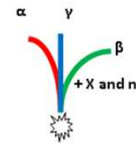


SOME EXAMPLES OF SENTENCING WASTE AT THE EXCLUDED/VERY LOW LEVEL WASTE BOUNDARY



Pete Burgess, Radiation Metrology Ltd
(radiationmetrology@gmail.com)

Problem 1

- Cambridge University had worked with Pa-231 in a fume cupboard which had discharged into a ventilation duct made from asbestos cement
- The university wished to remove the ducting both to reduce the radiological risk and to upgrade ventilation to modern standards
- The Environment Agency were keen this happened and supported the project
- The aim was to treat the debris as asbestos, rather than as radioactive, waste



3

Relevant legislation (then)

- Asbestos – Hazardous Waste Regulations
- Radioactivity – The Phosphatic Substances and Rare Earths Exemption Order
- Allowed disposal without a permit of material contaminated up to 14.8 Bq/g elemental (not isotopic) activity for a range of specified naturally occurring radioelements
- Two polonium isotopes (see later) hence the head of chain activity limit = 7.4 Bq/g if in equilibrium
- Anything above treated as Low Level Waste
- Now replaced in UK legislation with something more sensible

The duct material

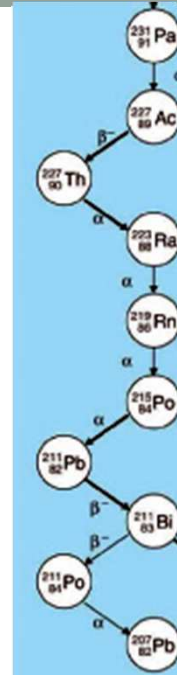
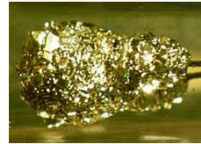
- Urastone, a popular cement – asbestos composite
- (At least until we realised how nasty asbestos is)
 - “Former Asbestos Worker Receives £62,000 Mesothelioma Compensation ”
- Used for many cast products
- A difficult monitoring surface – not smooth and rather dusty



4

Pa-231 Decay Scheme

- Decay product of U-235
- Simplified version with major decays only
- Actinium-227, the first decay product, has a 22 year half life
- The rest are quick – days at most
- Work was performed about 40 years ago
- UKAEA separated material from 1960?
- Radon isotope has a 3.9 s half-life, hence limited escape
- Hence significant ingrowth



5

6

Available radiations

- Several alpha decays – difficult to monitor on dusty, porous surfaces
- One low energy beta – Ac-227 to Th-227 – too low to be useful
- Two energetic beta emitters
 - Pb-211, mainly at 1372 keV E_{max}
 - Tl-207 at 1423 keV E_{max}
- Hence a mixture of energetic beta emitters and alpha emitters

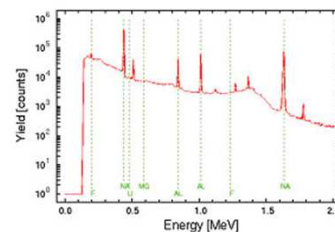
Surface monitoring equipment used

- Thermo BP19 and Ludlum 44-142
- Both respond efficiently to the beta emitters expected and, less efficiently, to alpha emitters
- **Alpha and low E beta response suppressed using 8 mg cm⁻² plastic sheet – reduces surface condition dependence**
- Response to Cl-36 (0.714 MeV) and Sr-90 + Y-90 (0.54 + 2.27 MeV) determined with the plastic sheet in place
- Measured response = 80 % of open window values



Fingerprint

- Age of the material was not well defined
- Samples were taken for gamma spectrometry
- Results were rather variable
- First set – Pa-231, Th-227, Ra-223, Rn-219, Pb-211 and Bi-211 reported
- None are prolific gamma emitters
- Ratio of decay product to head of chain ranged from an impossible 1.11 to 0.64
- Average was 0.85 = 60 years since separation
- Seemed too old



9

Gamma lines

- Pa-231
 - 300 keV, 5 %
- Th-227
 - 236 keV, 12 %
 - 256 keV, 7 %
 - 300 keV, 4.5%
 - 330 keV, 4 %
- Ra-223
 - 144 keV, 3.2 %
 - 154 keV, 5.6 %
 - 269 keV, 13.7 %
 - 323 keV, 3.9 %
 - 338 keV, 2.8 %
- Note the overlap between the head of chain and Th-227

10

Second analysis

- Th-227/Pa-231 = 0.54 ± 0.04
- = 21 to 27 years
- Ra-223/Pa-231 = 0.69 to 0.76
- = 37 to 45 years
- Range reported = 21 to 60 years!
- Probably conservative value used for monitoring assumed an ingrowth of 65 % = 33 years
- Beta response = $0.65 \times (0.91 + 0.997) \times 0.8 \times 25$
 $\text{cps/Bq/cm}^2 \text{ Pa-231} = 25 \text{ cps/Bq/cm}^2$

11

Instrument limiting count rates

- Each material thickness and density was assessed
- A decision was made on whether the material would be single or double sided contaminated
- Limiting value calculated on 5 Bq/g average activity
- Example
 - **Urastone duct**
 - **4 g/cm²**
 - **Single sided**
 - **Surface activity limit = 20 Bq/cm²**
 - **Instrument response used = 19 cps/Bq/cm² Pa-231**
(conservative, in case ingrowth was lower than we thought)
 - **Limit = 380 cps**

Soft waste

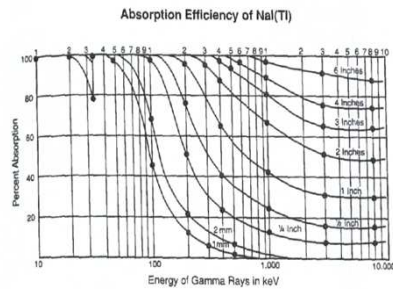
- Any asbestos clean-up produces soft waste
- Coveralls, gloves, filters etc.
- Monitored as a bulk material in plastic bags
 - Taken as 40 cm in diameter x 30 cm deep
 - Density of 0.2 (typical for soft waste)
- Detector selected was a Mini Instruments (now Thermo) 44B
- 2 mm thick, 32 mm diameter sodium iodide scintillator



12

Calculated response

- Response calculated on the basis that all the photons would escape
- Low Z material
- Most interactions are Compton
- Decrease in photon numbers balanced by the increase in detection efficiency for the lower energy
- (Cs-137 source covered by 10 cm of sand gives the same count rate as an open source!)



- 14.8 Bq/g Pa-231 and 65 % ingrowth gave 34 cps above a typical background of 5 to 10 cps
- Monitoring results were expected to be well below the limit

13

Soft waste bag results

- No need to add any statistical offset as each waste mass was small
- Provided the bags mainly had trivial levels of activity then the bulk activity would clearly comply

Item Reference	Mass (kg)	Material types Swabs/vacbags etc.	North	South	East	West	Top	bottom	Average CPS
545	10	Coveralls	Nil	Nil	Nil	Nil	Nil	Nil	Nil
46	3	Coveralls	Nil	Nil	Nil	Nil	Nil	Nil	Nil
47	4	Mixed	10	20	10	20	30	70	25

14

15

Conclusion 1

- Virtually all the solid materials complied with the PSRE limits after, at most, minor cleaning such as wiping and vacuuming
- All the soft waste complied
- Some of the vacuum cleaner bags were low level radioactive waste

S 65	10	40	60	200	200	1K	2K	Filters	Vac BAG
S 66	10	NIL						mixed	

May 2015

Radiation Metrology Ltd

Problem 2

- We decided to look at the problem of over-painted plutonium contamination
- The alphas don't make it through a layer of paint
- X-rays and gamma rays may do
- So what's left?
 - **10-21 keV x-rays from Pu alpha emitters and Am-241 ingrown from Pu-241**
 - **26.3 and 59.5 keV gamma rays from Am-241**

1
6

FIDLER probe

- Field Instrument for Detection of Low Energy Radiation – FIDLER
 - Designed for detection of low energy (5-150 keV) photons
 - Thin slice of NaI(Tl)
 - Relatively insensitive to higher energies
 - Therefore fairly low background
 - Thick glass light guide coupling it to a big photomultiplier
 - Makes it a bit on the heavy side
 - Gives good performance for spectrometry



1
7

Spectra - spectrometry or not?

- We connected the FIDLER up to an MCA (multi-channel analyser)
- This allowed us to see the energy distribution of the signal and the noise
- In understanding the problem and setting regions of interest looking at a spectrum was very helpful.
- Discriminators could be used to set a single region of interest.

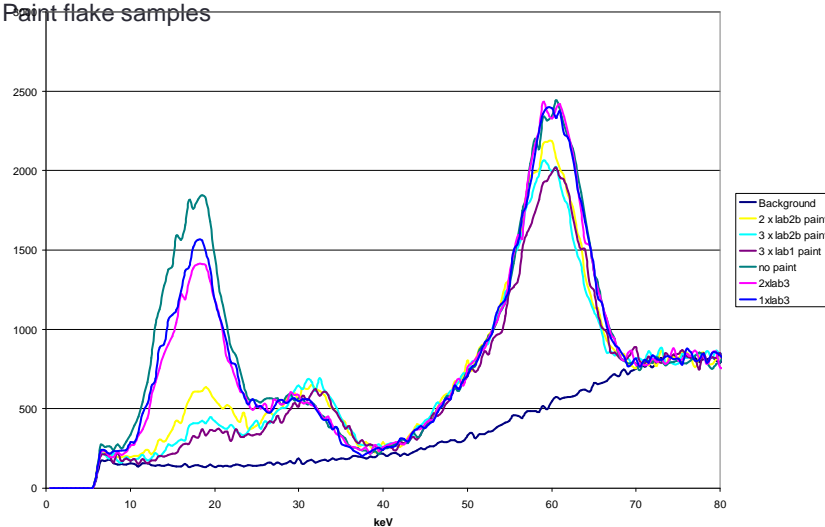
19

Setup

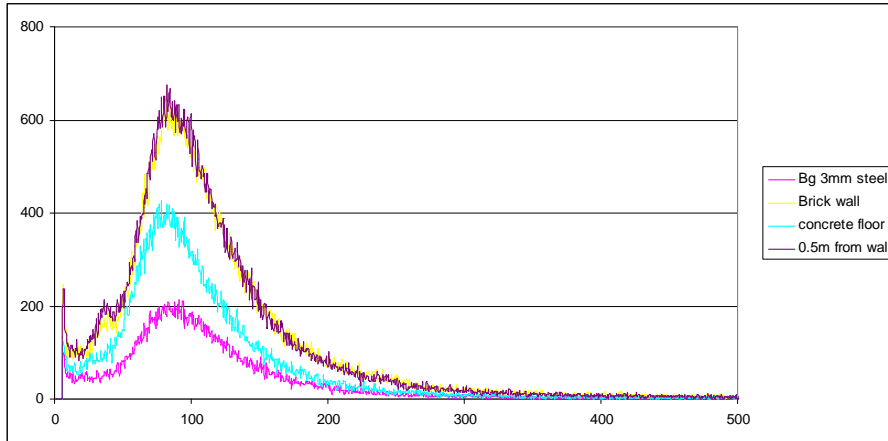
- 2 sets of measurements using an Am-241 source
- Real paint samples from a facility
 - Paint samples were removed from a building that had been in use for approx. 50 years
 - Paint samples used as absorbers in front of an Am-241 point source
 - The composition of these samples was determined using X-ray fluorescence analysis
- Modern leaded paint
 - Modern leaded paint was painted onto paper and a pvc sheet
 - This was then used as an absorber in front of an Am-241 point source
 - The composition of this paint was obtained from the manufacturer
- Background measurements
 - Backgrounds were taken on a variety of substrates

Spectra

- Paint flake samples



Background spectra



2
1

Limits of detection Am-241

- Using Currie's method, we calculated the maximum missable activity on a brick wall for a point source directly under the detector and for activity per unit area over the probe area.
- Two counting windows

Energy range	Counting time s	Bq cm-2	point source Bq
10 – 80 keV	1	1	120
	100	0.1	10
40-80 keV	1	2	240
	100	0.2	20

2
2

23

Application

- Low energy X-rays are often attenuated
- Through non-lead based paints there is very little attenuation of the 59.5 keV gamma
 - The limits of detection quoted for the 40-80 keV range would therefore be applicable for contamination under such paint.
- Through lead based paints there is attenuation of the 59.5 keV gamma
 - Detection is clearly possible but the thickness and composition of the paint would be needed to estimate activity.
- As there is very little attenuation of the signal by air
 - The detector could be moved away from the surface to monitor a larger area in one measurement

24

Alternative detectors

- We chose the FIDLER as it offers good spectral performance and good portability
- Other lighter NaI detectors are available with poorer performance, but with advantages such as lower weight.
- The fundamental requirements are a large area, a thin window and a thin (2 mm maximum) crystal
- High resolution HPGe systems can also be used, offering advantages in resolution but disadvantages in price and weight.

25

Conclusion

- X and gamma measurements can be very useful in dealing with Pu and Am-241, but you have to understand any paint that might have been painted over the contamination

Problem 3, Winfrith EAST building

- Built to treat 300 m³ of sludge from the SGHWR
- Correctly, the Steam Generating Heavy Water Reactor
- Britain's version of the Candu
- Very high maintenance, very high operator doses



A relatively new building

- Built 2001 to process radioactive sludges from existing concrete storage tanks
- The sludges were agitated, filtered and concentrated prior to encapsulation in a cement matrix within 500 litre stainless steel drums.
- Only a very small number of minor incidents inside the cell line
- Finished sludge encapsulation in April 2010
- Much considered to be “clean”




Active areas and components

- Input piping
- Mixing tanks
- Dirty side ventilation
- Some equipment transferred from other buildings
- Removed as far as possible early in the project
- Reducing the chance of cross-contamination and background increase



29

Aim – to demonstrate that the material is indistinguishable from background

- Conventional monitoring involves a ratemeter based measurement and the user's judgement
 - Background or not background?
 - Difficult to quantify
 - Difficult to audit
 - Poor sensitivity
- 
- This project uses integrated counts over a defined area
 - Easy to calculate statistical uncertainty

30

Point of suspicion

- Growing in popularity as an idea in the UK
- Numerically the same as Currie Critical Limit
- But easier language
- “Do we think this might well not be background?”
- Prompts an action – remeasure and average
- This project the point of suspicion is set at the mean + 2 sigma
- Expect 1 measurement in 40 will exceed

31

Approach

- Identify a fingerprint
- Choose an instrument and detector
- Work out the response to the fingerprint
- Identify clean reference samples for each object category
- Identify a low background area
- Perform a 100 second count
- This becomes the reference for that sort of object and the next 2 hours
- Look up the Point of Suspicion on a spreadsheet
- Any measurement that equals or exceeds that, repeat and average
- If still above the Point of Suspicion, deem it contaminated

32

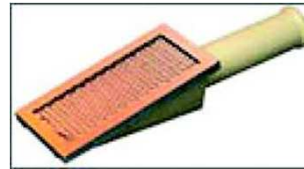
Fingerprint

- A mixture of activation products and fuel leakage
- Modified by the pond water treatment process
- Established by measurement of the sludge
- Only 0.06 % alphas
- 11 % Ni-63 – very difficult to detect beta
- 60 % Cs-137- medium energy beta
- 22 % Co-60 – low energy beta
- 4 % Fe-55 – low energy X-ray
- 3 % minor contributions
- **The Cs-137 is easy to detect**

33

Instruments used

- Larger, flat areas – Thermo DP6 dual phosphor scintillator
 - Convenient area – 100 cm²
 - Good alpha and medium to high energy beta efficiency
 - About 40 % for alpha and 50 % for beta, 2 π
- Connected to an Electra ratemeter – dual channel, selectable beta channel width and integrating time
- RSRL standard instruments



34

Performance of the DP6 for direct measurement

- Averaged over the probe area
- DP6 beta fingerprint 12 cps/Bq/cm²
- Maximum acceptable beta background = 6 cps
- For a 30 second count, background mean = 180 counts
- 1 sigma (σ) = $\sqrt{180} = 13.4$ counts = 0.45 cps
- MMA corresponds to $\approx 4 \sigma = 1.8$ cps
- Corresponding fingerprint activity = 0.15 Bq/cm²
- For all barring thin material, this would meet the then SoLA Exemption Limit of 0.4 Bq/g

35

Surveyors reaction

- Surveyors initially resisted the approach
- They didn't like the idea of integrating over significant areas having spent their careers surveying small areas by ear
- "But what if all the activity is in one spot"
- Good question – if it is, then ALARP suggests we remove it
- So keep listening – if there seems to be an unusual count rate, check it's real
- If not, just start again!

36

Operations for objects

- Each object had a unique reference number
- Description recorded – material, form etc
- Measured and weighed
- Select a suitable background object
- Measure the background over 100 seconds
- Look up the corresponding point of suspicion on a spreadsheet
- Divide the object surface into convenient areas, up to about 0.5 m² for large, flat things
- Make the measurement
- Record the result
- If it complies, into the acceptable pile for disposal as exempt waste

37

Operations for buildings

- Basically the same approach
- Identify areas likely to be clean – no history of spills etc
- Check the count rate is basically the same over the walls
- Check it's the same over the floors
- Check it's the same over the steel doors
- The major confounding factor were the door apertures
- Rooms with small door area had higher backgrounds
- K-40 in the building material, checked with an Exploranium GR-135

38

Result

- **All the material expected to be clean complied with the criteria**
- Not as expensive as some people imagined
- Possible improvement would be to use a beta detector with a thinner scintillator.
- This reduces the background and, hence, the monitoring time for a given MMA
- Now we'd work to EPR, which uses nuclide specific Out of Scope Levels
- This procedure would still be sufficient

39

Acknowledgements

- Example 1: Clare Irving, Centurion RPA
- Example 2: Rob Clarke, Mike Davies, Nuvia, Ian Croudace, GAU
- Example 3: Michelle Skelland, then of RSRL, the lady who turned the theory into a result

40

Currie – the man who brought sense to this sort of measurement

- Limits for qualitative detection and quantitative determination. Application to radiochemistry
- [Lloyd A. Currie](#)
- *Anal. Chem.*, 1968, 40 (3), pp 586–593
- DOI: 10.1021/ac60259a007
- Publication Date: March 1968