Environmental gamma spectroscopy: Opportunities and plans for studies with Boulby

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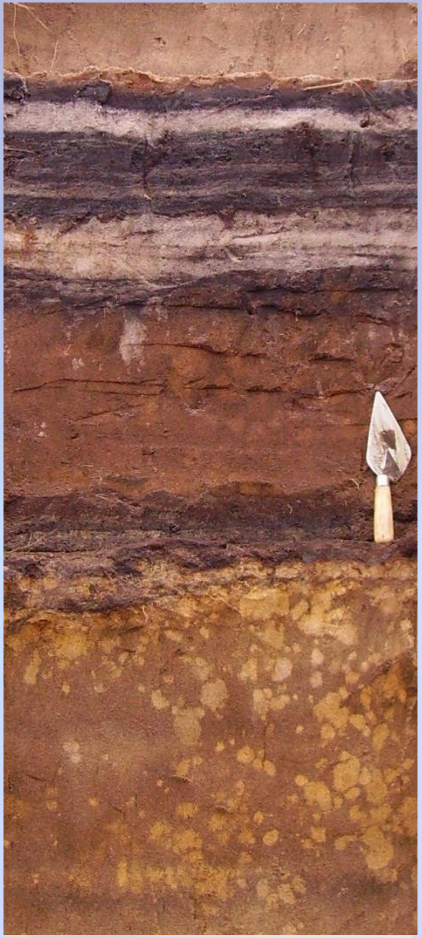
Outline

- Importance of chronology to understanding the interaction between human activity and environmental change

- Environmental radioactivity and process rates

- Low energy, low level, spectrometry and dating recent sediments
  - *OSL dating and the environmental* dose rate
  - *210-Pb dating using CRS model and the challenge of reducing detection limits*
  - *^{32}Si dating – outline of detection schemes for a radiometric chronometer to bridge the age gap between pathways and detection*

- Work underway at Boulby (STFC Futures) and future plans
  - Can we do better with an underground component?
  - How to couple surface and underground laboratories?



Climate change, population & environmental pressure, food & water supply

- Population : now > 7 b potentially rising to 9 b? Stabilisation at 7.8 b?
- IPCC IV, 2007 Group I : “*The Physical Science Basis”*
  - “Very likely” (>90%) that temperature rises are the result of increases in heat trapping gases due to human activity
  - 6 scenarios – 1-6.4ºC rise over next century
- IPCC IV, 2007 Group II : “*Climate Change: Impacts, Adaptation and Vulnerability”*.
  - Water Resource
    - Loss of glacier fed water resources; increased evaporation rates; reduced summer river flows
  - Agriculture - Loss of food production in seasonally dry low latitude zones
    - Africa – loss of rain fed crops in arid zones (?50%by 2020) –
    - loss of mangrove and coral systems (coastal fishing)
    - Asia - “high risk of food shortages” (decline of production, urbanisation, population growth)
    - Possible growth in mid latitude areas (lower frost risk, more water)
      - Potentially offset by increased losses due to extreme weather

LWEC, global security themes

- Importance of understand human response to rapid environmental change

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Introducing SUERC

The Scottish Universities Environmental Research Centre is the successor to the Scottish Universities Research & Reactor centre

Part of the University of Glasgow

Located on Scottish Enterprise Technology Park, East Kilbride.

A unique UK resource for isotope science

Research groups

Isotope Geoscience
Environmental Geochemistry
Isotope Biology
Environmental Physics

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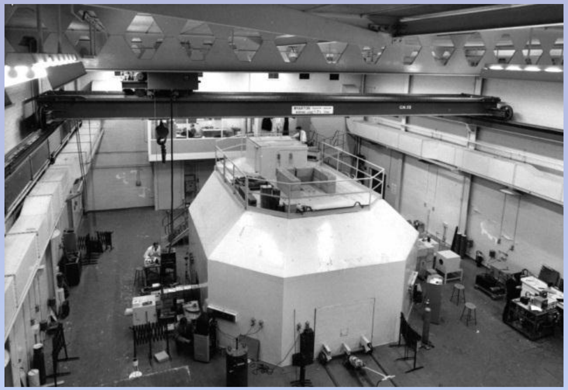

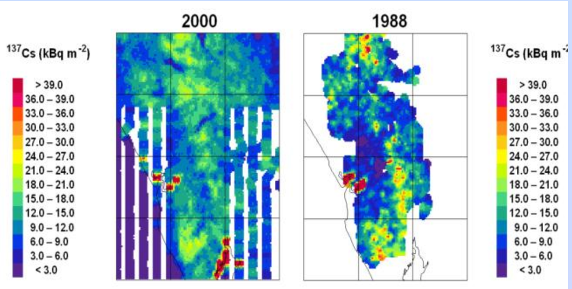

SURRC 300 kW research reactor

- opened by John Cockcroft in 1963
- neutron facilities, isotope production, and training in nuclear science
- decommissioned from 1995
- successful site delicensing 2008

Accelerator Mass Spectrometry

- established 2000
- 5 MV Pelletron, dual ion source AMS
- 1 MV single stage AMS
- The most productive facility in Europe (^{14}C, cosmogenic nuclides, ^{129}I etc)

Hosts 5 NERC NSS Facilities

Radiocarbon dating
^{39}Ar-^{40}Ar dating
Cosmogenic Isotope Facility
Stable Isotope Geoscience support
Stable Isotope Life Science support

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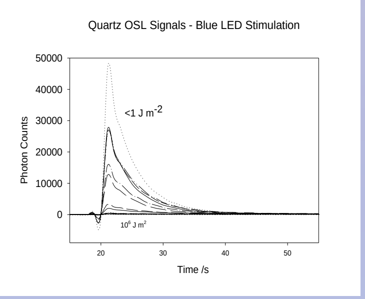
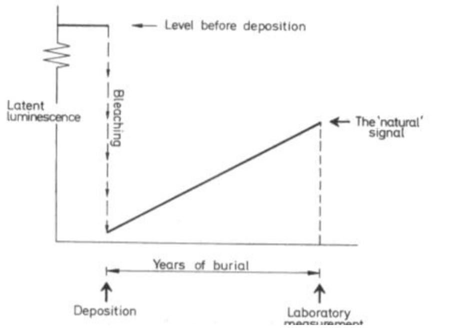
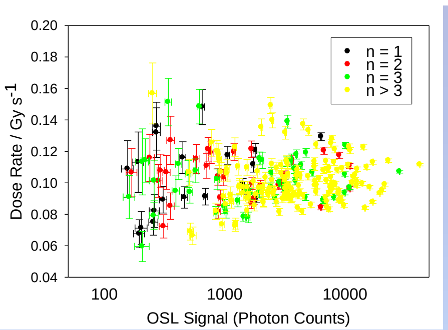
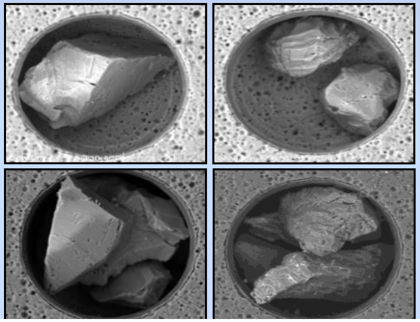


Luminescence dating

- **Age (ka) = Dose (Gy)/Dose Rate (mGy a^{-1})**
- Dose (Gy) – determined from stimulated luminescence measurements of natural minerals
- Dose Rate (mGy a-1) determined by radiometric and radioanalytical measurements coupled to a microdosimetric model
- Age range : 0-10^6 a
- Applicability : heated materials, light exposed sediments
- Uncertainties – combining sample derived (variable from case to case) and fixed (calibration) uncertainties
- **Challenges : reduction of errors from smaller (complex) samples**

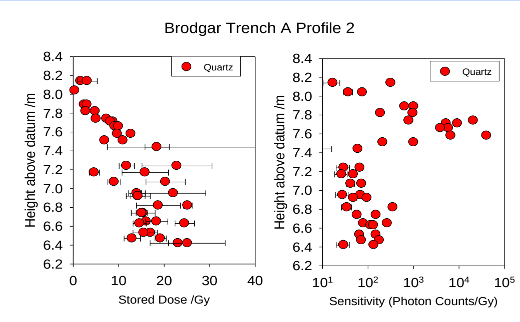


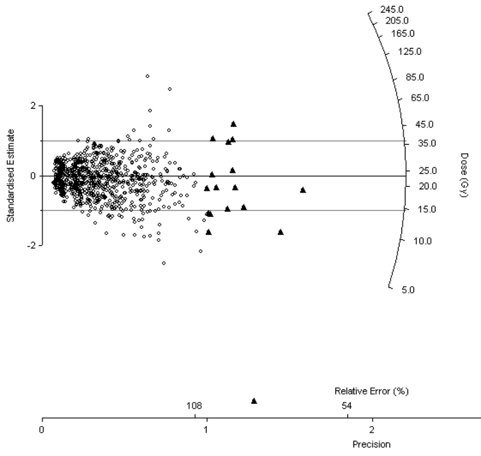




Sanderson, D.C.W., Murphy,S., 2010, Using simple portable OSL measurements to understand complex sediment sequences for luminescence dating, Quaternary Geochronology 5, 299-305

Age / Years	± 5%	± 10%	± 15%
10	0.5	1	1.5
100	5	10	15
1000	50	100	150
10000	500	1000	1500
100000	5000	10000	15000





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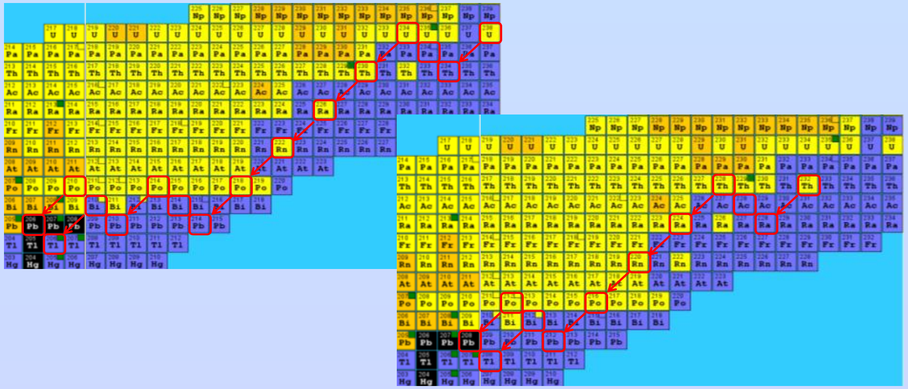
Environmental Radioactivity & Radiation Dose Rate determination

- Dose contributions to luminescence dating
- ^{40}K (beta, gamma)
- U and Th and their decay products (alpha, beta, gamma)
- Alpha and beta radiation from within the sample
- Gamma and cosmic radiation from the environment



Methods used by SUERC

In-situ gamma spectrometry

to record the gamma dose rates from the excavation environment (typically 20-30% of the total dose rate)

High Resolution gamma spectrometry in the laboratory

to determine the U,Th, K mixture of the sample and to model alpha, beta and gamma dose rate contributions

Thick Source beta Counting

direct measurements of beta dose rates from dry samples

Modelling

water content corrections, cosmic ray dose rates, effective dose rates

(NAA, ICPMS)

Can we improve dose rate measurements underground?

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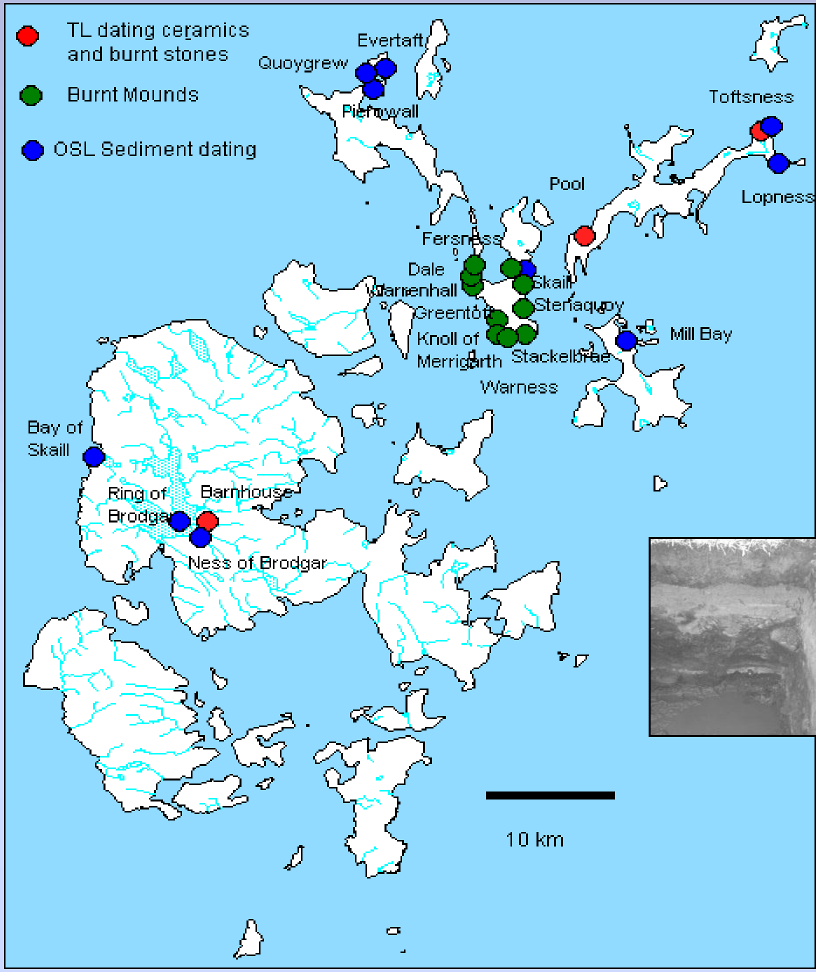
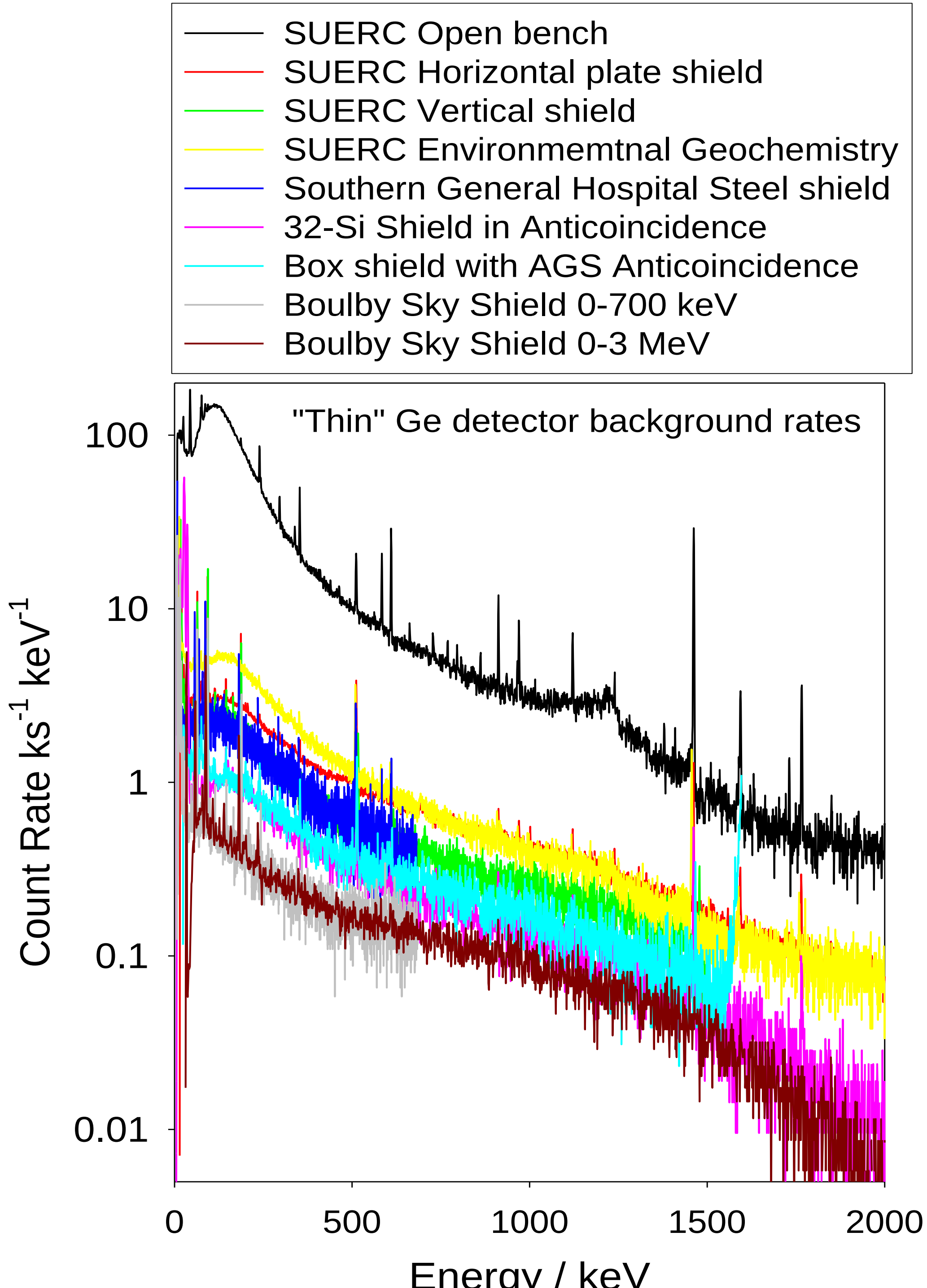
Luminescence dating and Neolithic Orkney

- Heated materials - Pool, Toftsness, Barnhouse
  - ceramics and hearthstones (TL from 1980s; published 2007)
  - Study of firing temperatures (Joel Spencer PhD thesis 1996)
  - Combined TL and OSL on heated materials (Iona Anthony, 2003, PhD thesis, Luminescence Dating of Scottish Burnt Mounds : New Investigation in Orkney and Shetland, University of Glasgow) oai:theses.gla.ac.uk:1632
- OSL dating of sediments - Landscape, environment, site formation processes
  - Wind blown sands on archaeological sites - storminess, sediment supply, links to Atlantic climate system, human impacts, records of abandonment (Anne Sommerville, 2003. PhD Thesis, University of Glasgow)
- Ditch fill sediments from the Ring of Brodgar
  - Exploratory work at the Ness of Brodgar
  - Environmental section on Stronsay
  - New initiative on surface exposure dating

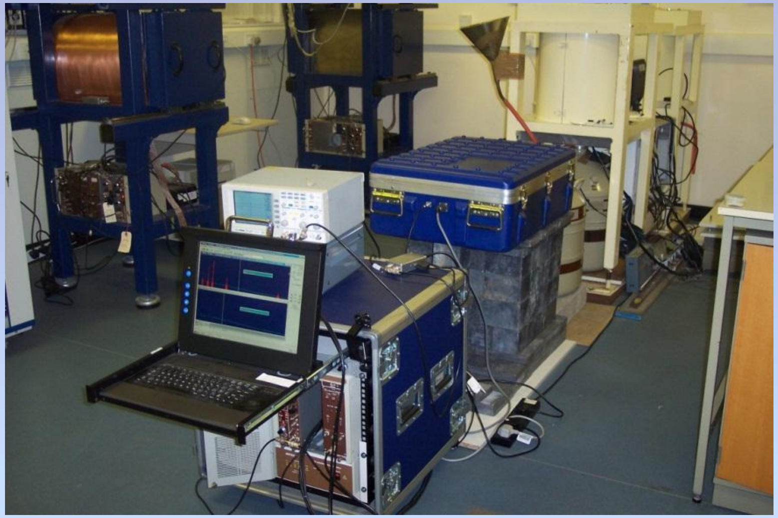


Sommerville et al, 2007, *The Holocene 17(5)1-11*

Tofts Ness (lower sand)	602	0.986 ± 0.076	4.67 ± 0.16	4735 ± 400	2735 ± 400 BC	2260 ± 100 BC	H4 2350-2250 BC
	603	0.602 ± 0.068	2.47 ± 0.13	4100 ± 510	2100 ± 510 BC		
	612	0.92 ± 0.045	3.25 ± 0.09	3530 ± 200	1350 ± 200 BC		
	613	0.963 ± 0.033	5.06 ± 0.13	5255 ± 220	3255 ± 220 BC		
	616	1.546 ± 0.052	6.72 ± 0.28	4345 ± 230	2345 ± 230 BC		
	617	1.552 ± 0.053	6.24 ± 0.21	4020 ± 190	2020 ± 190 BC		







Comparison of background rates for thin "n" type Ge detectors

Boulby is best (so far) !

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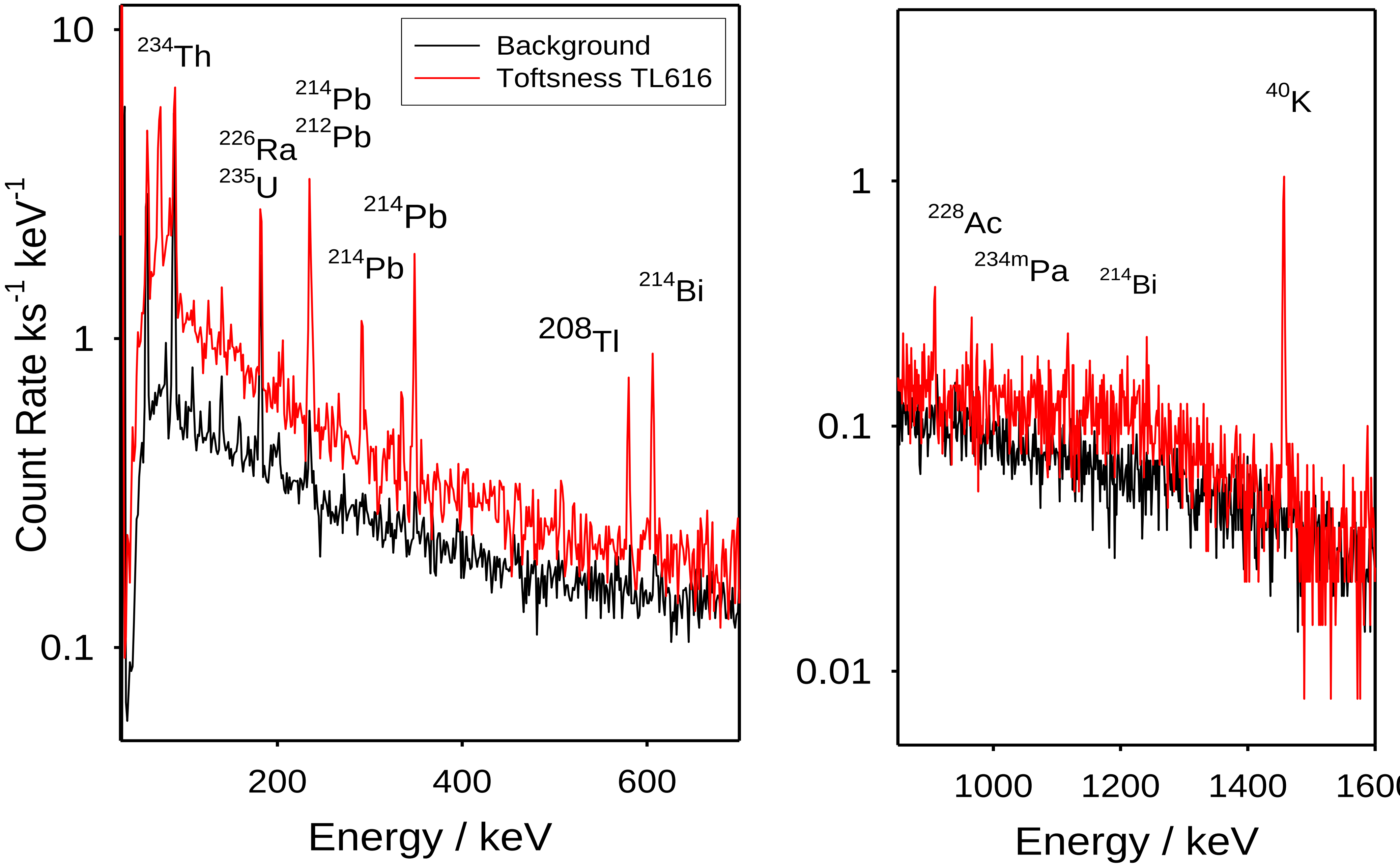
Analysis of small OSL dating samples

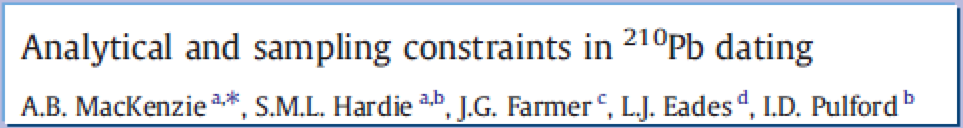


SUTL616 Toftsness lower sand – 78ks analysis at Boulby (LoAx 20 g) Dec 2012

	Activity Concentration		Concentration		Dose Rates /mGy/a					
Source	Bq/kg	Error	%/ppm		Alpha	Error	Beta	Error	Gamma	Error
K	580.33	50.29	1.88	0.16			1.56	0.13	0.45	0.04
U	27.21	3.21	2.20	0.26	6.12	0.72	0.32	0.04	0.25	0.03
Th	18.81	9.08	4.64	2.24	3.43	1.65	0.13	0.06	0.24	0.11
				Total	9.55	1.80	2.01	0.15	0.94	0.13

SUTL616 Toftsness lower sand – 100ks analysis at SUERC (GMX1 20 g) Feb 2001

	Activity Concentration		Concentration		Dose Rates /mGy/a					
Source	Bq/kg	Error	%/ppm		Alpha	Error	Beta	Error	Gamma	Error
K	575.63	27.52	1.86	0.09			1.54	0.07	0.45	0.02
U	36.12	3.86	2.93	0.31	8.13	0.87	0.43	0.05	0.34	0.04
Th	14.35	7.12	3.54	1.76	2.61	1.30	0.10	0.05	0.18	0.09
				Total	10.74	1.56	2.07	0.10	0.97	0.10



Analytical and sampling constraints in ^{210}Pb dating

A.B. MacKenzie [a,*], S.M.L. Hardie [a,b], J.G. Farmer [c], L.J. Eades [d], I.D. Pulford [b]

Science of the Total Environment 409 (2011) 1298–1304

“210Pb dating provides a valuable, widely used means of establishing recent chronologies for sediments and other accumulating natural deposits. The Constant Rate of Supply (CRS) model is the most versatile and widely used method for establishing 210Pb chronologies but, when using this model, care must be taken to account for limitations imposed by sampling and analytical factors. “

“For a core that is of sufficient length to extend to the depth at which unsupported 210Pb activity reaches the limit of detection, negligible, or near negligible, d deviation of CRS ages would be generated if the limit of detection was 1 Bq kg−1 or better. However, with a limit of detection of 10 Bq kg−1, bias towards erroneously old values could be generated for ages older than about 80 years.”

^{210}Pb dating

- **radon decay product with 22.5 a half life**
- **deposited unsupported as aerosols**
- **Depth profile of activity concentration can be used to establish sedimentation rates over c. 100-200 a**
- **Low energy gamma emitter**

Can we produce < 1 Bqkg^{-1} detection limits to meet the MacKenzie et al criteria?

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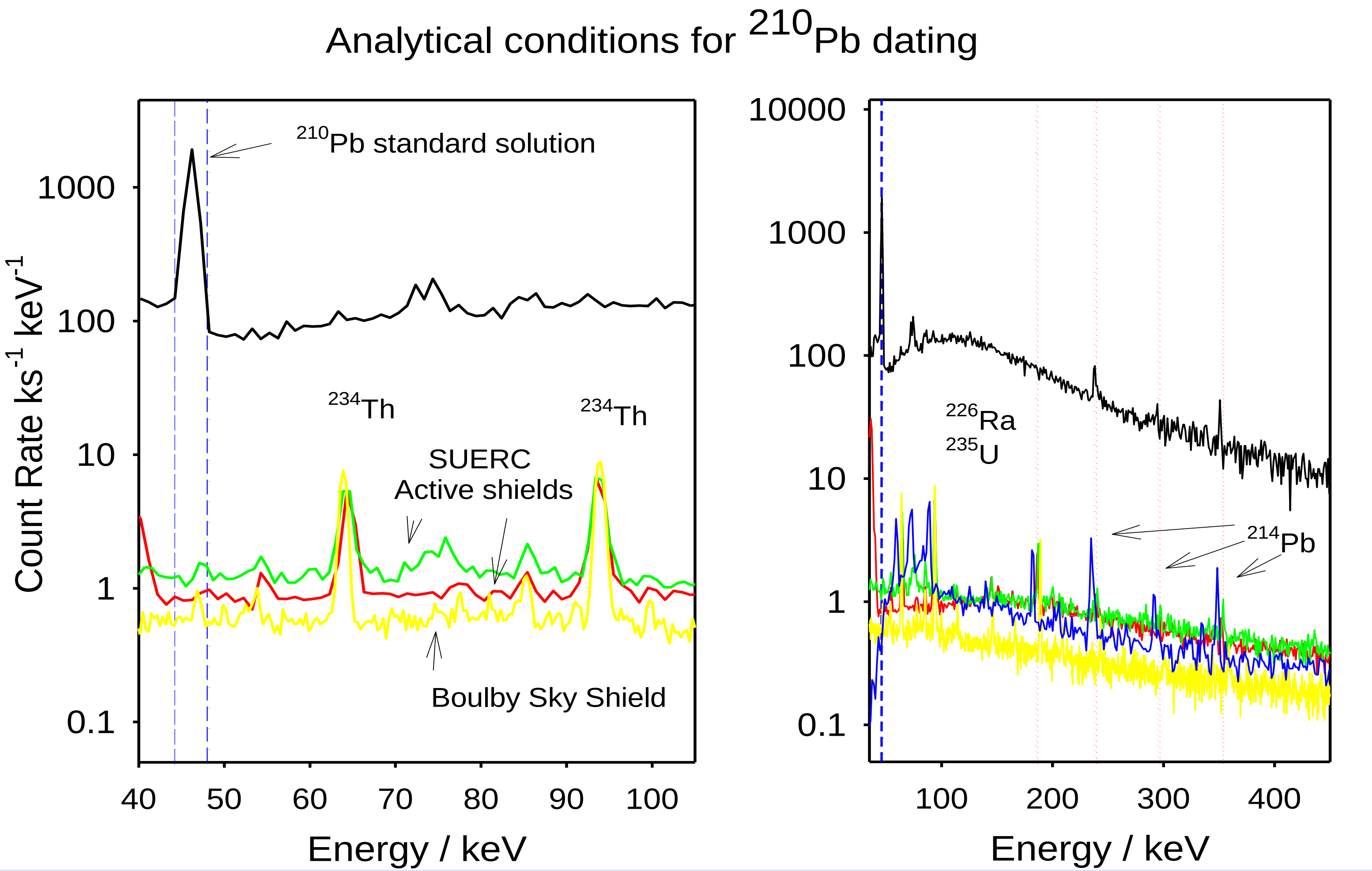
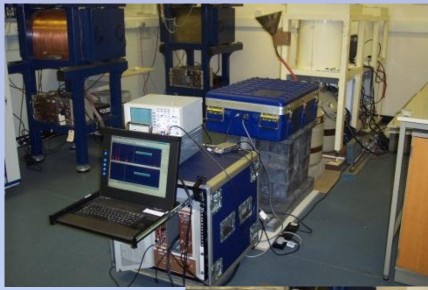
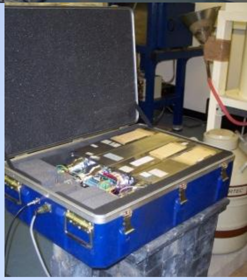
^{210}Pb gamma spectrometry for unsupported and supported components



Analytical conditions for ^{210}Pb dating





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Initial detection limit estimates for 46.5 keV ^{210}Pb line

Detection limits in Bq kg-1	SUERC				Boulby	
Counting time	32-Si AC system		Box Shield AC		Sky Shield	
ks	5 g	20g	5g	20g	5g	20g
1	30.0	14.0	46.4	21.6	25.4	11.9
10	10.2	4.7	6.0	2.8	3.7	1.7
100	3.9	1.8	1.9	0.9	1.5	0.7
1000	1.9	0.9	1.5	0.7	1.3	0.6
10000	1.3	0.6	1.5	0.7	1.3	0.6

Can we produce < 1 Bqkg^{-1} detection limits to meet the MacKenzie et al criteria?

Yes ! (
– but more work needed to assess the effect of supported activity on this
further detector improvements are planned)

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^{32}Si dating: The missing radiometric chronometer?

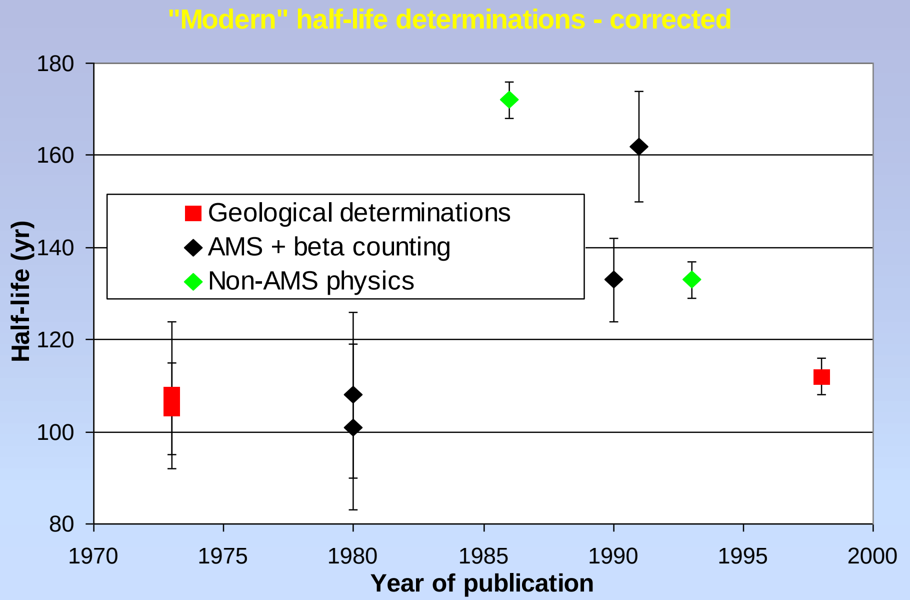
- Production by spallation of Ar in atmosphere [e.g.: $^{40}Ar(n,2\alpha n)^{32}Si$]
- Half life: 140 ± 7 Years (Morgenstern et al., 1996)
- Beta decay

$$^{32}Si \xrightarrow[t_{1/2} = 140\ y]{\beta^- (E_{max} 225\ keV)} {}^{32}P \xrightarrow[t_{1/2} = 14.3\ days]{\beta^- (E_{max} 1710\ keV)} {}^{32}S$$

- Promising half life for dating in the age gap
- Potential use as an environmental process tracer
- But natural activity and abundance levels have greatly limited applications

- NERC grant (NE/B50606X/1) Development of an ultra low-level system for analysis of ^{32}Si from environmental samples

- Summarise this and indicate how we might examine a role for underground analysis



"Modern" half-life determinations - corrected

The analytical challenges

Observed activities of ^{32}Si in natural media

Matrix	^{32}Si activity	Reference
Rain water	13.1 mBq m^{-3}	Morgenstern et al., 2000
Lake water	1.3 Bq kg^{-1} SiO_2	Nijampurkar et al., 1998
Lake sediment	1 Bq kg^{-1} SiO_2	Nijampurkar et al., 1998
Marine sediment	2.2 mBq kg^{-1} sediment	De Master and Cochran, 1982

First analysis of natural ^{32}Si: Lal et al. (1960)

^{32}Si by AMS limited by :

^{32}S isobaric interference
Presence of stable Si
$^{32}Si/Si$ ratio of 10^{-15} or greater
Required which restricts analysis to matrices suchas rainwater, snow, ice and some fresh waters.

Radiometric analysis

Liquid scintillation spectrometers, surface barrier detectors and gas proportional counters have been used but:

Best available background count rates (4 – 9)x10^{-4} cps.
Detection efficiencies 35 – 66%
Large sample sizes
Only one sample can be analysed at a time

Approach taken in project NE/B50606X/1
Develop a multi-sample array of beta spectrometers for parallel analysis of many samples, background mimimise by active and passive shielding

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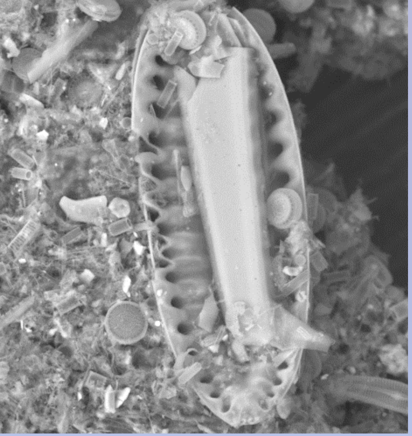
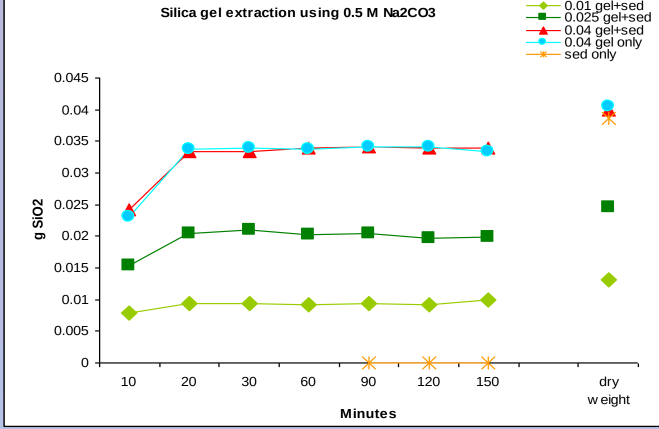


Sample Chemistry based on ^{32}P detection developed by A.B. MacKenzie and H. Rossitor



Stages:

1) **Silica extraction (0.5 M Na_2CO_3)**
2) **Storage for ^{32}P ingrowth (6-8 weeks)**
3) **Phosphorus separation ($CaMoPO_4$)**
4) **Source preparation and counting (6-8 weeks)**

Stage	Mass of silica	Activity of ^{32}Si /mBq	Mass of P	Activity of ^{32}P
^{32}Si extraction & purification	10 g	10 mBq	0	0↑
Storage for ^{32}P ingrowth	10 g	10 mBq	↑ 2.6 mg + carrier	↑10 mBq
^{32}P precipitation	0	0	2.6 mg + carrier	10 mBq
Source preparation	0	0	2.6 mg + carrier	10 mBq↓

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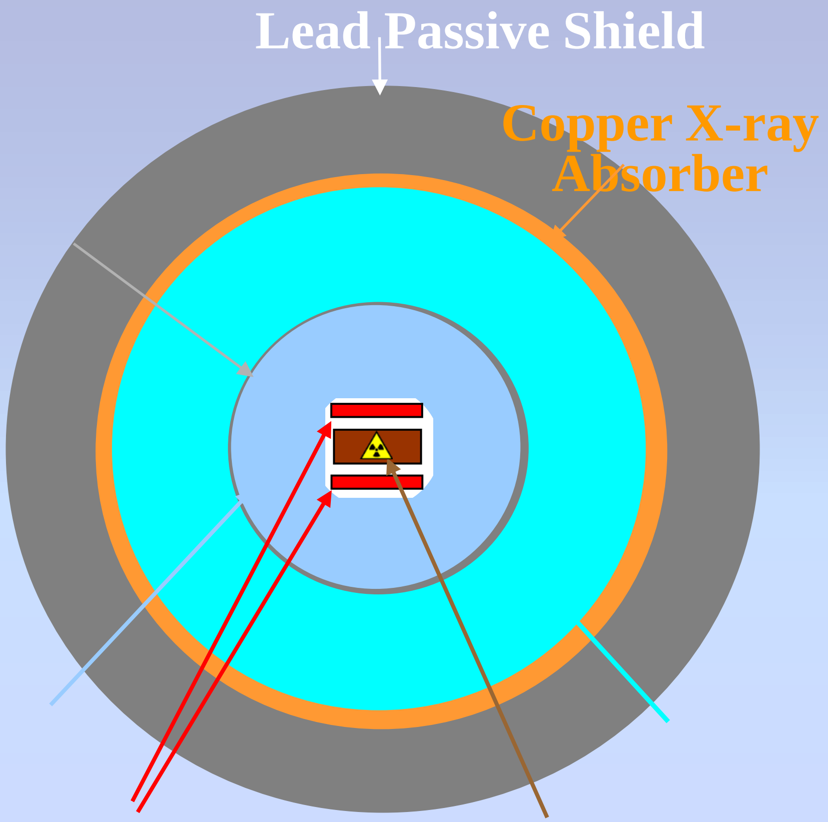
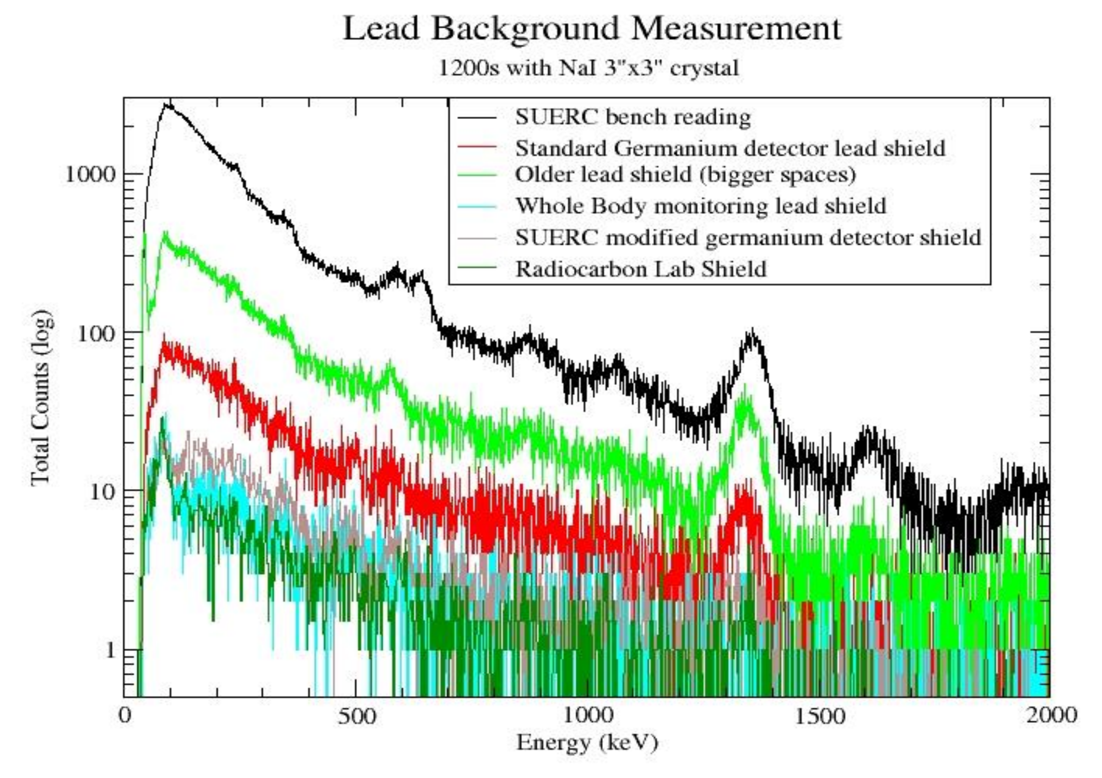
Detector Concept

- Use of Photodiodes
  - 100 mm^2 diodes
  - New wafer designs
- Passive shielding
- Active shielding
  - NaI vs CsI well detectors
- Pre-amplifier development
- Multichannel nucleonics system



Passive shielding



- **Analysis of the purity of possible lead sources and shield designs.**

➢ **Decision made to develop custom cast ring shields using recycled lead bricks from the SURRC Whole body Monitor**

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Active shielding

Shield I: NaI

Blind well
Dimensions
75 x 200 mm

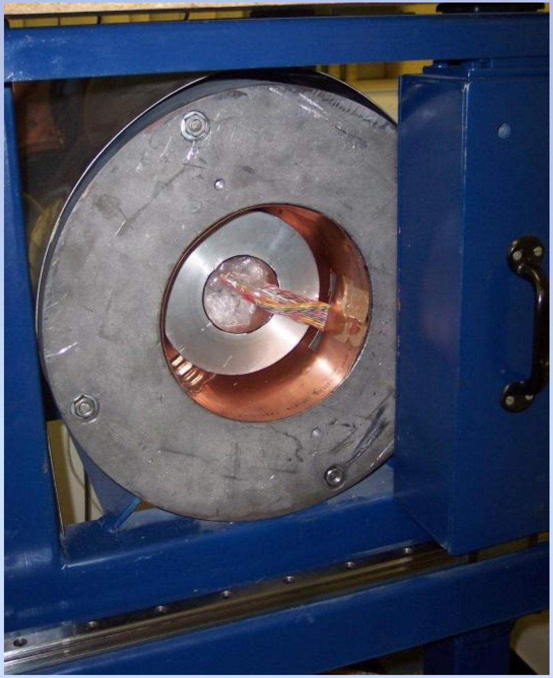
Single PMT from
Rear face

Which is best?

Peak to compton
considerations

Activation of ^{134}Cs

Ionic radius of ^{40}K



Shield 2: CsI

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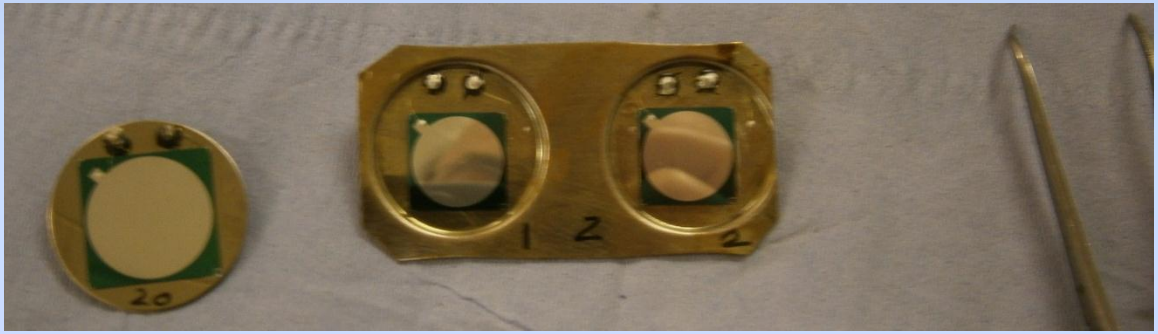
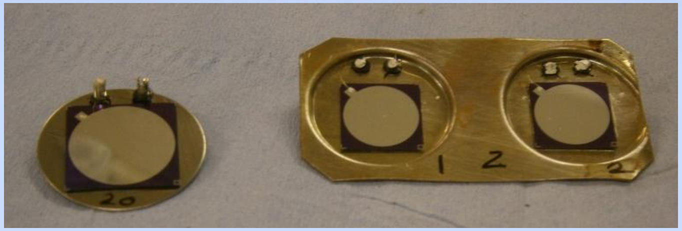
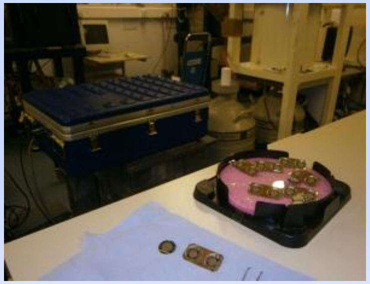
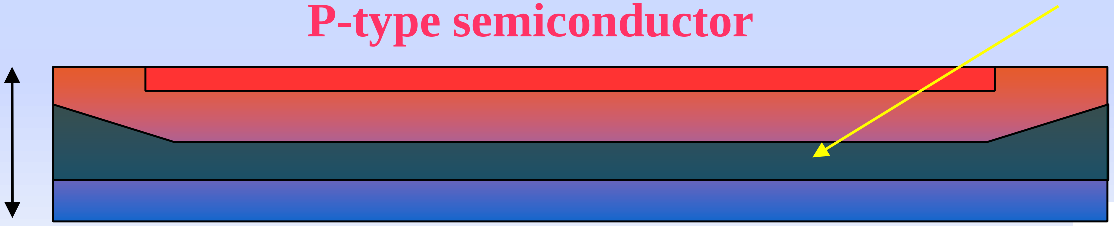
Silicon Photodiodes

- Good efficiency for x-rays and beta particles
- Stable charge output with low levels of inherent noise (for <1-2 cm^2)
- pure material – (bare diodes have v. low internal background)
- easily scaled up for use in arrays
- custom diodes made (Semefab, Glenrothes) on alpha free substrates









Depletion Layer

P-type semiconductor

N-type semiconductor

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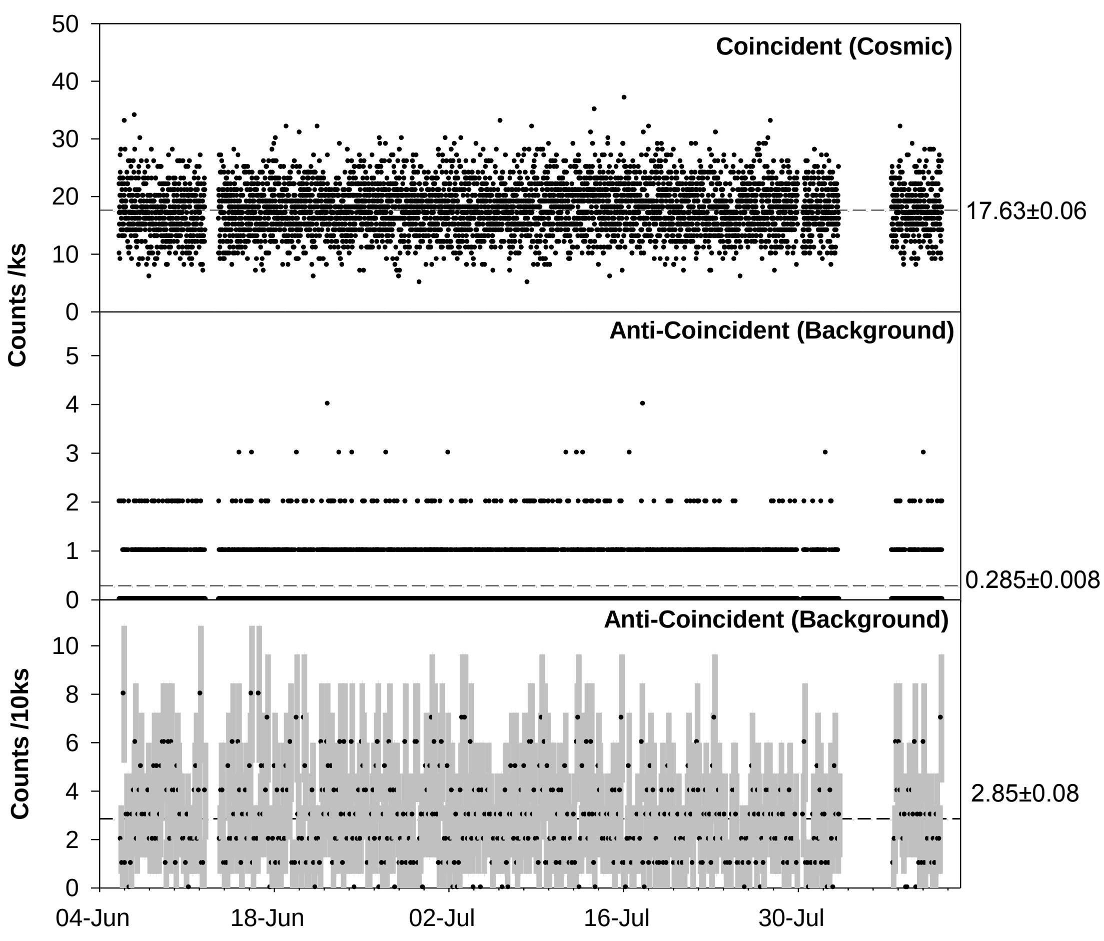
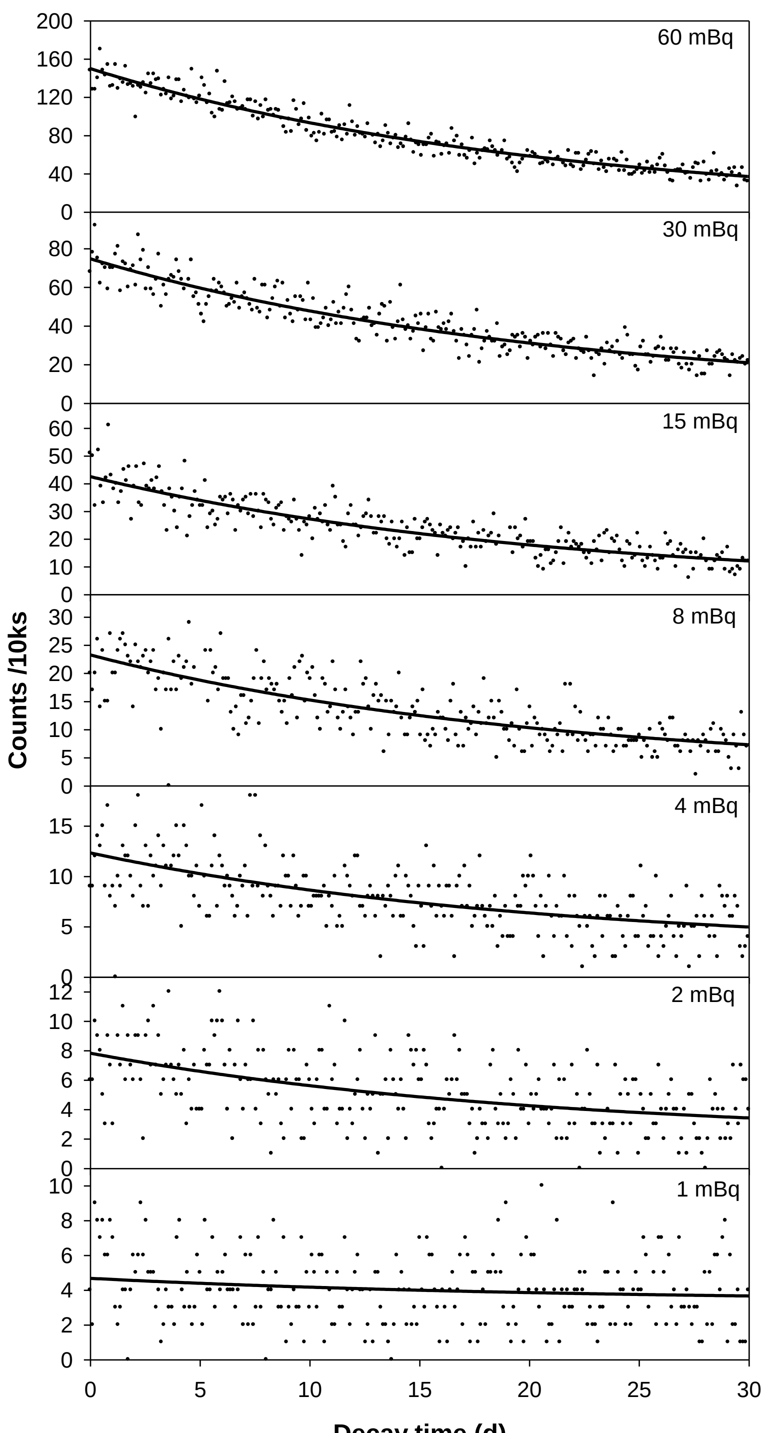
Measurements of ^{32}P Decay





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Detection Limits (mBq) for ^{32}P achieved in NE/B50606X/1

Activity (mBq)	**2 months counting**	
	Initial rate (10^{-4} s^{-1})	Background rate (10^{-4} s^{-1})
60	147±1	3.3±1.1
30	72.2±0.9	3.5±0.8
15	40.3±0.7	2.6±0.6
8	20.9±0.5	2.4±0.5
4	9.4±0.4	2.9±0.4
2	4.5±0.6*	2.9±0.7*

- **The current detection performance permits analysis of natural samples with 10-50 g of biogenic silica; a significant improvement compared with prior requirements for 100-500 g of biogenic silica**
- **Could we do better in the underground environment?**

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Summary and conclusions

- Work has been initiated between SUERC and the underground laboratory at Boulby to explore opportunities for low level spectrometry applied to environmental sciences
- In the first instance three examples relating to sediment chronometry have been examined
- Very promising initial results
- We plan to explore further background reduction within the prototype systems
- Also to review comparative performance with surface and other underground laboratories
- We would like to identify applications where the benefits of underground work can be optimally coupled to applications of high scientific quality and good fit to the priority challenge areas
- Finally it is important to build communities on the basis of effective interdisciplinary and interinstitutional collaboration, and to find good models for coupling surface and underground capabilities