Surveillance of Surface Radioactivity – The Role of Liquid Scintillation Counting.

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Waste Management Technology- Analytical Services

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Waste Management and Technology

Waste Management Technology (WMT) is a leading provider of specialised radioactive waste management services. Services include:

- **Waste management assessments and consultancy studies**
- **Waste sampling and characterisation**
- **Advanced waste treatment processes**
- **Recycling, recovery and re-categorisation (e.g.detritiation)**
- **Waste packaging and immobilisation**
- **Impact and fire testing of transport packages**
- **Radioactive materials transport**
- **High temperature chemistry**
- **Waste management plant optimisation, design review and validation**
- **Analytical and Radiochemistry**

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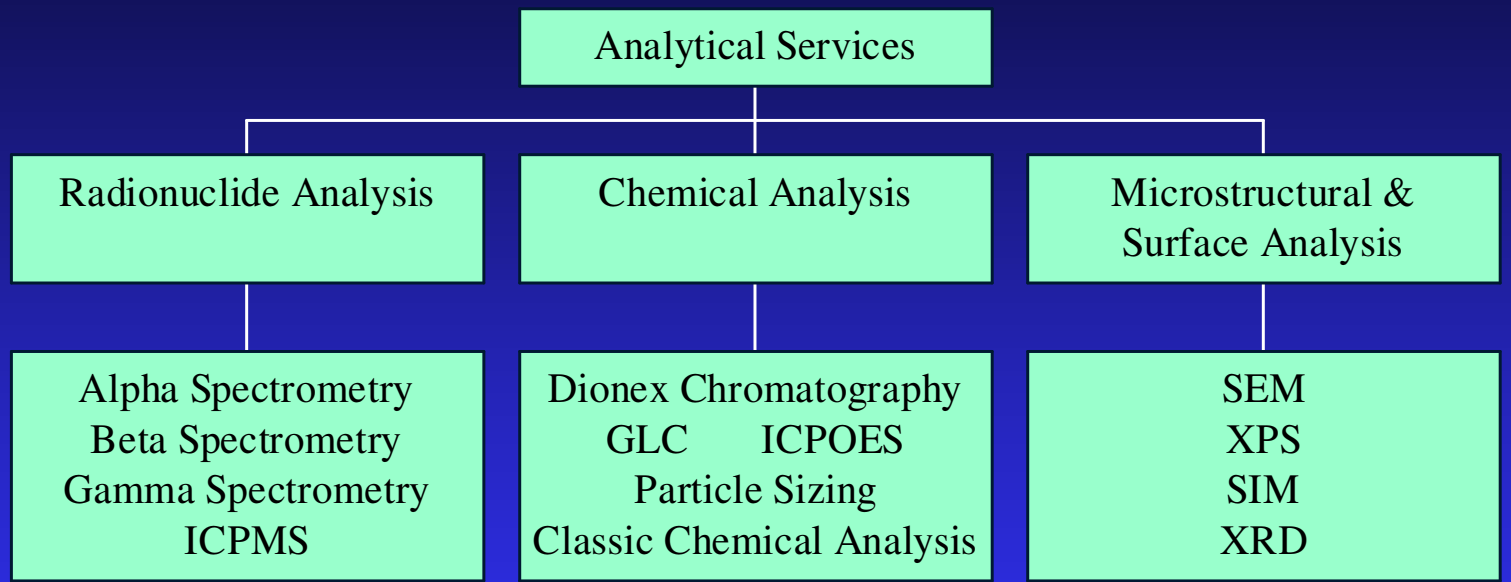
WMT – Analytical Services



The service focus is on providing analytical support to nuclear site decommissioning and operations, primarily radionuclide analysis supported by chemical, physical and microstructural analysis capability.

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The surveillance of surfaces contaminated with radioactivity is achieved essentially by :

- Direct monitoring with health physics monitors (scintillation probes, Geiger-Muller probes etc)

- The swabbing of known surface areas followed by assessment of the wipes for radioactivity:

  - By direct measurement, e.g., gas-flow proportional counters, gamma spectrometry

  - **Liquid scintillation Counting – generally for soft beta contamination e.g., Tritium or Carbon-14**

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Basic Approach for the Assessment of Soft Beta Contamination

1 Swab a known area of the potentially contaminated surface with a filter paper wipe

2 Place wipe in a liquid scintillation cocktail and count!

Straightforward and quick – therefore cheap!!

BUT IS IT ACCURATE?

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NO! NO! NO! NO! **NO!**

"Monitoring by wiping is one of the most uncertain measurements used in a scientific area where relative approximate measurements are the norm"

P Burgess "Summary of Pick-Up Efficiencies from Wipes" 8-12-05

Why?

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Pick-Up Factors

Depends on:

- Surface condition – smooth or rough (glass or brick)
- Type of material used to wipe the surface
- Dry or wet wipe? Solvent used?
- Chemical nature of the contaminant, I.e., affinity with the chosen wipe system (solvent)
- Actual area wiped, I.e., “ploughing a furrow” through the deposited material over a specified area.

1950's – R Dunster applied logic to the problem and decided that a reasonably pessimistic wipe-efficiency was 10%, this was widely accepted by the industry and is used today.

Numerous workers have since performed a number of experimental studies in an effort to further quantify this parameter for a variety of surfaces, wipe-systems, and contaminants.

1967 – Prince and Wang showed that for fibre-glass/"shell-stone" surfaces dry paper wipe removed 5-30% ^{3}H-Paraffin/^{3}H-acetate, wet wipes gave 5-32%.

1967 – Royster and Fish deposited 1 micron ThO_2 particles on various materials:

Fibreboard	"normal wipe"	24%
Perspex	"normal wipe"	71%
Concrete	"sticky paper"	44%
Painted Al	"sticky paper"	86%

1967 – Mitchell and Eurtsler deposited Be dust on wood, dry wipe removed 3% (21% of material removable by wiping and/or washing).

1989 – Kawano and Ebihara reported 13-90% for floor tile contaminated with "radiopharmaceuticals" using dry paper wipes.

1992 – Frame studied the issue and concluded that a wipe factor of 10% `was supportable in those situations where there is the absence of any site specific information.

1993 – Campbell contaminated a variety of surfaces (wood, paint, stainless steel, Formica, painted and unpainted lead) with either ^{3}H-thyimidine or ^{3}H-triolein. Dry wipes were on average 3% efficient for all surfaces, but this depended heavily on the surface condition, water gave 34-47%, methanol 16-23%.

So Dunsters "rule-of thumb" of 10% seems to be reasonably pessimistic and applicable to the real situation

But there are also uncertainties with respect to the detection system employed

Liquid Scintillation Counting Uncertainties.

Effect of source geometry on counting efficiency

Source Self-Adsorption – Filter medium “transparency”, degree of surface material on filter.

Chemical-Colour Quenching:

- Wipe condition – degree of soiling
- Solvents employed

Campbell et al (1993) and other workers have stated that quenching of greater than 20% is the norm.

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The WMT-AC Approach for The Assessment of ^{3}H Surface Contamination

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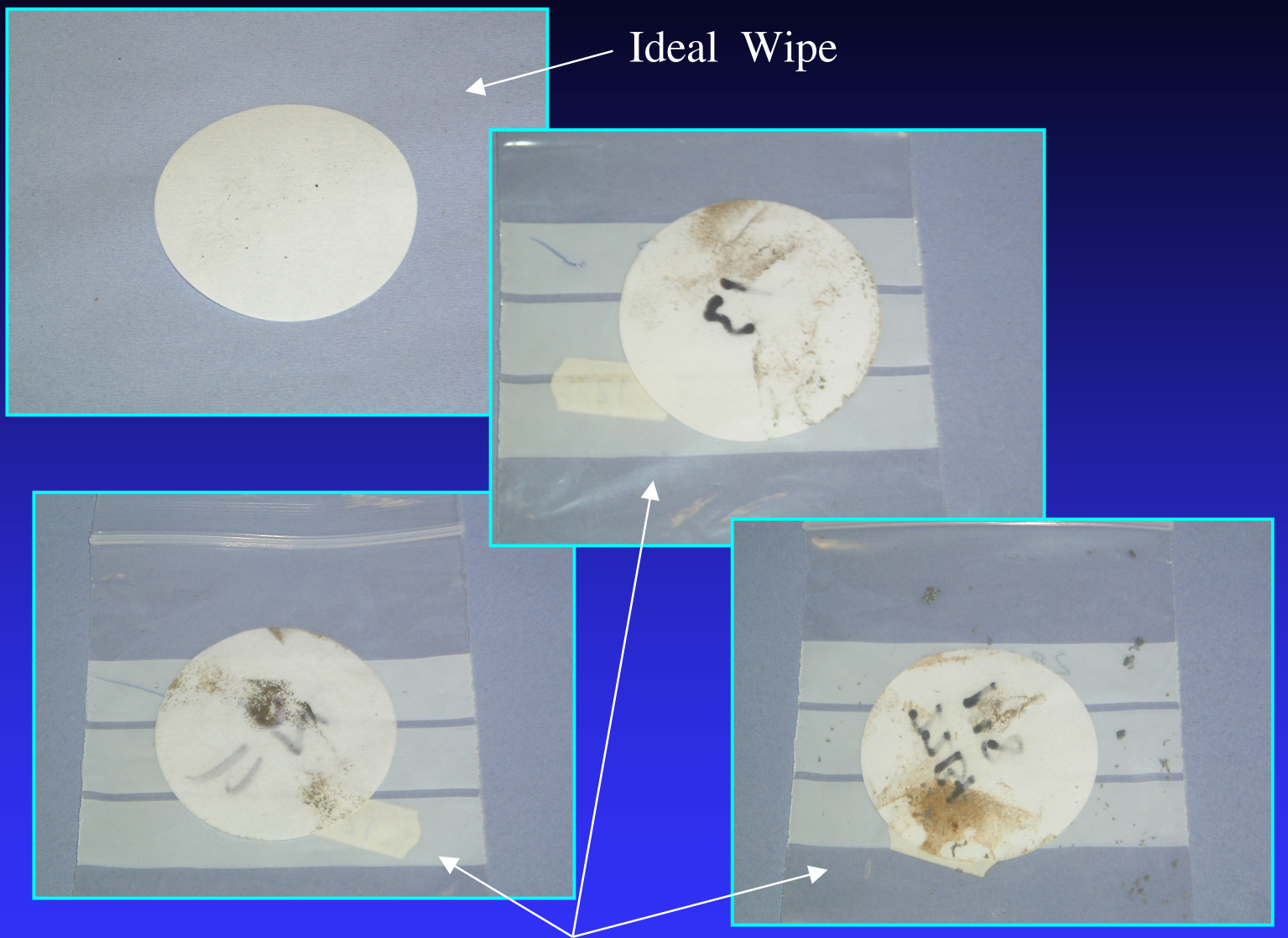
First Things First - Sampling

Specify wipe medium – 5.5 cm diameter filter papers preferred

If possible these are supplied by ourselves or we ask for at least 10-15 “clean examples” for preparation of blanks, quench curves

Require the filters to be despatched in labelled self-sealing plastic bags to preserve the analyte etc, where the filters are folded neatly in half to protect the “exposed surface”.

Specify that the filter paper is not to be marked with biro or felt-tip pen



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Source Preparation

Wipes that are too heavily soiled are rejected and analysed by an alternative method (combustion).

To ensure a reproducible source geometry the paper is folded in half (if this has not already been done) and positioned against the side of the vial as shown.

A Quench curve is established – colour and chemical quenching

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Add scintillant, and leave to dark condition

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Oh dear!

5 hrs

The curse of the marker pen strikes again

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So how does it perform?

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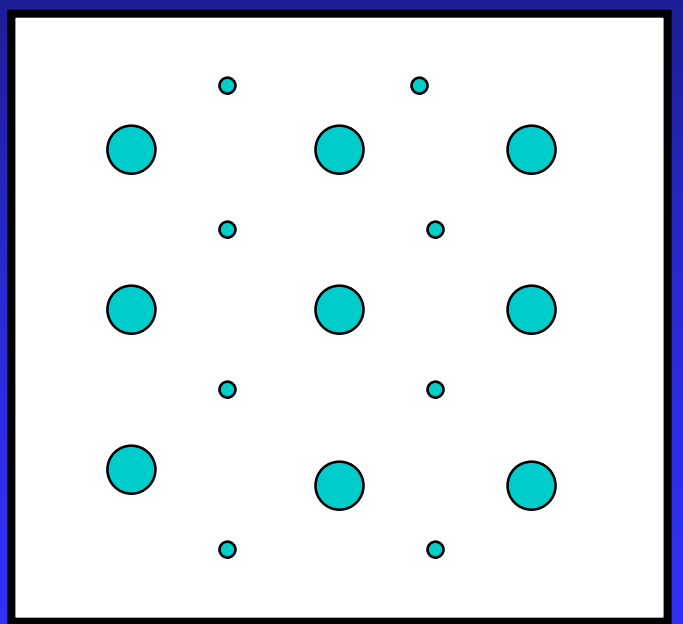
Method Test

Three sets of five un-patterned smooth porcelain tiles (156.25 cm^2) were each contaminated with 262 Bq of Tritiated Sucrose applied as a solution in demineralised water



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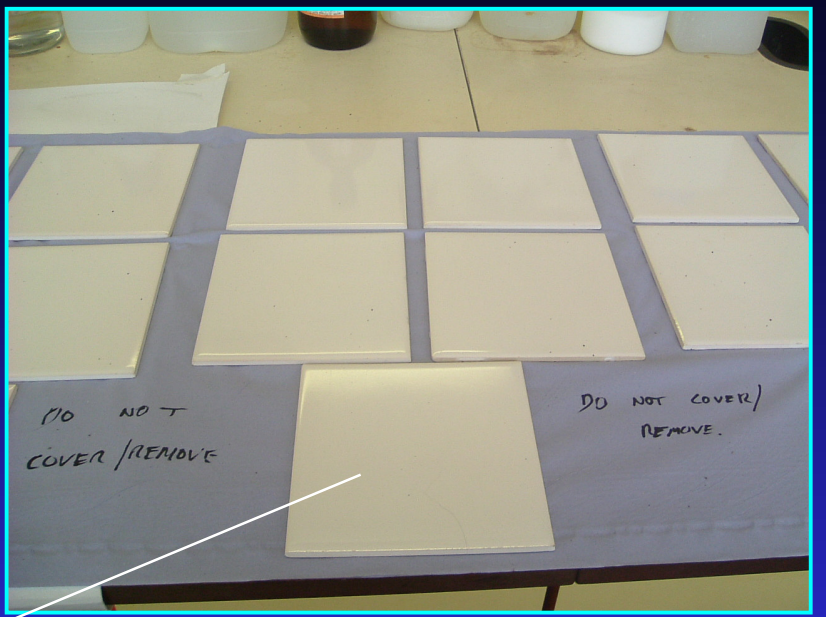
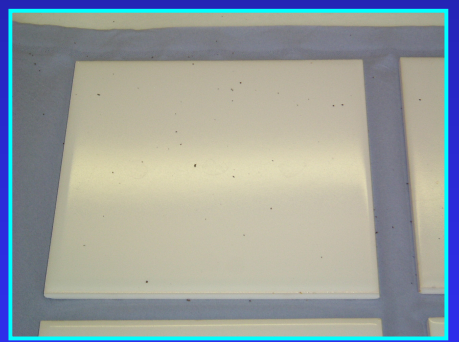
Each set of five tiles were wiped once by the same operator with 5.5.cm diameter Whatman Type 541 filter papers.

- One set were swabbed with dry papers
- One set were swabbed with water damped papers
- One set were swabbed with methanol damped papers

The prepared sources were then prepared as sources for liquid scintillation counting on an LKB-Wallac 1411 liquid beta scintillation counter





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Results

Sample ID	[^{3}H] dpm	SQP(E)	Using Dunsters 10% Wipe Factor: Implied [^{3}H] per tile	Implied Bq.cm^{-2}	Actual % Wipe Factor
Dry-1	772.4	791	129 +/- 3	0.8	4.9
Dry-2	485.9	793	81 +/- 2	0.5	3.1
Dry-3	601.1	792	100 +/- 2	0.6	3.8
Dry-4	1340.4	792	223 +/- 3	1.4	8.5
Dry-5	1107.3	793	185 +/- 3	1.2	7.0
Water-1	7320.8	786	1220 +/- 7	7.8	47
Water-2	7994.0	785	1332 +/- 8	8.5	51
Water-3	6749.8	785	1125 +/- 7	7.2	43
Water-4	6268.8	786	1045 +/- 7	6.7	40
Water-5	6326.9	786	1055 +/- 7	6.8	40
Methanol-1	2425.4	790	404 +/- 4	2.6	15
Methanol-2	2091.0	790	349 +/- 4	2.2	13
Methanol-3	2636.7	791	440 +/- 5	2.8	17
Methanol-4	2131.0	790	355 +/- 3	2.3	14
Methanol-5	3149.4	791	525 +/- 5	3.4	20

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But what if other species are present in the real condition?

Always interrogate the spectrum.

If necessary set windows manually to exclude interference, and assess sample specific % counting efficiencies by direct spiking and recounting – somewhat negates the “cheap and quick” argument, but still cheaper than alternatives such as combustion techniques.

In Conclusion

The technique as applied to ^{3}H monitoring is semi-quantitative at best, in most cases it is appropriate for monitoring relative levels of contamination – "Yes or No" conditions and will give a reasonable estimate of the degree of contamination

However, it is not defensible to use this technique to establish whether or not an object can be disposed of as "free-release" unless

- The limit-of-detection is sufficiently low to ensure confidence that the contamination is below the required criteria
- An appropriate upper value is designated above which repeat assessment/decontamination is performed, I.e.,

Assumed % Wipe Factor	Implied contamination level ($Bq.cm^{-2}$) for an observed level of 2 $Bq/100\ cm^2$
10	0.20 +/- 0.02
8	0.25 +/- 0.03
5	0.40 +/- 0.05
2	1.00 +/- 0.10

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