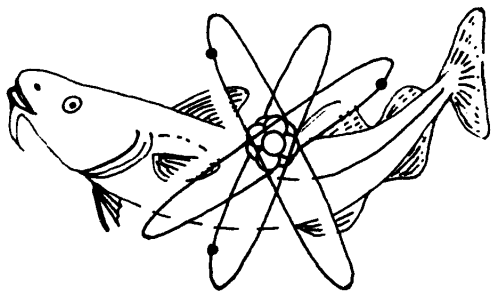
MINISTRY OF AGRICULTURE, FISHERIES AND FOOD

FISHERIES RADIOBIOLOGICAL LABORATORY



GROSS BETA COUNTING OF ENVIRONMENTAL MATERIALS

BY

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TECHNICAL REPORT FRL 3

LOWESTOFT
SUFFOLK

OCTOBER
1968

We are following a policy of making known to all concerned, not only the results of our monitoring of radioactive wastes discharged to surface waters and to the sea, but also the details of the methods used in evaluating these discharges. In this leaflet John Dutton deals with the question of gross beta radiation and its measurement, describing the techniques which have been developed at the Ministry's Fisheries Radiobiological Laboratory.

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1 Introduction

Most of the radioactive discharges authorized from sites in Great Britain contain beta-emitting isotopes, and the measurement of gross beta activity in environmental samples will, in many cases, enable a simple and rapid check to be made on the degrees of contamination. Two methods of measuring beta activity in environmental samples are in current use in this laboratory. The first is the "infinite depth" technique[1] in which a source presented to the detector (a 1 inch diameter GM tube with end-window of thickness 2 mg/cm^2) is indistinguishable from one of infinite depth in terms of beta emission; a source depth of 1.9 cm has been chosen, because the highest-energy beta emissions encountered in our environmental monitoring programme are the 3.6 MeV particles emitted during the decay of ruthenium/rhodium-106. The dried sample is packed into a cylindrical cardboard container and its count rate measured and compared with that of a potassium chloride standard, identically mounted. The activity of the sample is expressed as "potassium-40 beta equivalent pCi per gramme". The use of naturally-occurring potassium-40 as the standard is due to the fact that this nuclide constitutes the major portion of the radioactivity present in many of the samples.

A serious disadvantage of the infinite-depth method is that it discriminates against the lower-energy beta-emitting isotopes which make up the major portion of the discharges from the nuclear-powered generating stations operated by the Central Electricity Generating Board and the South of Scotland Electricity Board; this discrimination arises from the greater degree of absorption of the lower-energy beta particles in the source and, to a smaller extent, in the detector end-window. The use, as a standard, of a radioisotope whose β_{max} energy more closely matches the average energy of the radioisotopes discharged has been considered, but this does not in any way reduce the discrimination of the method against low-energy beta emitters, and ideally a method which is to measure the degree of contamination of the environment should be independent of the energy of the emission. An alternative gross-beta counting technique has therefore been introduced which, although not ideal, is certainly an improvement in terms of response to the energy of beta particle emission. This method involves the preparation of a thinly spread source (200 mg over 100 cm^2) whose count rate is compared with a similar potassium

chloride standard using a Beckman Widebeta II automatic proportional counter; the detector is 5 inches in diameter with an end-window of thickness 500μg/cm^2. Apart from its closer approach to an ideal response, the use of the automatic counter has obvious advantages over the older, manually-operated, equipment.

The discrimination against low-energy beta emitters by infinite-depth counting has been useful in certain cases: it provided a very simple check method for the presence of ruthenium-106 in certain materials in the marine environs of the U.K.A.E.A. fuel-processing plant at Windscale in Cumberland. This radioisotope is the critical isotope in the discharge, due to its high reconcentration in the seaweed Porphyra. Smaller amounts of other radionuclides are present in the weed: zirconium-95, niobium-95 and ruthenium-103 - all low-energy beta emitters - together with an almost constant amount of potassium-40 which has a β_{max} energy of 1.32 MeV. Cerium/praseodymium-144, a high-energy beta emitter, is not generally present in any significant quantity in the weed. When assayed by the infinite-depth method, therefore, the only significant counts arose from potassium-40 and ruthenium-106; thus, after correcting for the small amount of potassium-40 present in the sample, the activity remaining was due to ruthenium-106.

The change from infinite-depth to thin-source counting has meant that this check method is no longer applicable. It is, however, possible to substitute the external absorption facility available with the Beckman instrument for the self-absorption of the infinite depth source. The use of an external absorber of about 50 mg/cm^2 absorbs over 90 per cent of the low-energy beta emitters mentioned above, yet transmits about 70 per cent of the ruthenium/rhodium-106. The potassium-40 contribution is subtracted from the measured count rate and the resultant count rate is compared with a ruthenium-106 standard and expressed as "picocuries ruthenium-106 per gramme".

The relative response of each of the three counting methods to varying β_{max} energies is shown in Figure 1.

Details of the source preparation techniques are given in the appropriate sections of this report, but some general comments are necessary at this stage.

The radioactivity of silts and sediments may be adsorbed on the surface of these materials, and therefore the specific activity of particles in the original sample often increases with decreasing particle size. The preparation techniques described later involve grinding and sieving the sample, and it is therefore important to ensure that the whole of the portion selected is ground to pass through the sieve, because otherwise an unrepresentative sample may be counted.

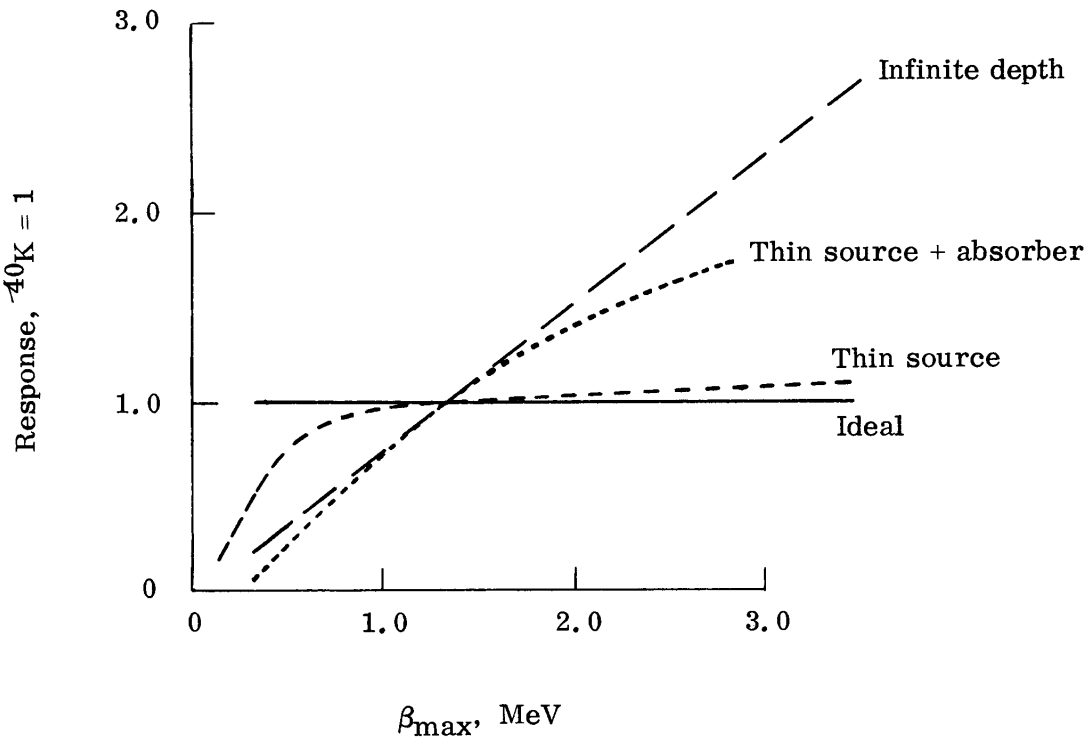

Figure 1 The relative response of different counting methods.

Some of the material for measurement is fibrous or oily (or both, e.g. the flesh of salmon and herring) and it is almost impossible to sieve it, especially for the thin-source count. In these cases infinite-depth counting is used.

The infinite-depth counting method is based on standard radiometric practice; the development of the thin-source counting method, in its application to environmental samples, was carried out at this laboratory by E. Reynolds (electronics) and J. W. R. Dutton (analysis).

2 Infinite-depth beta counting

2.1 Summary

The dried sample is assayed by measuring the beta activity of a 1 inch diameter infinite-depth source, using a 1 inch Geiger Müller tube, and comparing the count rate with that from a potassium chloride source identically mounted. The activity is expressed as "potassium-40 β equivalent pCi/g", dry for sands and sediments, and wet for other materials. The counting assembly, shown diagrammatically in Figure 2, is standard equipment.

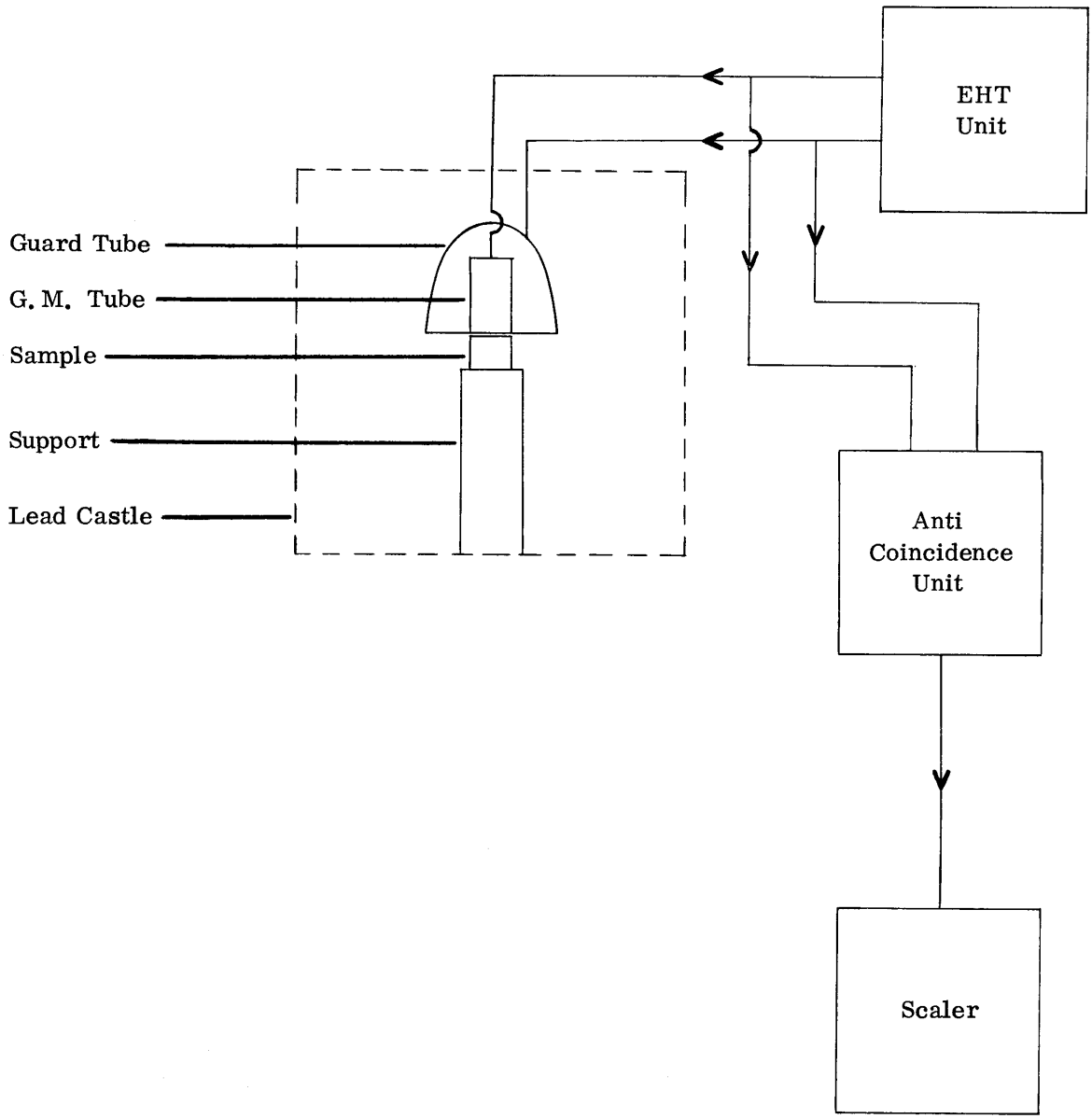

Figure 2 Schematic diagram of infinite-depth counting assembly.

2.2 Calibration

2.2.1 Preparation of the standard

About 30 grammes of analytical reagent grade potassium chloride are dried at 105-110°C and ground to pass though a BS60 sieve (250 μm). About 10 grammes of the dried material are then packed uniformly into a small cardboard container (2.8 cm diameter x 1.9 cm depth), the upper surface being level with the lip of the container.

2.2.2 Setting-up procedure

With the potassium standard in position, the EHT voltage is adjusted until no counts are registered. The voltage is then increased by 10V increments, counting the standard after each increment for five minutes, until the EHT is 150 volts above the starting voltage. A graph of counts obtained against applied EHT volts is plotted. The operating voltage is chosen on the plateau, 50 to 100 volts above the knee of the curve.

2.2.3 Energy response

Because the sources are thick, the degree of absorption of beta particles in the sample counted varies significantly with their energy. The response of this type of counting assembly to various isotopes is shown in Table 1.

Table 1 The response of infinite-depth counting equipment to various isotopes

Isotope	β_{max} MeV (major)	Efficiency	
		cpm/pCi per g dry	cpm/pCi, $^{40}K = 1$
$^{106}Ru/Rh$	2.0-3.6	0.53	2.3
$^{144}Ce/Pr$	2.98	0.60	2.6
^{40}K	1.32	0.23	1.0
^{137}Cs	0.51	0.09	0.4
^{60}Co	0.31	0.06	0.3
^{54}Mn	-	0.01	0.04
Background	1 to 1.5 cpm		

2.3 Measurement of sample activity

2.3.1 Preparation of the sample

About 30 grammes of the dried sample are ground to pass through a BS60 sieve. About 10 grammes of the sample* are then packed into a 1 inch diameter container as in paragraph 2.2.1.

*e.g. Porphyra, 7 g; Fucus, 10 g; mussels, 7 to 8 g; sediment, 7 to 8 g; sand, 14 to 15 g.

2.3.2 Sample counting

The background count rate of the equipment is measured using an empty container (= B cpm), and the sample and standard are counted to the statistical precision required (= N and M cpm respectively). After correction of the count rates for background, the activity of the sample is expressed as potassium activity giving rise to β radiation, using the formula:

$$^{40}\text{K } \beta \text{ eq. pCi/g dry} = \frac{N - B}{M - B} \times 3.9 \times 10^{2}.$$

The results are converted to wet weight if required.

The activity of potassium chloride is taken as 3.90×10^{2} pCi/g; this is the activity giving rise to the emission of beta particles.

2.4 The determination of ruthenium-106 activity in laverbread

The potassium-40 equivalent activity is calculated for the dry sample as in paragraph 2.3.2, and then converted to activity per gramme of wet material. The contribution from natural potassium is then subtracted (2.5 pCi/g is an average value for laverbread), and the nett potassium-40 equivalent activity is converted to ruthenium-106 by correcting for the overestimation of that isotope (a factor of 2.3, see Table 1):

$$^{106}\text{Ru pCi/g wet} = \left[(^{40}\text{K } \beta \text{ pCi/g dry} \times D) - 2.5 \right] \times \frac{1}{2.3}$$

where D is the dry:wet weight ratio of the sample.

3 Thin-source beta counting

3.1 Summary

The dried sample is assayed by measuring the count rate of a 200 mg source spread over 100 cm^2, using an automatic 5 inch proportional gas-flow counter, and comparing it with that from a potassium chloride source identically mounted. The activity is expressed as "potassium-40 β equivalent pCi/g", dry for sands and silts, and wet for other materials.

3.2 Automatic counter*

The counting instrument in use at this laboratory is a 5 inch gas-flow detector operating in the proportional region. An automatic

*Beckman Widebeta II supplied by Beckman Instruments Limited.

sample-changing device will accept 60 $4\frac{1}{2}$ inch diameter planchets, which are stainless steel trays 0.0148 inch thick (28 SWG) with a 1/8 inch high lip; the thickness is sufficient to ensure saturation backscatter. The trays should be free from flaws causing bowing and dishing. A sample of the steel to be used for the trays should first be obtained to ensure that it is not contaminated with cobalt-60 or other radioactive impurities sometimes found in stainless steel. A simple schematic diagram of the counting apparatus is shown in Figure 3.

3.3 Setting-up and operating procedure

Because the detector is operated in the proportional region, considerable care must be taken in selecting the EHT so that the detector is operating on the plateau for all the beta-emitting isotopes present in the sample. The settings for the detector, guard and alpha discriminators are governed by the operating requirements, and no settings are given in this report. The operating modes are set out in Table 2.

Table 2 Operating modes for the thin-source counting equipment

Switch	Setting
Preset minutes	200*
Preset counts	10^{4}*
Count mode	Nett
Count reject	Off
Automatic background subtractor	Off
Detector selector	Upper detector
Control selector	System control
Analyser mode	β plateau
Plateau point	Operate
α discriminator	On
Detector type	Window detector
Gas flush delay	Inoperative
Live time gate	On
Print out	Teletype**

* These settings can be varied to obtain the required precision.
**A modified Addo machine is used at this laboratory.

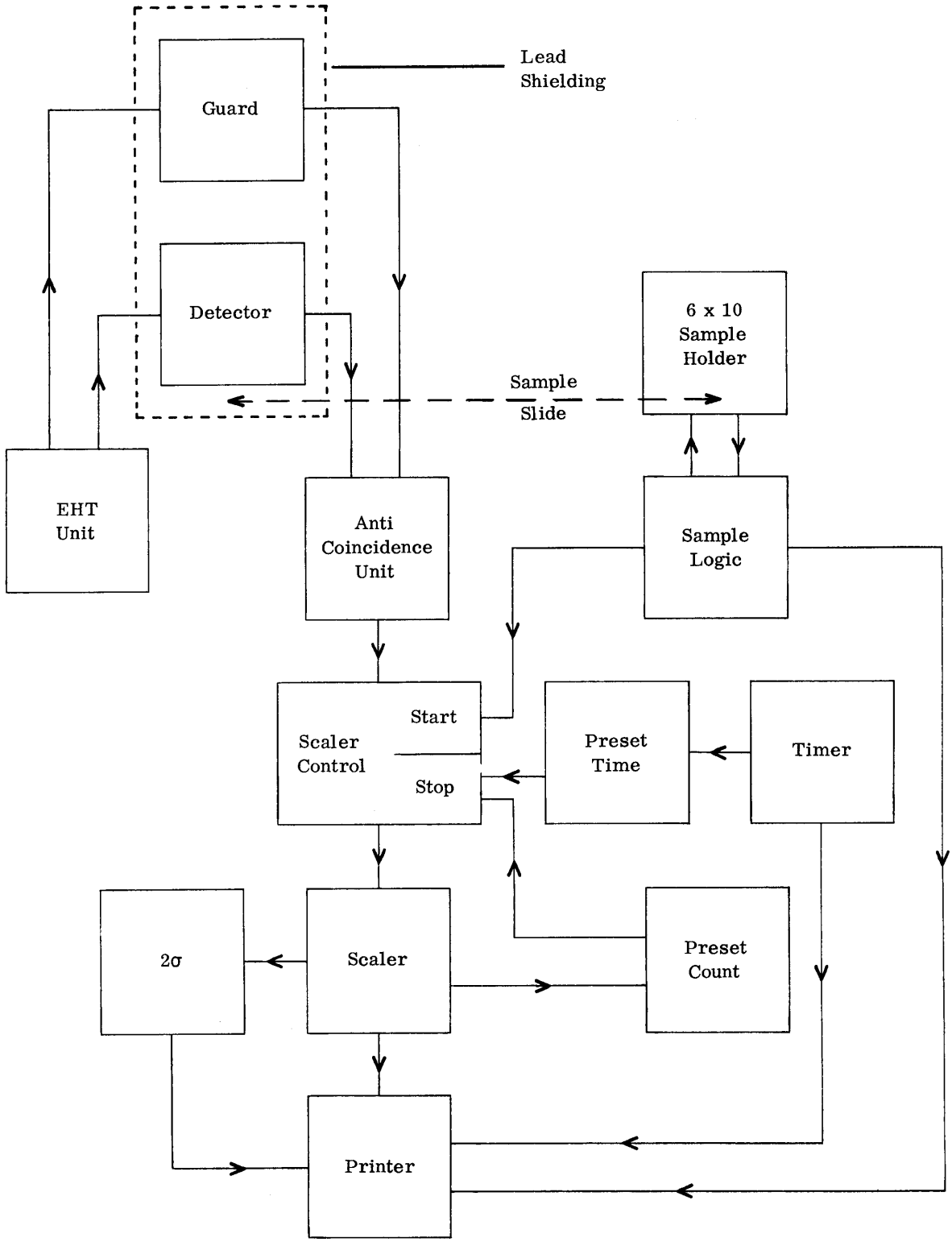

Figure 3 Simplified schematic diagram of thin-source counting assembly.

3.4 Preparation of the standard

About 10 grammes of analytical reagent grade potassium chloride are dried at 105-110°C and ground to pass through a BS240 sieve (66 μm); 0.2 grammes of the dried material are then placed in the centre of a $4\frac{1}{2}$ inch diameter stainless steel tray and a small volume of acetone added. The tray is swirled gently to spread the sample evenly and allowed to stand on a level surface until the acetone has evaporated. Six such standards are prepared.

3.5 Preparation of the sample

About 10 grammes of the dried sample are ground to pass through a BS240 sieve; 0.2 grammes of this material are taken and a source prepared in the same way as the potassium chloride standard (see paragraph 3.4).

3.6 Counting

Each turret of the sample changer is loaded with 10 trays (8 samples, 1 standard and 1 background tray) and the counter is set to operate. The counts from the sample, standard and background are automatically printed out, together with the length of count in each case (this is generally 200 minutes or the time taken to reach 10^4 counts).

The activity in the dry sample is calculated from the following equation:

$$^{40}\text{K } \beta \text{ eq. pCi/g dry} = \frac{S/T_s - B/T_b}{K/T_k - B/T_b} \times 3.9 \times 10^2$$

where S = counts obtained from the sample source over T_s minutes
B = counts obtained from the background over T_b minutes
K = counts obtained from the standard over T_k minutes;

it is then converted to wet weight if required.

3.7 Energy response

The response of the counting assembly to several radionuclides encountered in the aquatic environment is shown in Table 3.

Table 3 The response of the thin-source counting equipment to various isotopes

Isotope	β_{max} MeV	Efficiency		
		cpm/pCi	cpm/pCi per g dry	cpm/pCi, $^{40}K = 1$
$^{106}Ru/Rh$	2.0-3.6	1.39	0.277	1.04
$^{144}Ce/Pr$	2.98 & 0.32	1.78	0.356	1.34
^{40}K	1.32	1.33	0.266	1.00
^{137}Cs	0.51	1.05	0.209	0.79
^{60}Co	0.31	0.59	0.117	0.44
^{95}Nb	0.15	0.21	0.042	0.15
^{54}Mn	-	0.02	0.003	0.01
Background	5-6 cpm			

4 Thin-source beta counting with absorber

The determination of ruthenium-106 in laverbread.

4.1 Summary

The dried sample is assayed by measuring the count rate of a 200 mg source spread over 100 cm^2, using an automatic 5 inch proportional gas-flow counter with a 52.8 mg/cm^2 copper absorber between the source and detector end-window. After subtracting the contribution from the natural potassium in the sample, the ruthenium-106 content is calculated by comparing the corrected count rate with that of a ruthenium-106 standard identically mounted and counted. The activity of the sample is expressed as "pCi ruthenium-106/g wet weight".

4.2 Operating conditions

The operating modes for the counter are the same as in paragraph 3.3, except that the analyser mode switch is set to "β with absorber" (or "β then β with absorber" if both a gross beta and a ruthenium-106 beta count are required). A 52.8 mg/cm^2 absorber is placed on the counting slide.

4.6 Energy response

The energy response is shown in Table 4.

Table 4 The response of the thin-source counting equipment (with absorber) to various isotopes

Isotope	β_{max} MeV	Efficiency		
		cpm/pCi	cpm/pCi per g dry	cpm/pCi, $^{40}K = 1$
$^{106}Ru/Rh$	2.0-3.6	1.00	0.201	1.67
$^{144}Ce/Pr$	2.98 & 0.32	0.86	0.172	1.43
^{40}K	1.32	0.60	0.120	1.00
^{137}Cs	0.51	0.13	0.027	0.22
^{60}Co	0.31	0.03	0.005	0.04
^{95}Nb	0.15	0.01	0.002	0.02
^{54}Mn	-	0.01	0.002	0.02
Background	5 cpm			

REFERENCE

1. U.K.A.E.A. PG Report 403W (1962). Analytical method for the determination of gross beta activity in vegetation, seaweed, fish flesh, shore sand, silt and seabed mud by the infinite depth technique.

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