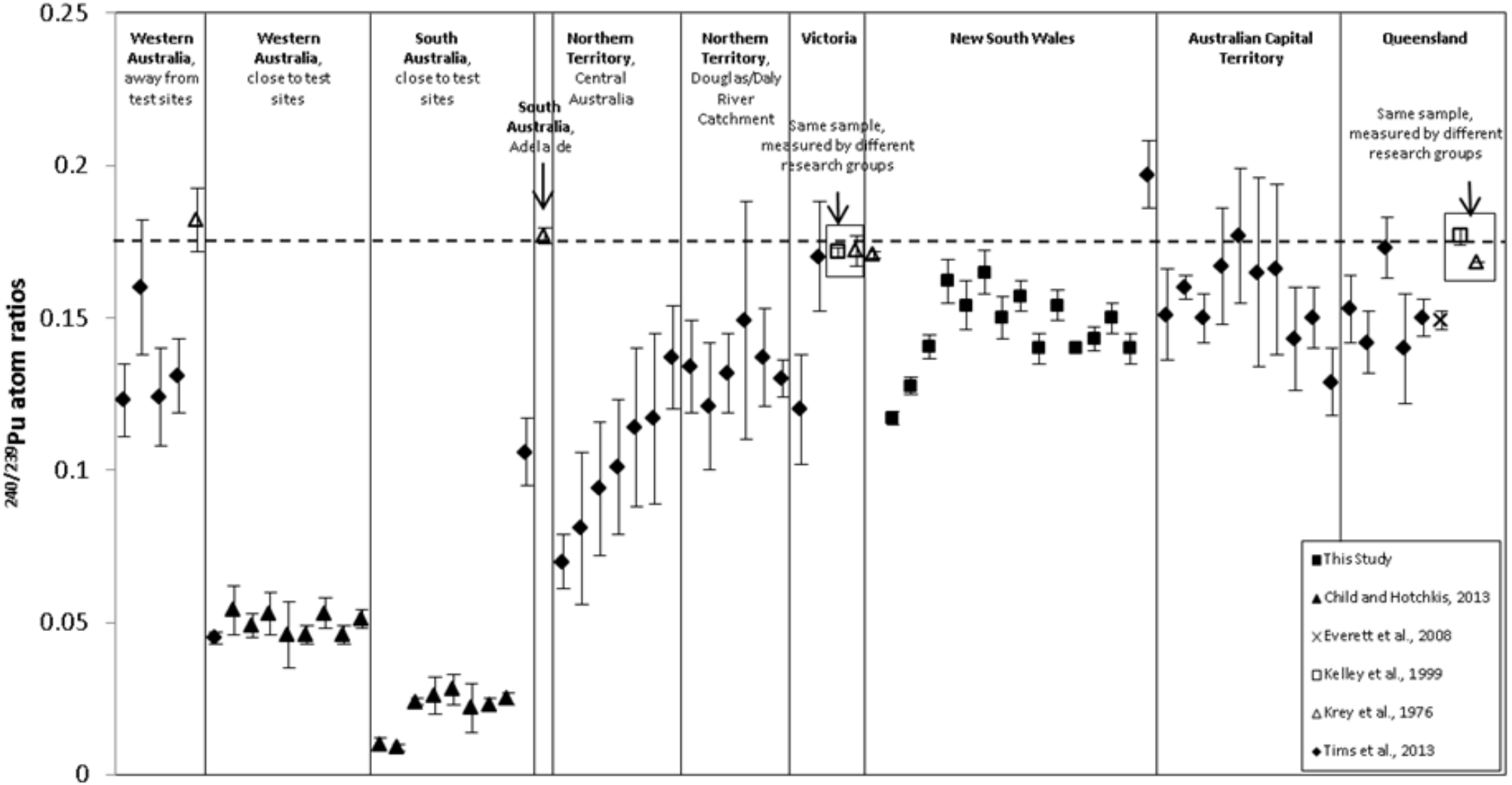
7 nuclear tests at the Maralinga test site, 2 at Emu, South Australia, 1950s to early 1960s

- series of ‘safety trials’ involving conventional explosives and about 22kg Pu at Maralinga

Plutonium fallout across Australia

240/239 isotope ratios

B. Smith et al., 2016.

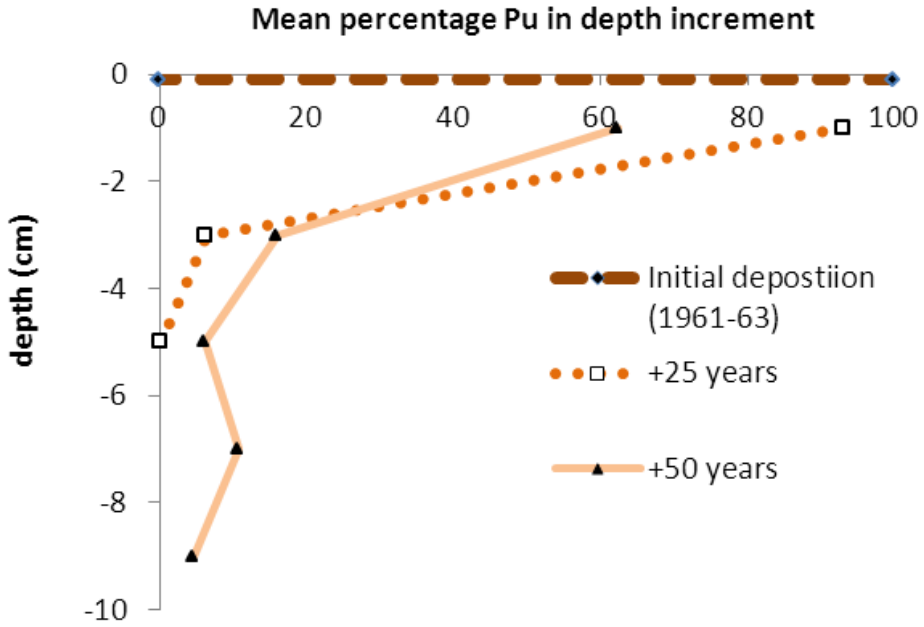


Pu at Maralinga test site

We have measured U and Pu isotopics in soils and biota

Soil profiles can be compared to earlier data to learn about migration through the soil

M.P. Johansen et al. / Journal of Environmental Radioactivity 131 (2014) 72-80

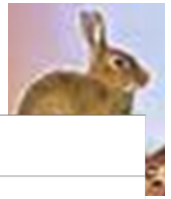


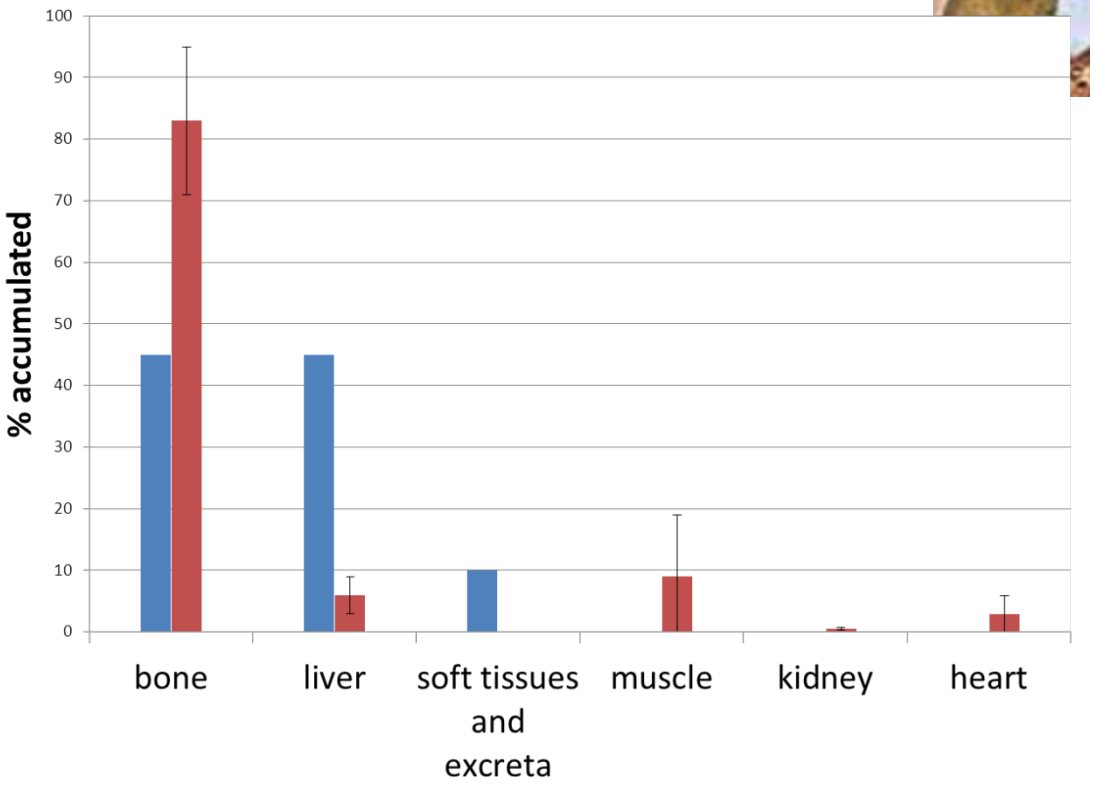
Pu at Maralinga test site

Distribution in mammalian organs:

- bone vs. liver distribution contrasts with existing mammal data
- related to physico-chemical form of Pu and ingestion route

Denise Greig © Australian Museum





Percentage burdens of $^{239,240}Pu$ in tissues of Taranaki wildlife mammals ( ) compared with ICRP 19 and ICRP 30 values ( ) (recommended for human worker protection). n=7 for Taranaki mammals.

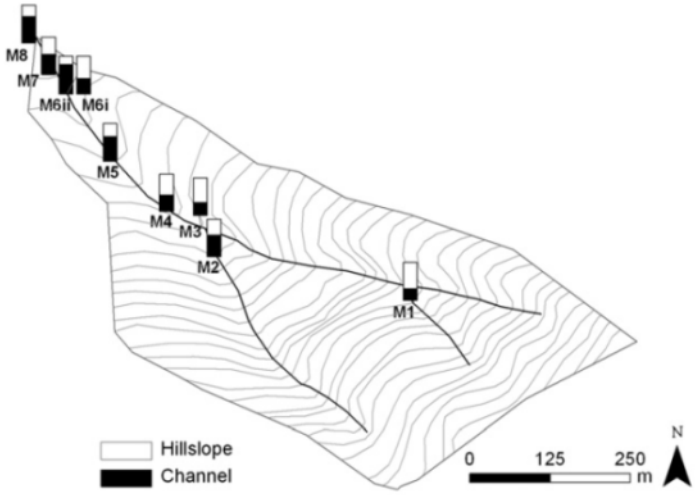
M.P. Johansen et al. 2016

Applications – plutonium AMS

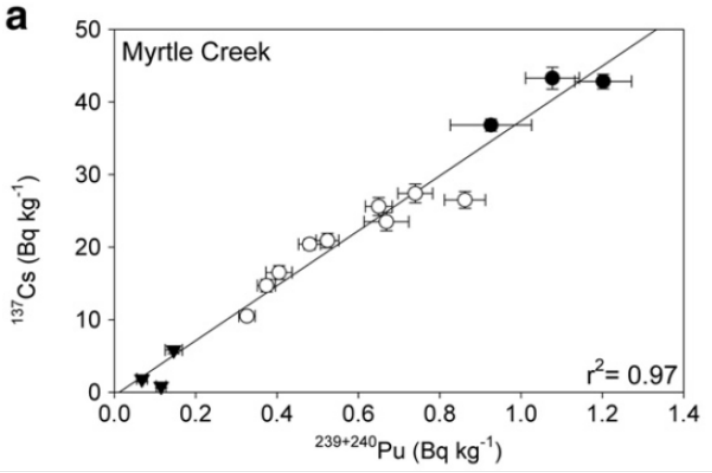
Soil erosion following bushfires + intense rain events

H.G. Smith et al., Quantifying sources of fine sediment supplied to post-fire debris flows using fallout radionuclide tracers, Geomorphology 139–140, 403-15 (2012).

Contour map of the Myrtle Creek catchment. Sampled sediment deposition sites are shown, with the relative contribution from hillslope and channel bank source contributions.



Correlation between ^{137}Cs and Pu



Myrtle Creek and some sampling sites

Applications – plutonium AMS

Understanding soil erosion and sedimentation through source apportionment

Hobgen, S.E., Myers, B.A., Fisher, R.P. and Wasson, R.J., 2014. Creating a sediment budget in a data poor context: an example from eastern Indonesia. Geografiska Annaler: Series A, Physical Geography, 96, 513–530. doi:10.1111/geoa.12076

Hobgen et al., to be published.

Location: Indonesian island of Sumba

Fig. 2. Landscape and vegetation of Kambaniru River catchment, East Sumba. Photograph: B. Myers.

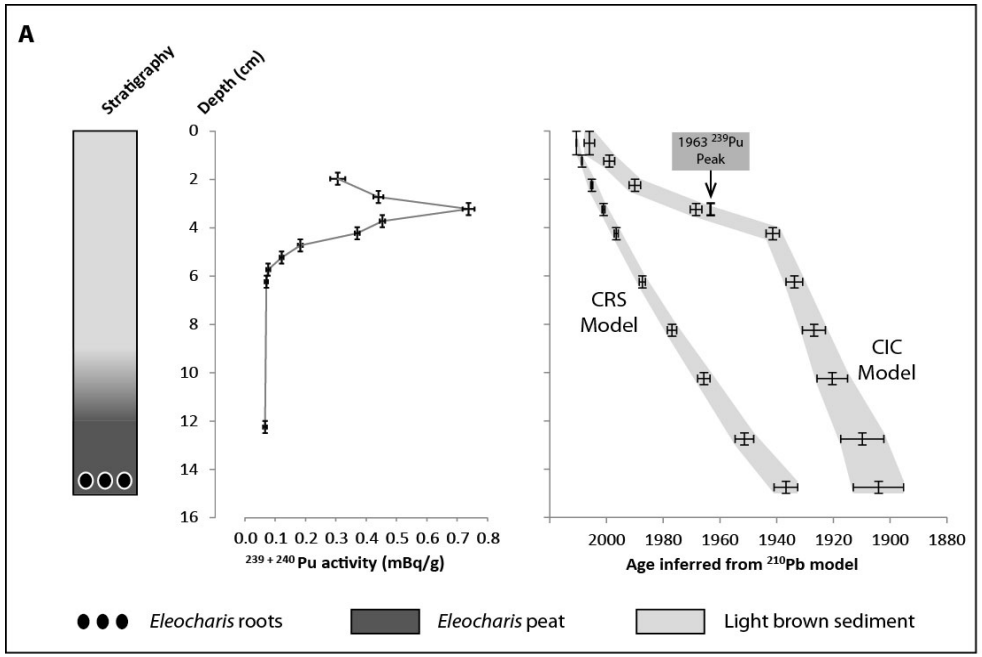
Table 5. Mean concentrations of radionuclide tracers in the <63 µm fraction of surface and subsurface soil samples.

	^{137}Cs (Bq kg^{-1})	^{239}Pu (fg g^{-1})	^{210}Pb$_{(ex)}$ (Bq kg^{-1})
Surface soils	1.9 ± 1.4 (n = 27)	35.8 ± 33.8 (n = 18)	70 ± 36 (n = 27)
Subsurface soils	<0.3 (n = 5)	7.2 ± 6.6 (n = 5)	–4 ± 12 (n = 5)
River sediments	0.9 ± 0.4 (n = 15)	14 ± 5.4 (n = 11)	43 ± 33 (n = 15)
Weir sediments	0.7 ± 0.3 (n = 2)	10.5 ± 3.0 (n = 2)	40 ± 8 (n = 2)

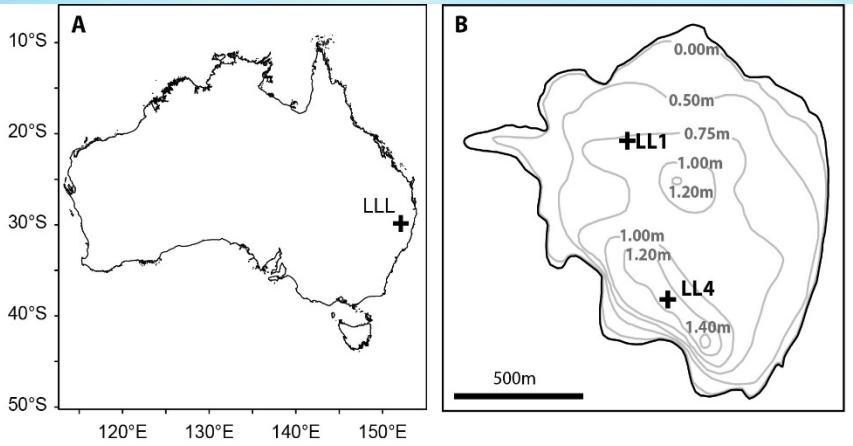
Applications – plutonium AMS

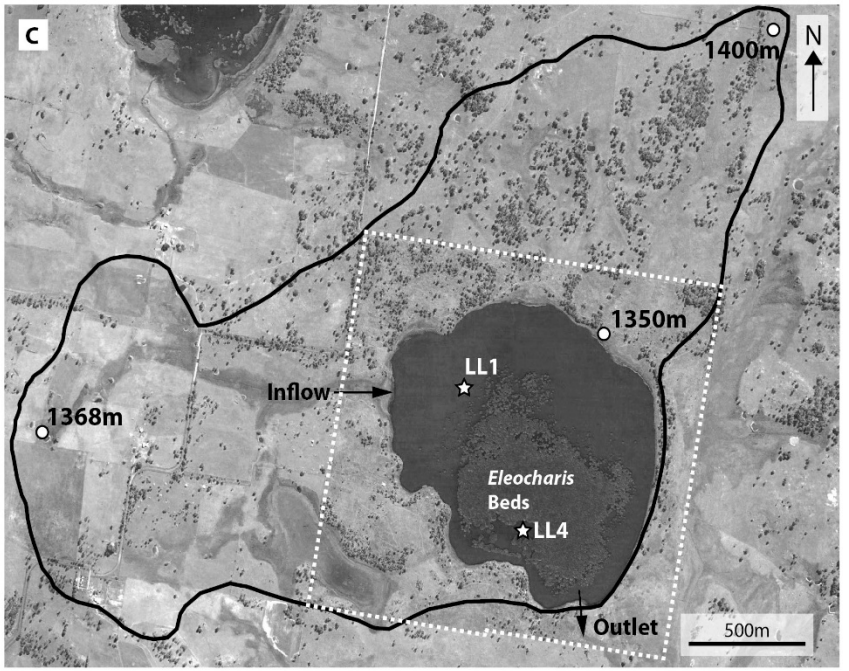
Dating recent sediments for environmental studies

C. Woodward et al., 19th century native forest clearance caused a major change in hydrological regime in a sub-humid Australian wetland (Little Llangothlin Lagoon), 2016, in press.



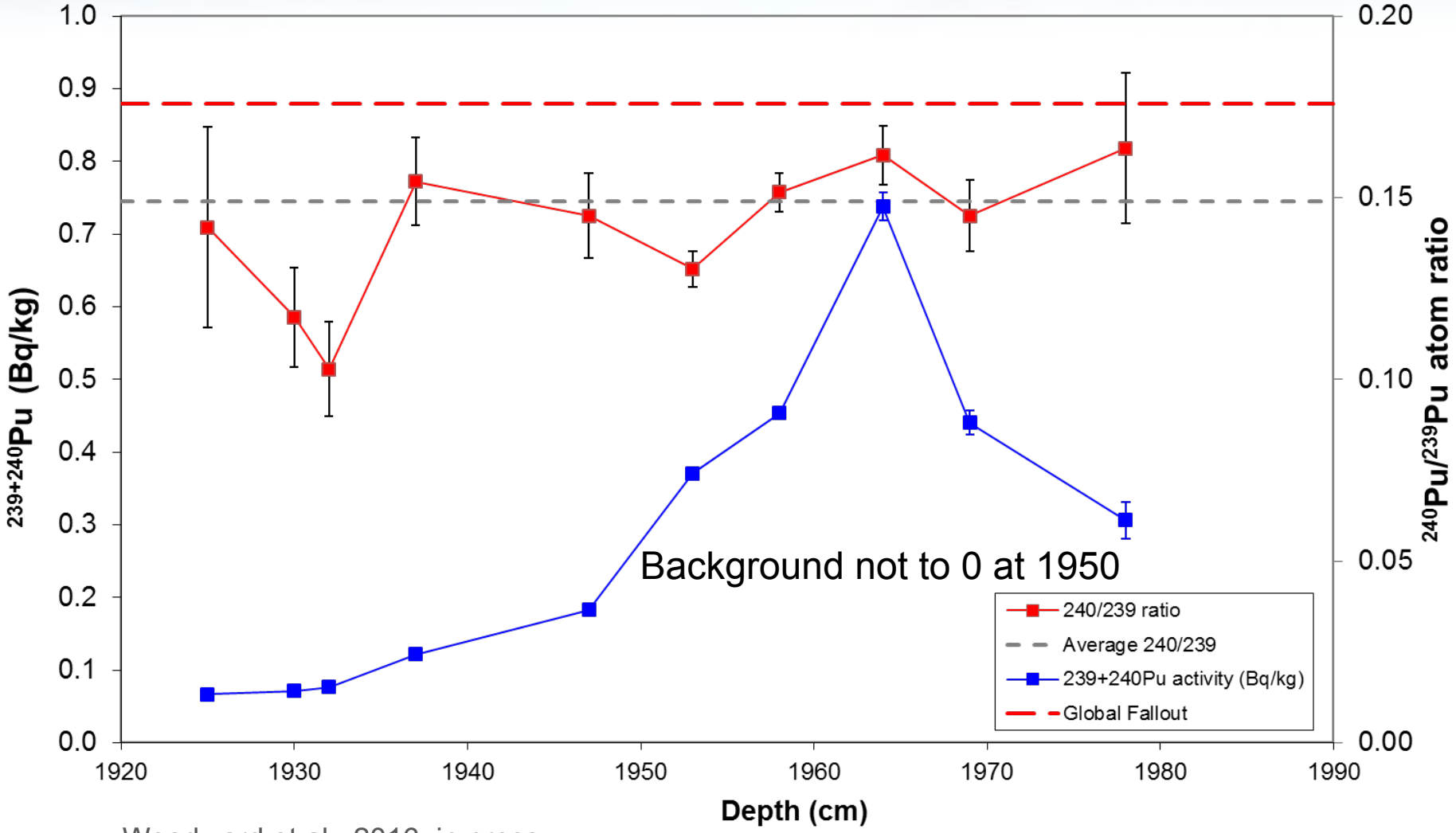
Plutonium shows well-defined bomb-pulse peak. This allows us to confirm the validity of ^{210}Pb dating of the sediment core based on constant initial concentration (CIC) model.





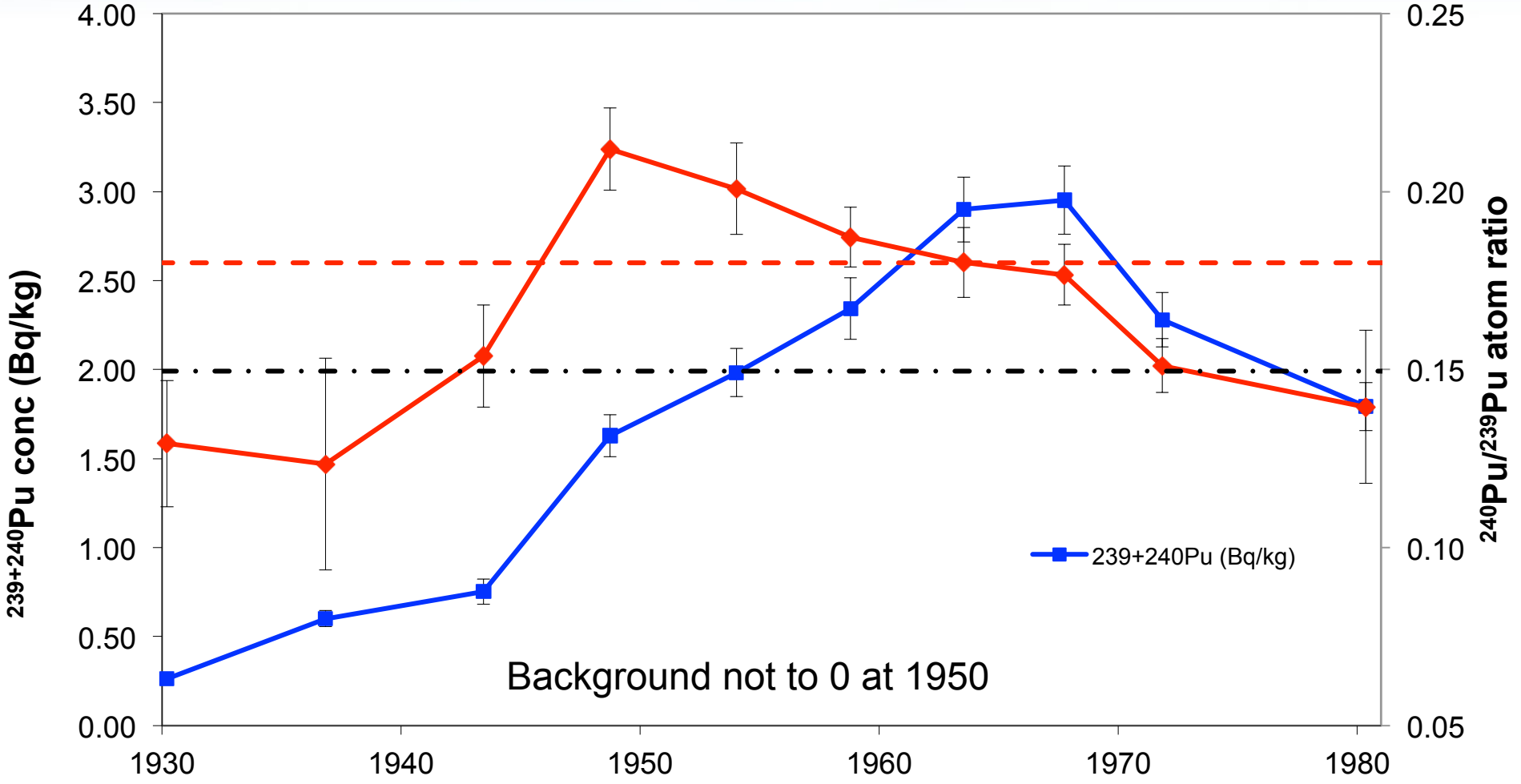
Location: northern tablelands of NSW

Llangothlin Lagoon, Northern Tablelands, NSW



Woodward et al., 2016, in press

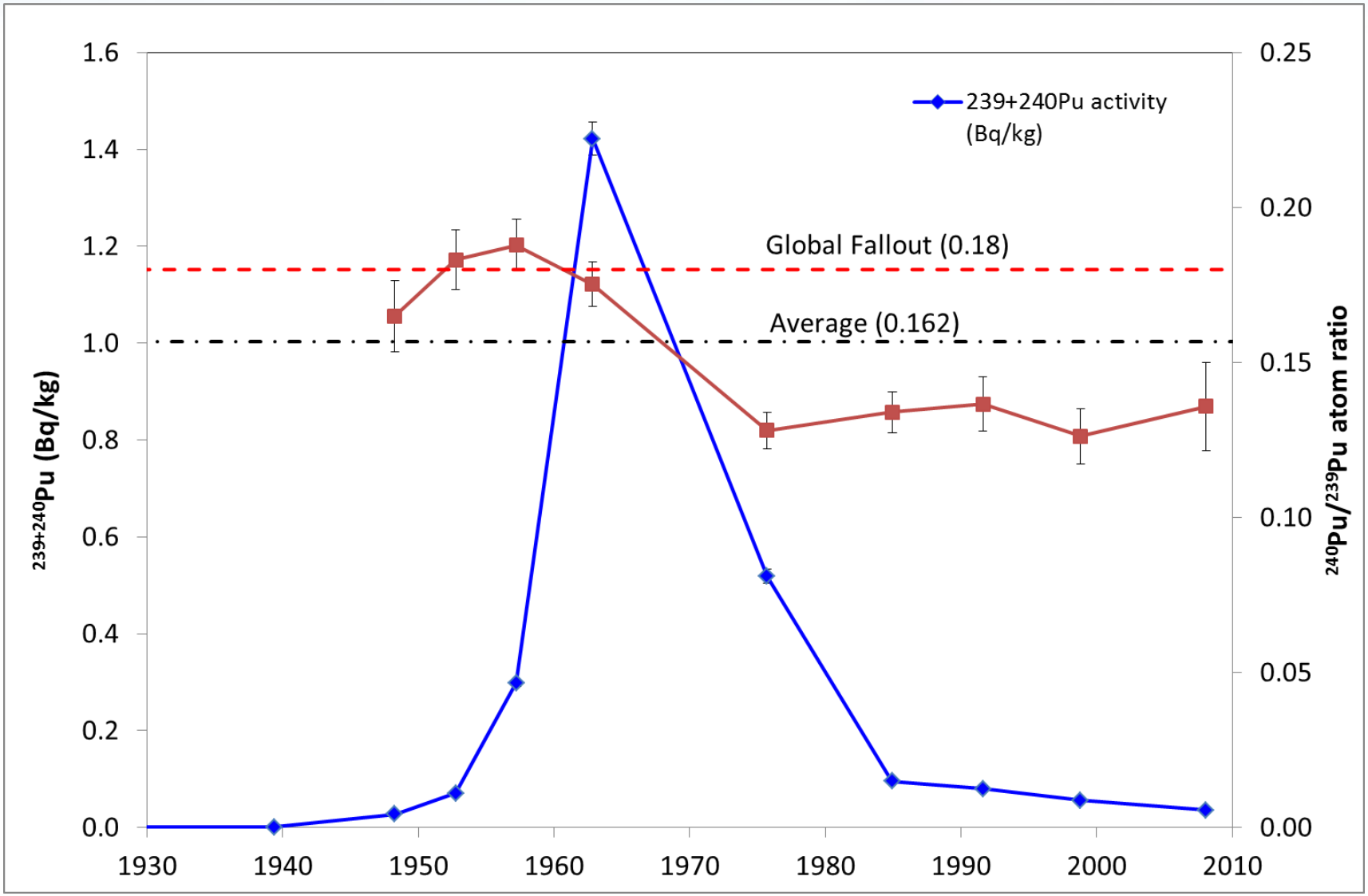
Bathurst Harbour, Tasmania



Harrison et al., to be published

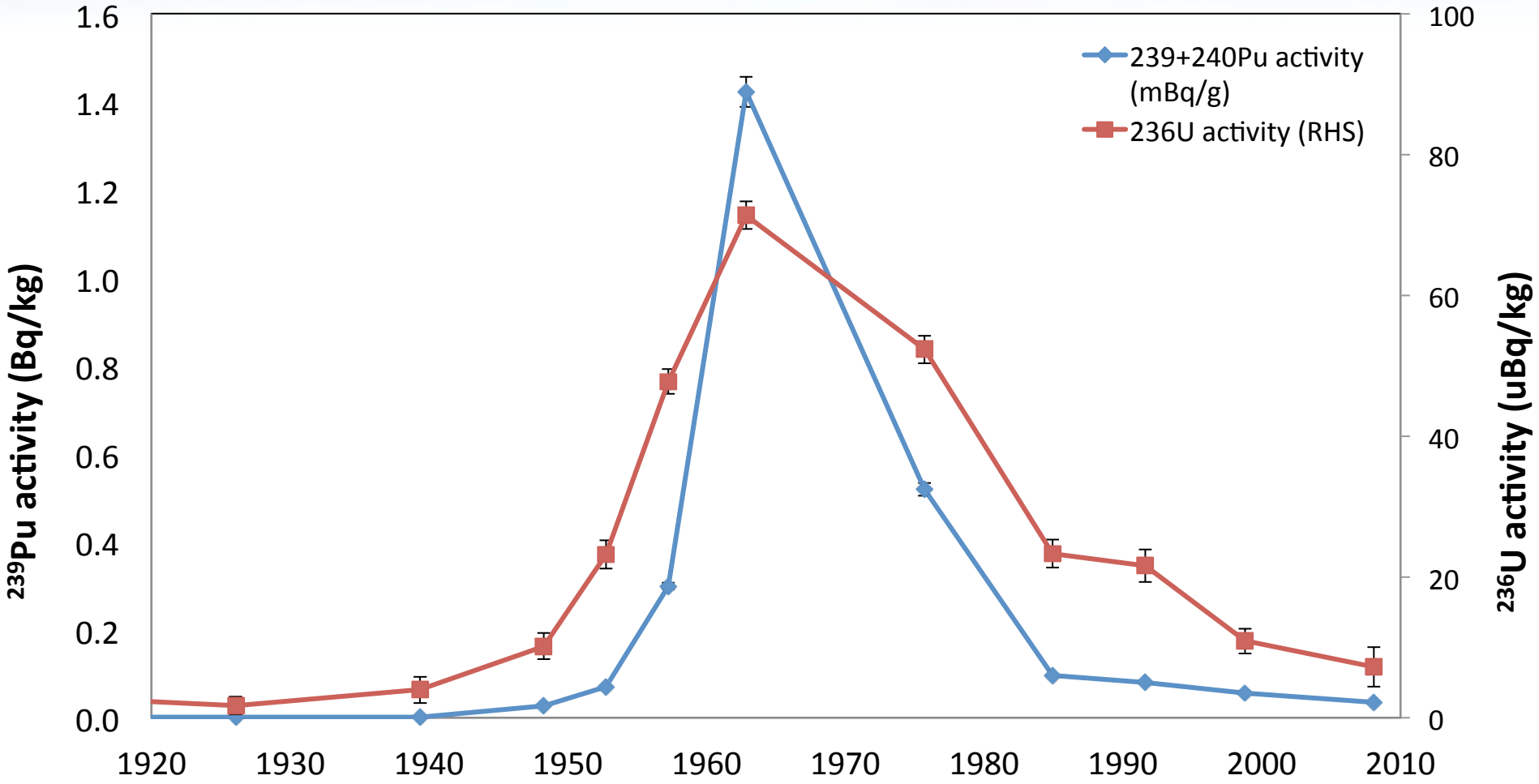
Mangrove sediment, North Coast, NSW

Jon Knight, Griffith University



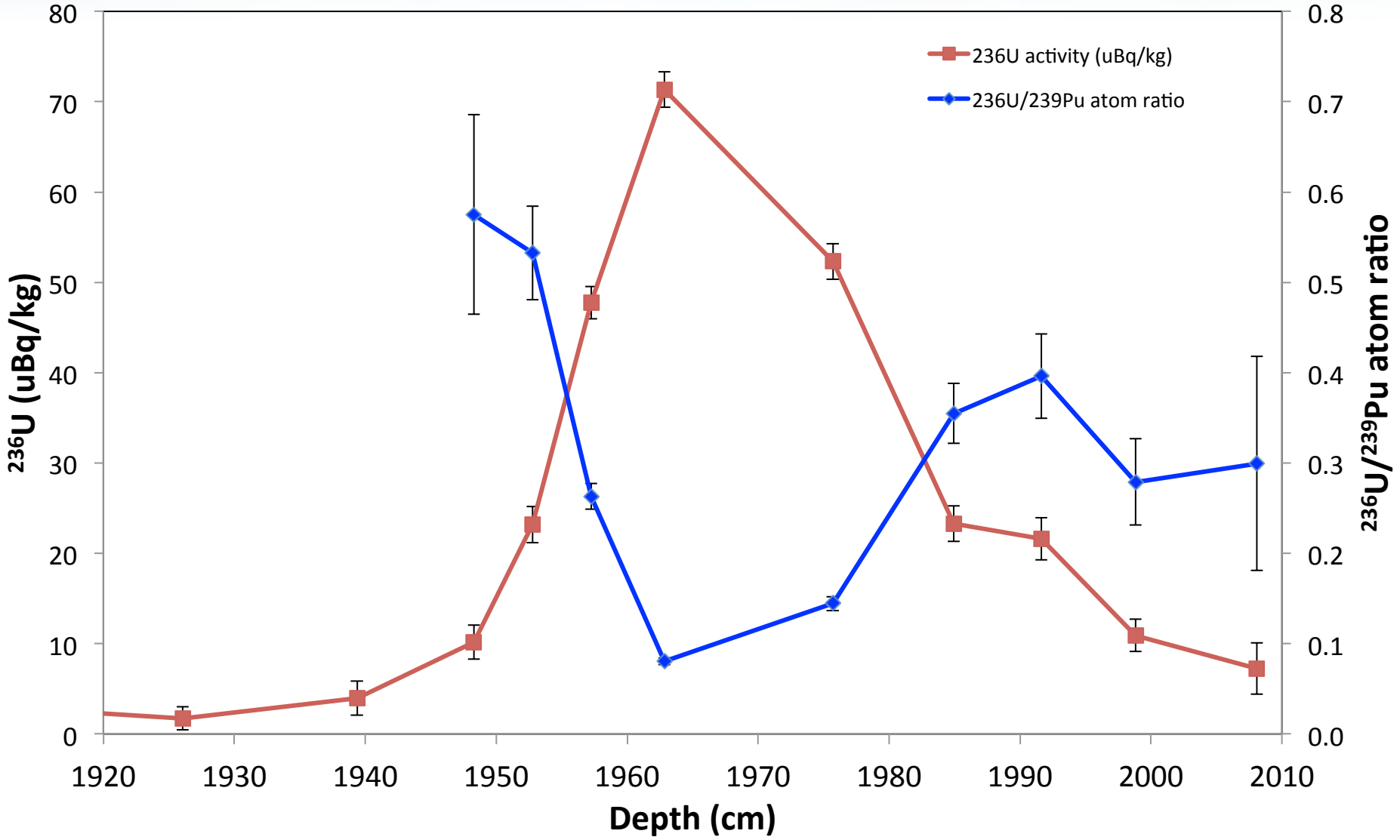
Mangrove sediment, North Coast, NSW

Jon Knight, Griffith University



Mangrove sediment, North Coast, NSW

Jon Knight, Griffith University



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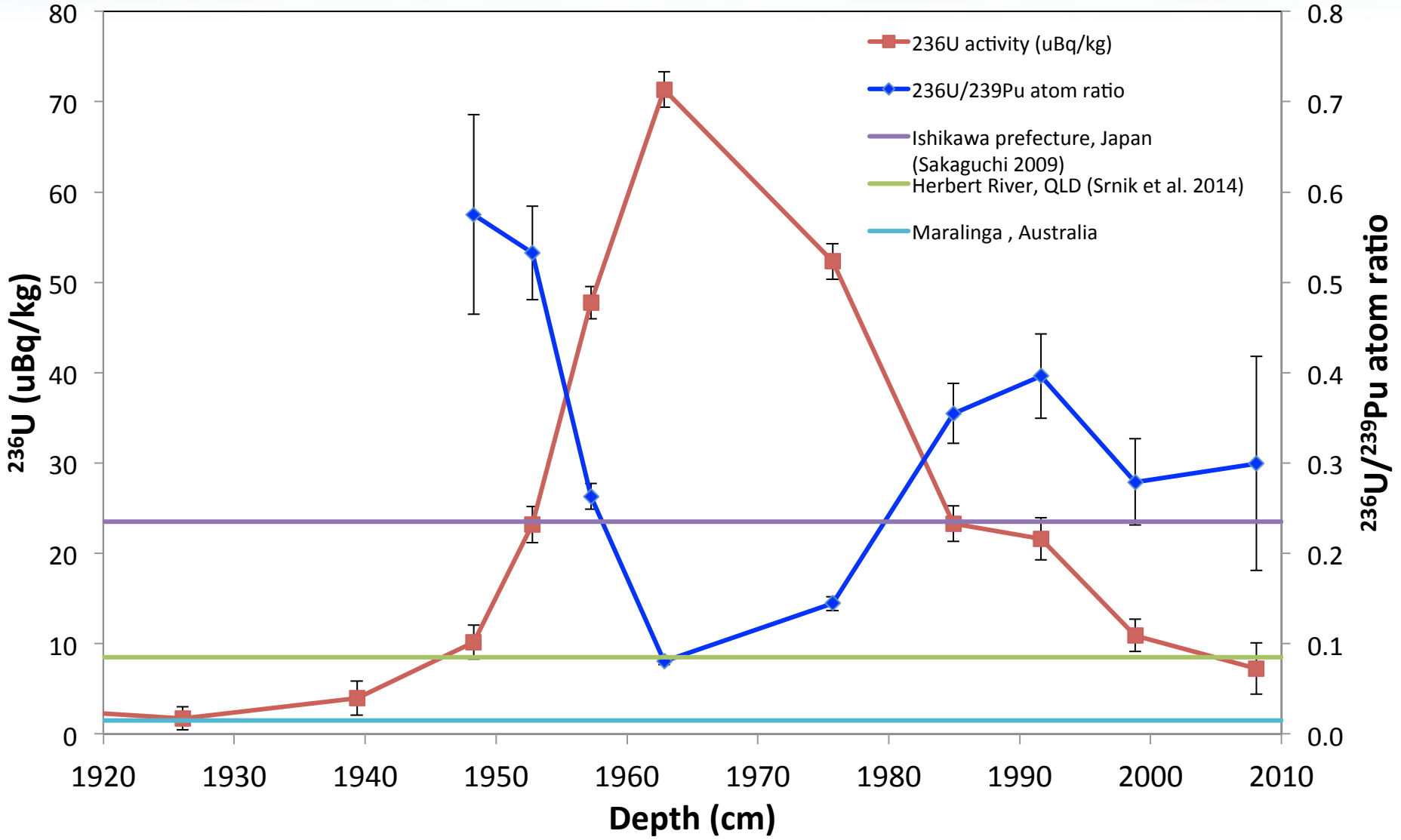
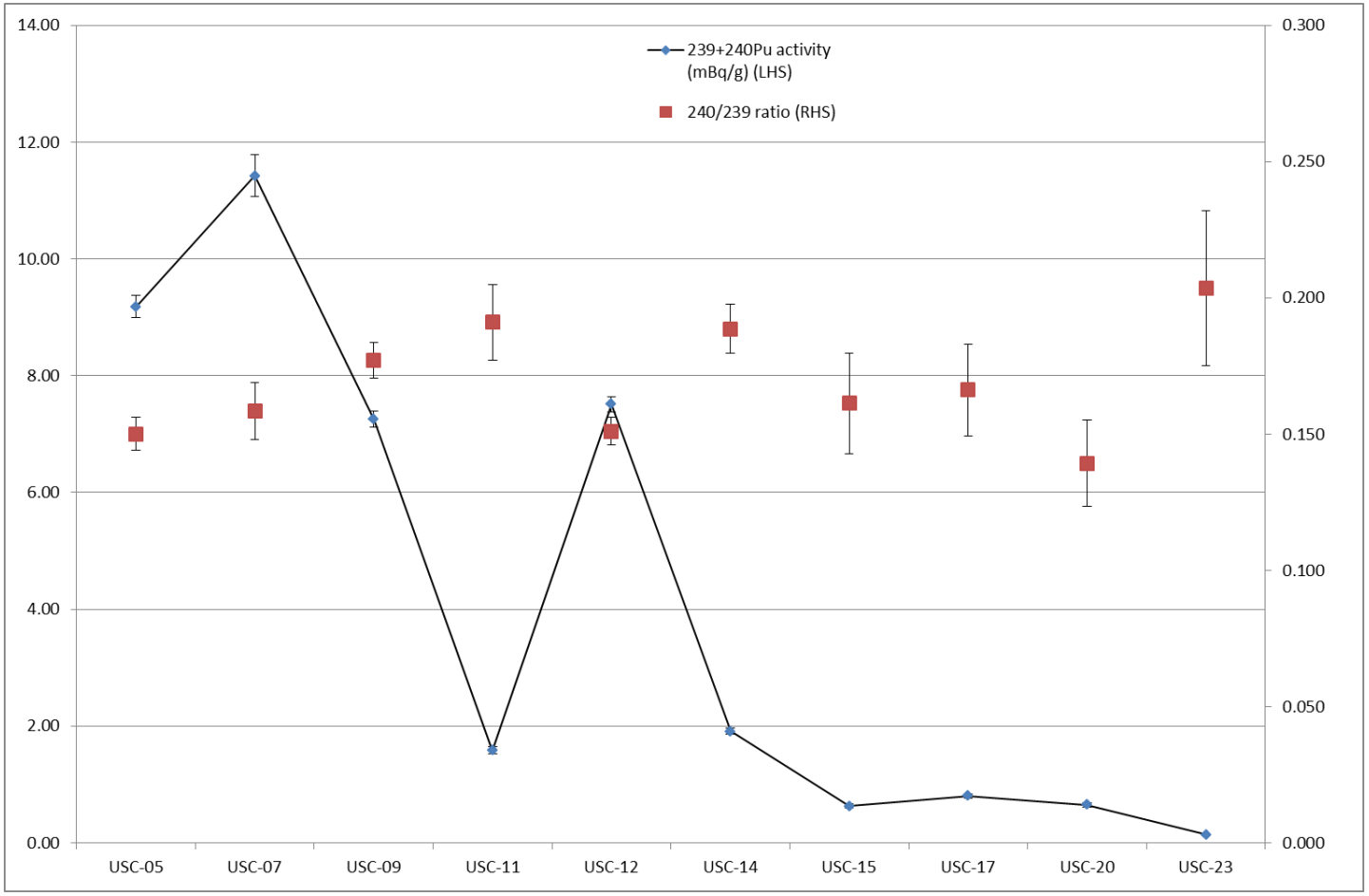


Peat core, Snowy Mts, NSW

$^{239+240}$Pu concentrations and ^{240}Pu/^{239}Pu ratios



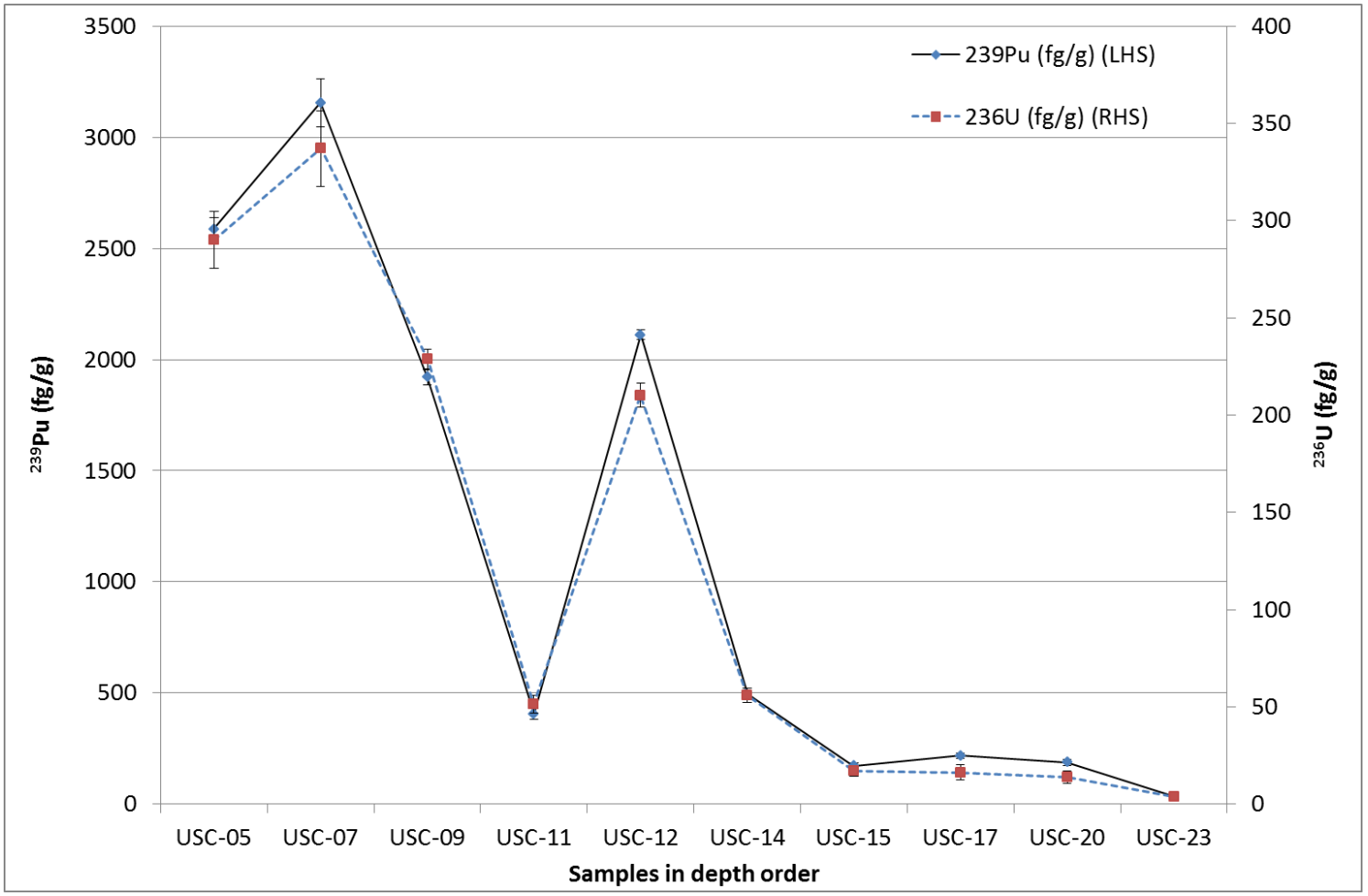
Marx et al., to be published

Peat core, Snowy Mts, NSW

^{239}Pu and ^{236}U concentrations

^{236}U/^{239}Pu ratio
0.105±0.017

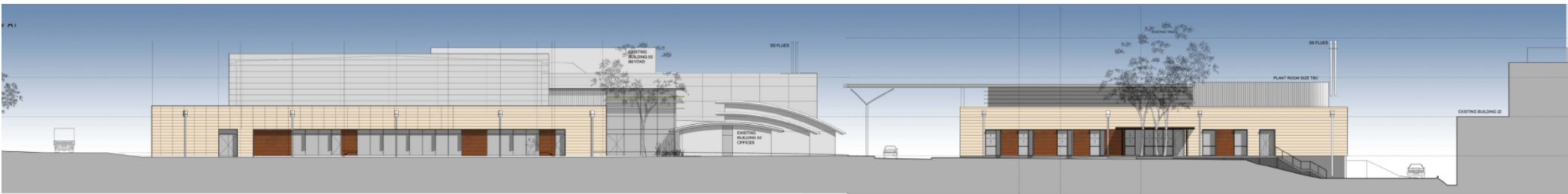
No differentiation, suggesting particles remain intact



Marx et al., to be published

Further work

- Study high-resolution records of fallout to understand influence of different test programs
- Study the form of Pu in fallout at Maralinga (for safety trials and for nuclear tests)
- Study uptake in the marine environment (Monte Bello)
- Study forms of fallout at Monte Bello islands (predominantly calcareous rocks and soils)



Acknowledgments

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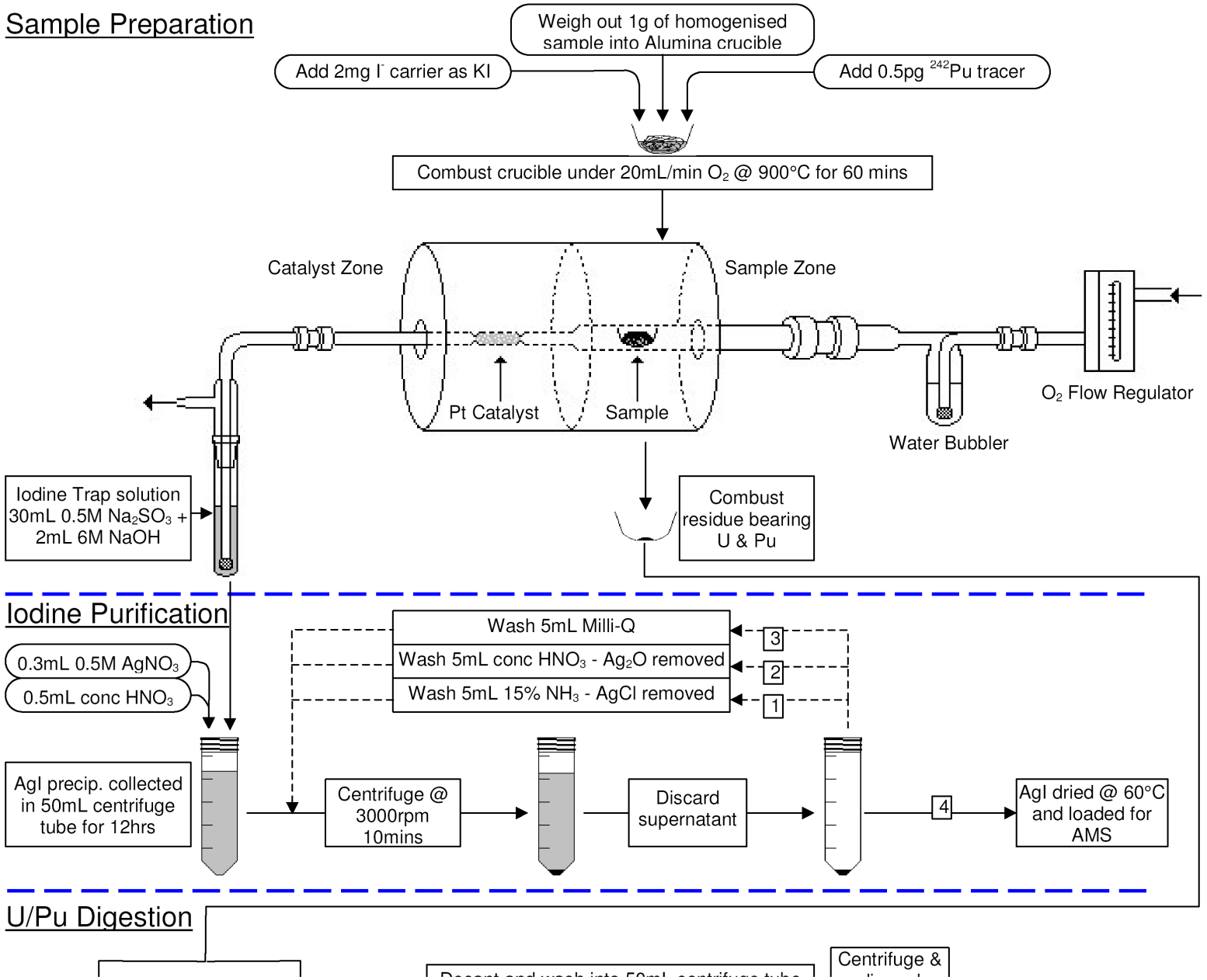
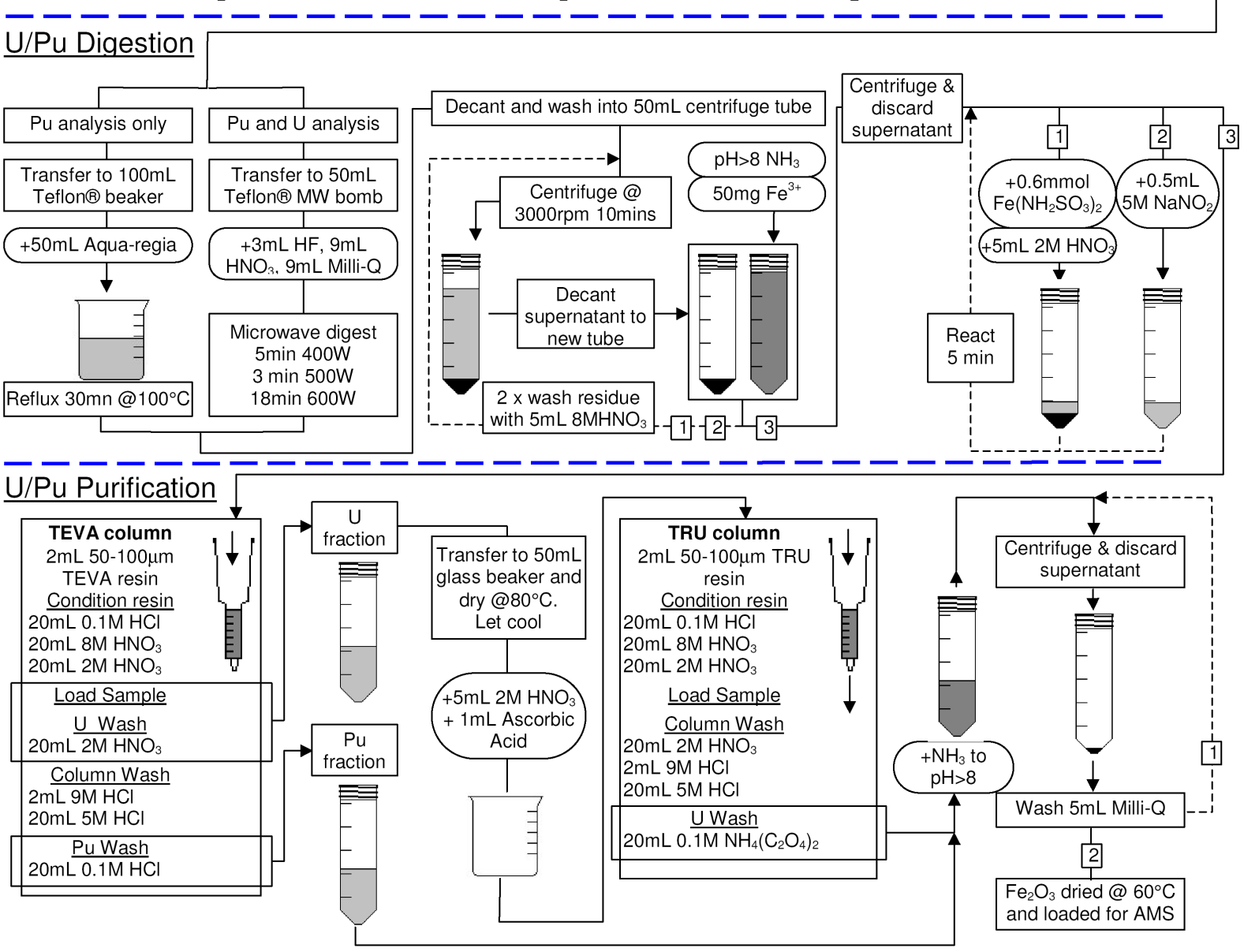
- Collaborators: D.P. Child, M.P. Johansen, K.M. Wilcken, V. Levchenko, T.E. Payne, E. Davis, J.J. Harrison, S. Thiruvoth, K. Saunders and K.L. Wilsher (ANSTO), R. Kitchen, T. Hauser (National Electrostatics Corp.), B. Smith, S. Marx (University of Wollongong), J. Knight (Griffith University), A. Wallner (ANU)

Thank you for your attention!

Sample Preparation



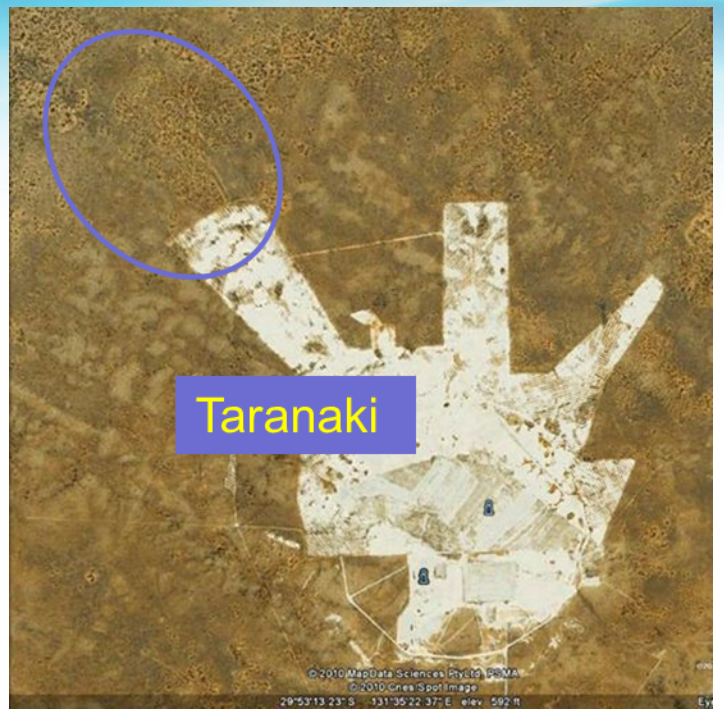

U/Pu Digestion
Pu analysis only
Pu and U analysis
Transfer to 100mL Teflon® beaker
Transfer to 50mL Teflon® MW bomb
+50mL Aqua-regia
+3mL HF, 9mL HNO_3, 9mL Milli-Q
Reflux 30mn @100°C
Microwave digest 5min 400W 3 min 500W 18min 600W
Decant and wash into 50mL centrifuge tube
Centrifuge @ 3000rpm 10mins
pH>8 NH_3
50mg Fe^{3+}
Decant supernatant to new tube
2 x wash residue with 5mL 8MHNO_3
1
2
3
Centrifuge & discard supernatant
+0.6mmol $Fe(NH_2SO_3)_2$
+0.5mL 5M $NaNO_2$
+5mL 2M HNO_3
React 5 min
U/Pu Purification
TEVA column
2mL 50-100µm TEVA resin
Condition resin
20mL 0.1M HCl
20mL 8M HNO_3
20mL 2M HNO_3
Load Sample
U Wash
20mL 2M HNO_3
Column Wash
2mL 9M HCl
20mL 5M HCl
Pu Wash
20mL 0.1M HCl
U fraction
Pu fraction
Transfer to 50mL glass beaker and dry @80°C. Let cool
+5mL 2M HNO_3 + 1mL Ascorbic Acid
TRU column
2mL 50-100µm TRU resin
Condition resin
20mL 0.1M HCl
20mL 8M HNO_3
20mL 2M HNO_3
Load Sample
Column Wash
20mL 2M HNO_3
2mL 9M HCl
20mL 5M HCl
U Wash
20mL 0.1M $NH_4(C_2O_4)_2$
+NH_3 to pH>8
Centrifuge & discard supernatant
Wash 5mL Milli-Q
1
2
Fe_2O_3 dried @ 60°C and loaded for AMS

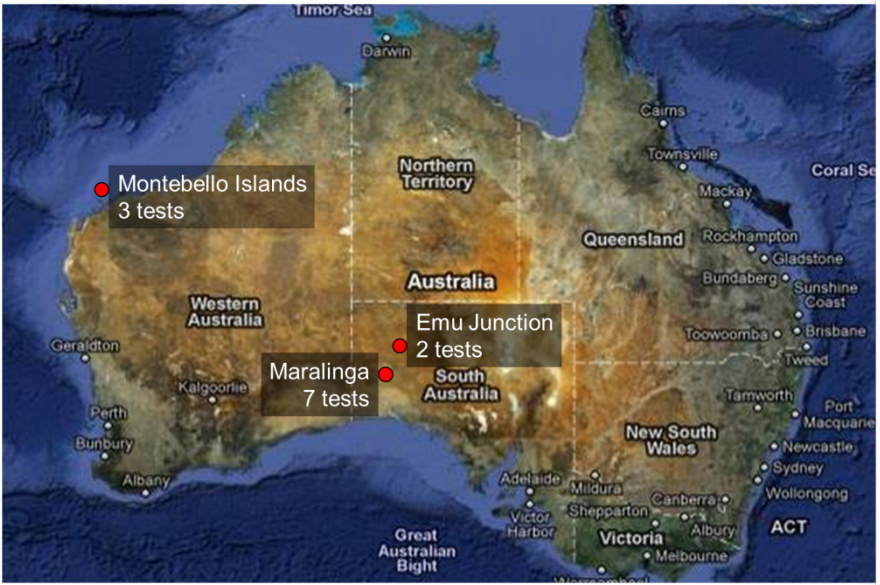
Pu at Maralinga test site

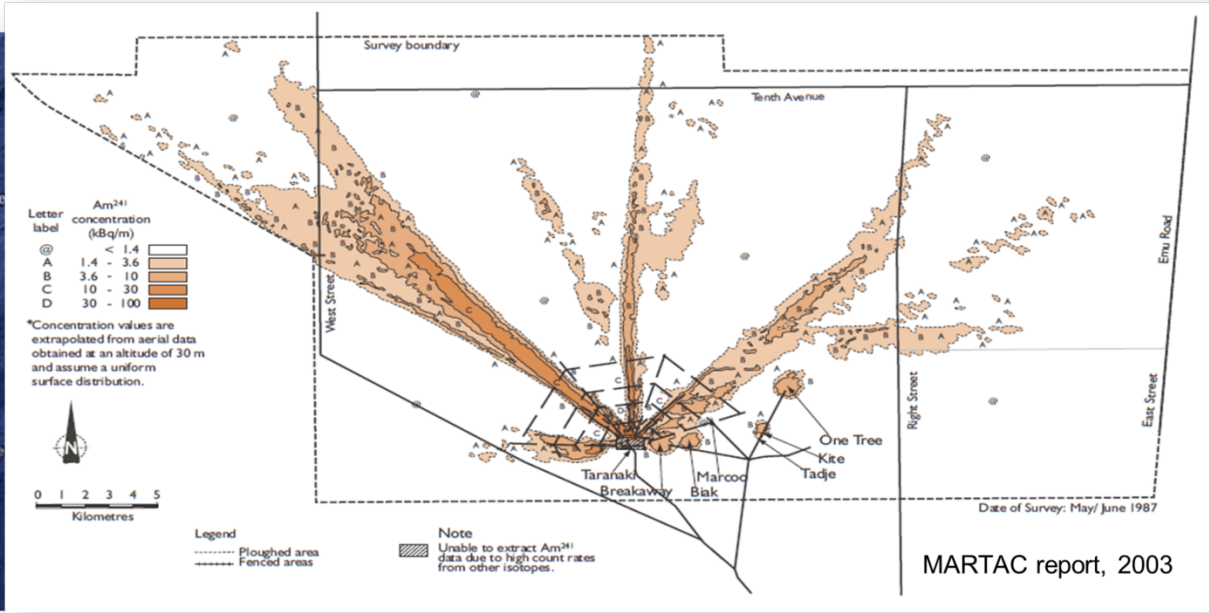
7 nuclear tests at the Maralinga test site, South Australia, 1950s to early 1960s

- series of 'safety trials' involving conventional explosives and about 22kg Pu at Taranaki
- several attempts at site remediation
- semi-arid environment

→ new data on Pu migration and uptake







MARTAC report, 2003

Pu at Maralinga test site

Concentration Ratio (CR) =

$$\frac{\textit{(activity conc. whole organism)}}{\textit{(activity conc. in soil)}}$$

Mammals, lizards and arthropods sampled

- concentration ratios compared to existing data for Pu
- differences to other data may be explained in terms of different physico-chemical forms of Pu

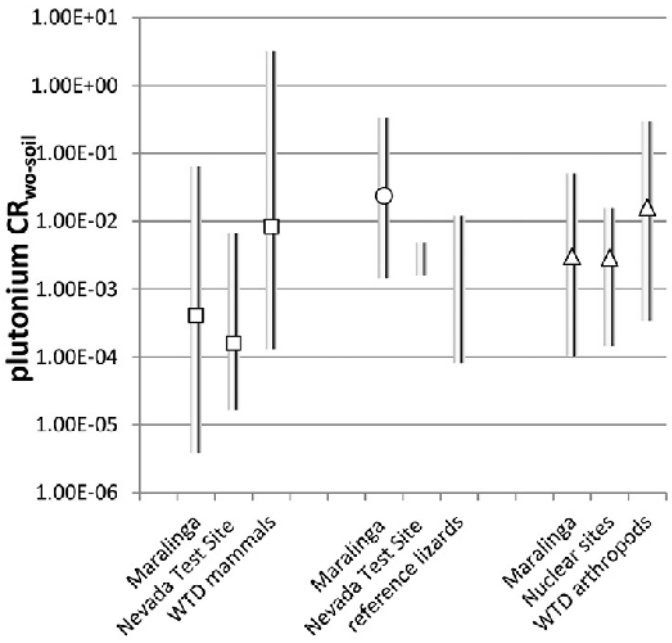
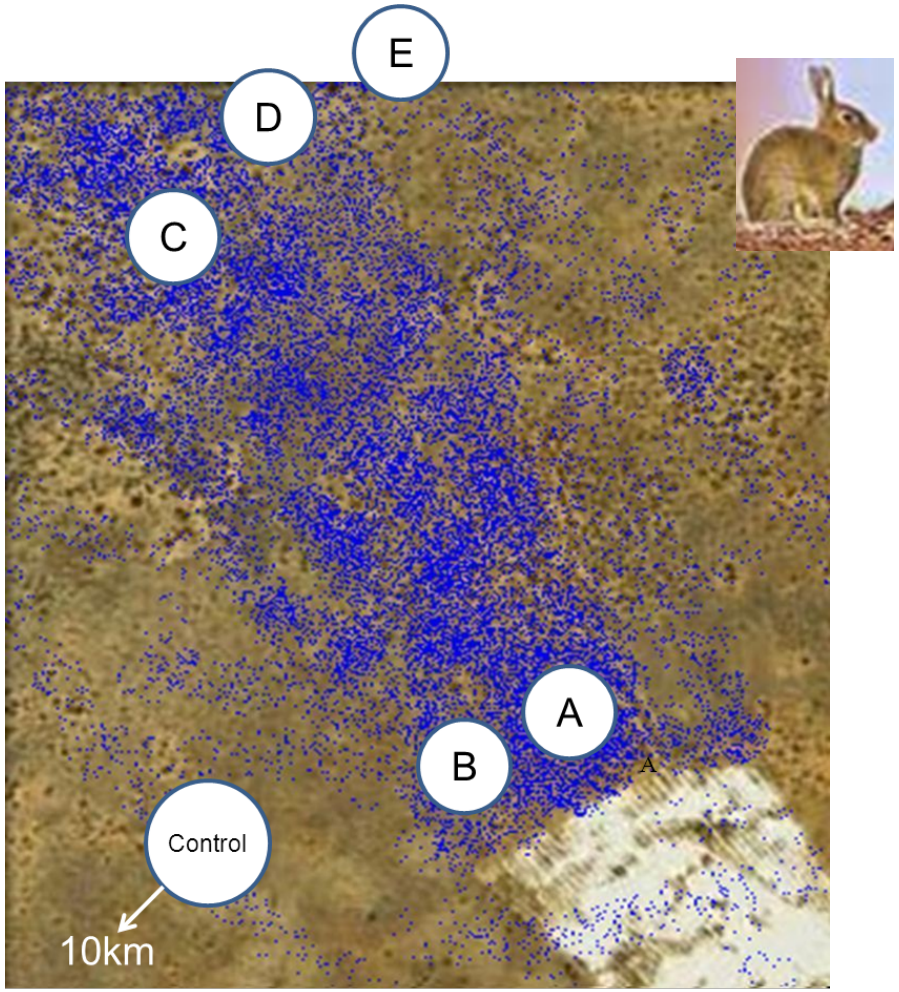


Fig. 4. Comparison of Pu $CR_{wo\text{-}soil}$ values from Taranaki, Maralinga, with reference values. Symbols are geometric means where available (□) mammals, (○) reptiles, and (△) arthropods (whiskers are minimum–maximum ranges). [1] Johansen et al. (2012, 2013); [2] www.TheWildlifeTransferDatabase.org; [3] Wood et al. (2008, 2009a).



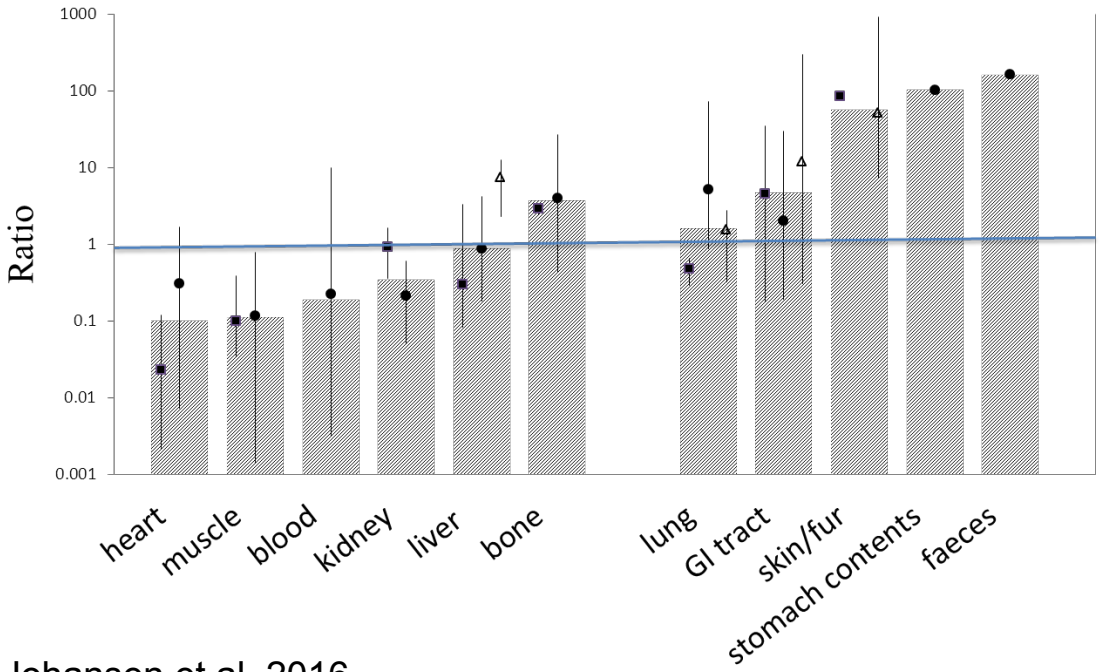
M.P. Johansen et al. / Journal of Environmental Radioactivity 131 (2014) 72-80

Pu at Maralinga test site

Distribution in mammalian tissues measured

- for red kangaroo, European rabbit; sandy inland mouse
- absorbed Pu is a fraction of the total Pu present
- implications for predator-prey relations and potential human diet

Denise Greig © Australian Museum



M.P. Johansen et al. 2016

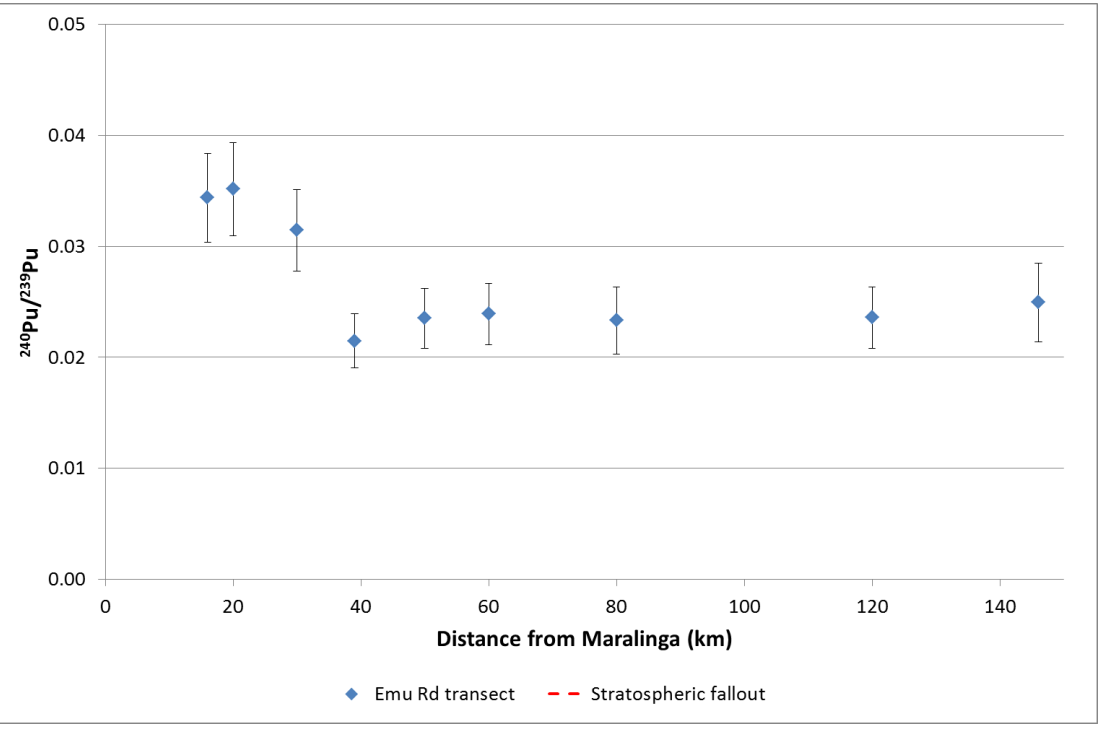
Pu at Maralinga test site

7 nuclear tests at the Maralinga test site, 2 at Emu, South Australia, 1950s to early 1960s

- series of 'safety trails' involving conventional explosives and about 22kg Pu at Taranaki (also HEU)

M. Johansen et al., 2015.

→ new data on Pu migration and uptake in wildlife



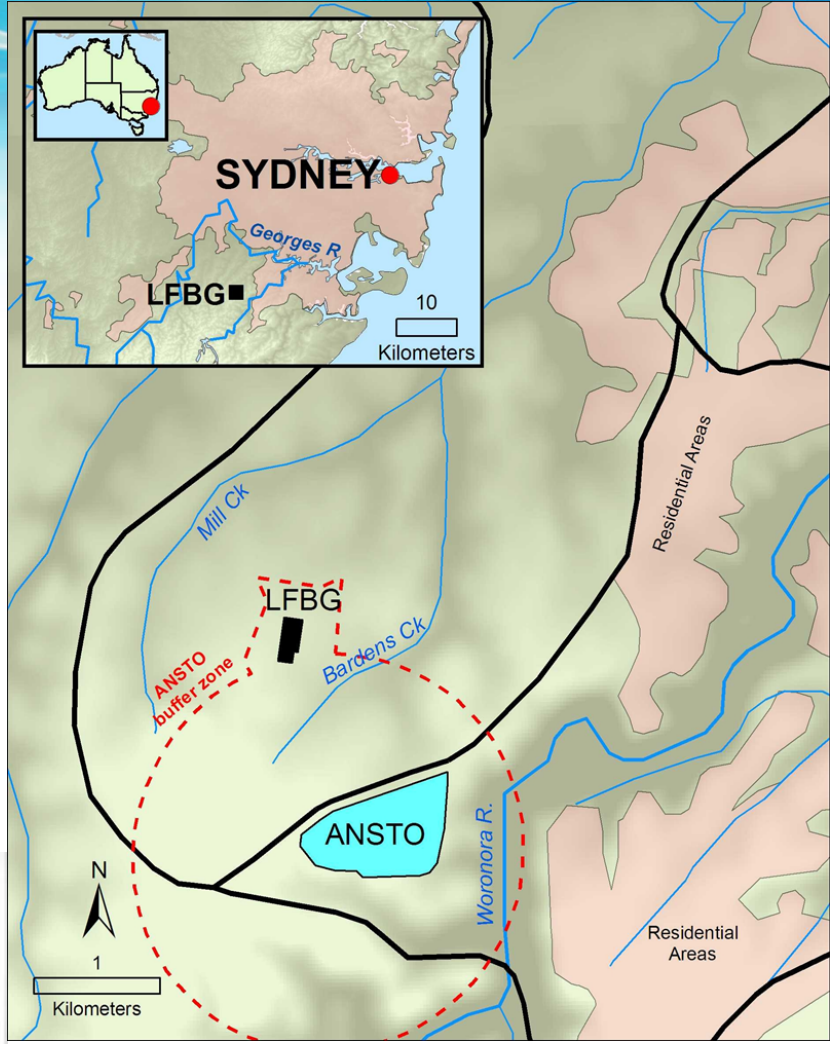
U, Pu by AMS: LFBG study

Little Forest Burial Ground

Used in 1960s to dispose of contaminated waste from the Lucas Heights site

- series of trenches dug 1960-68
- variety of waste including tritium, beryllium and activated materials
- waste included a few grams of Pu and ^{233}U

See T. Payne et al, Environ. Sci. Technol. 2013, 47, 13284.

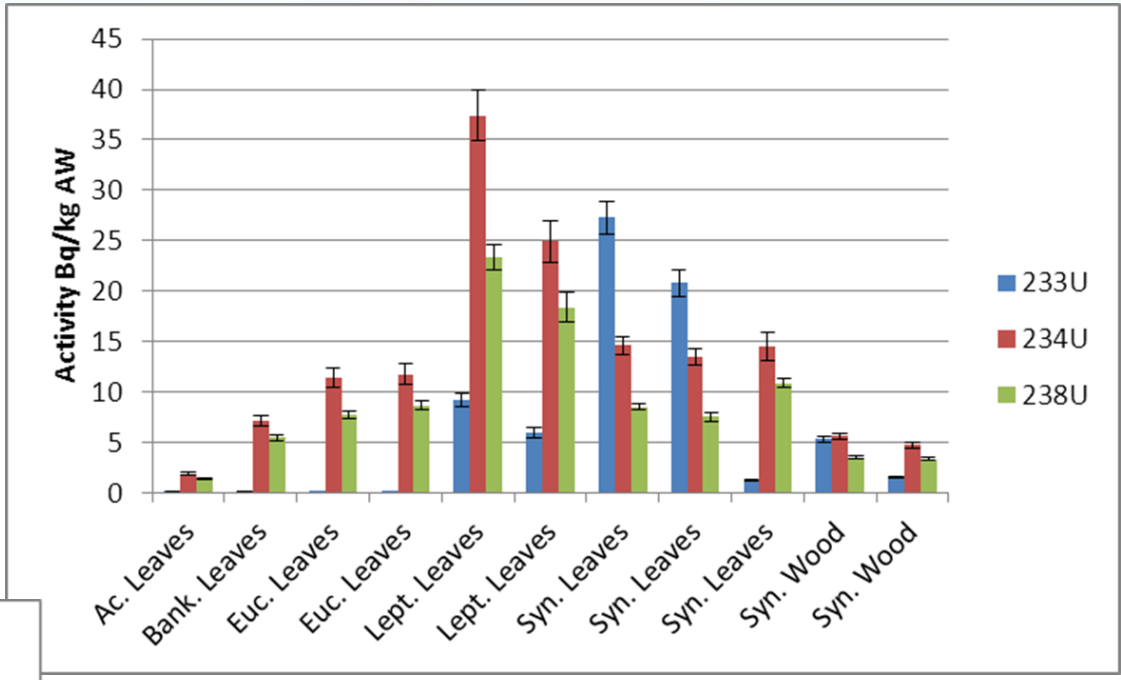


Location on edge of Sydney

U, Pu by AMS: LFBG study

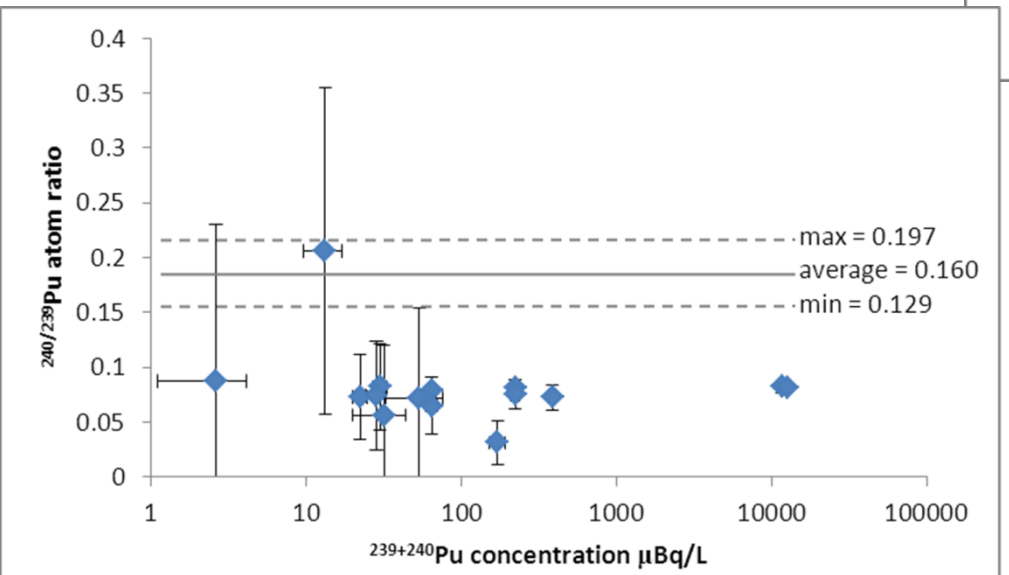
Little Forest Burial Ground

- data demonstrates very low mobility of U and Pu, but some evidence of movement to around the site boundary
- groundwaters show $^{240}Pu/^{239}Pu$ ratio distinct from fallout
- vegetation shows uptake of ^{233}U and Pu (and similar distinct ratio)



U isotopes in vegetation

K. Wilsher et al., 2015, to be published.



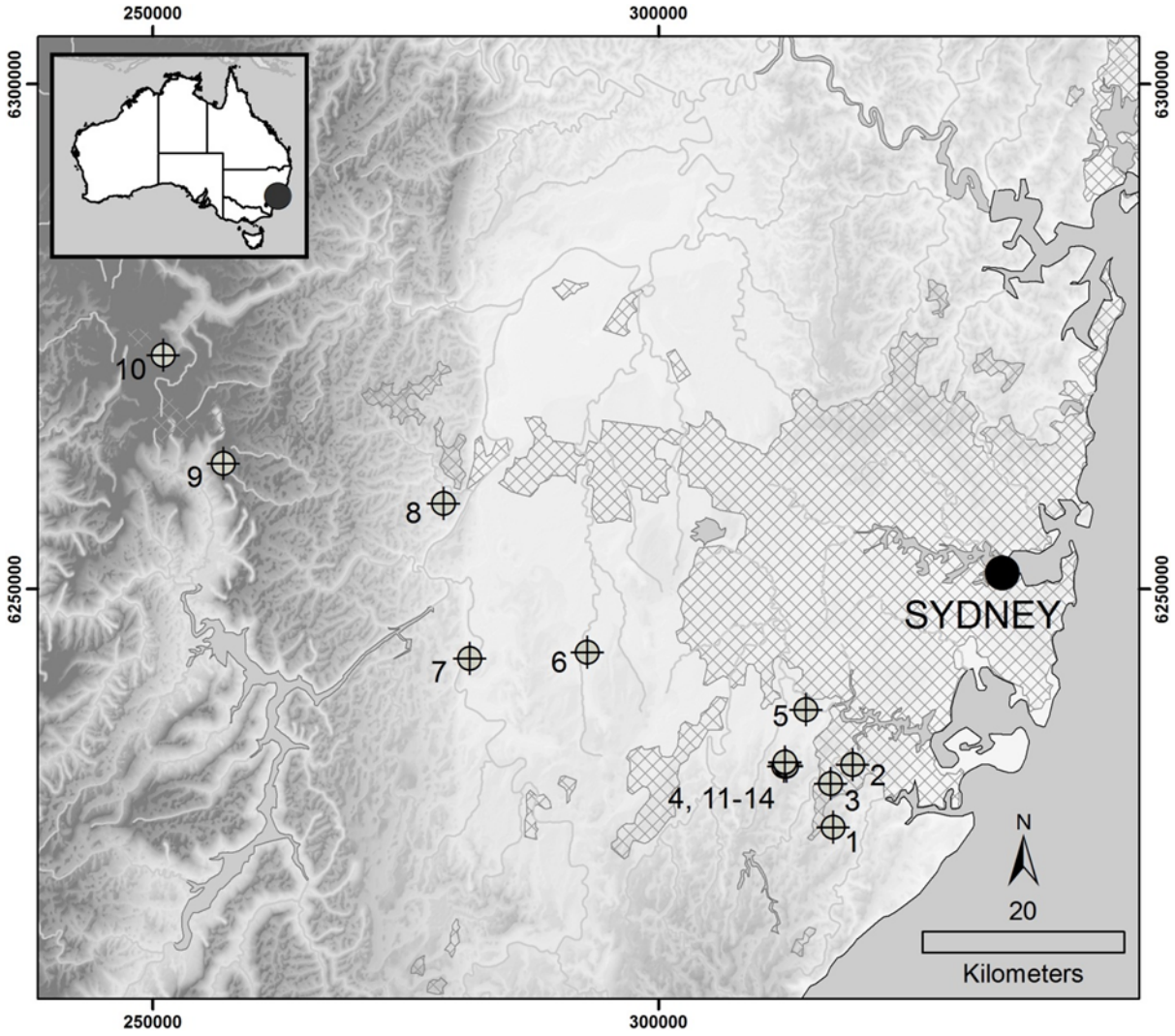
Pu isotope ratio vs concentration for LFBG groundwaters

'average' is regional average fallout

Pu Sydney baseline study

Transect running SE to NW across the Sydney basin

B. Smith et al., 2014, to be published.



Mangrove sediment, North Coast, NSW

Jon Knight, Griffith University