V. GENERAL ASPECTS OF ^{14}C TECHNIQUE

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INTERNATIONAL COMPARISON OF PROPORTIONAL GAS COUNTERS FOR ^{14}C ACTIVITY MEASUREMENTS*

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In early 1982 we attempted to compile information on ^{14}C counting equipment from all laboratories listed in RADIOCARBON. A gratifying number of laboratories cooperated in communicating their data: more than 65% of those in which proportional counters are in operation. Our compilation covers more than 80% of all gas counters used for ^{14}C dating. Unfortunately, the number of parameters determining counter quality as well as the spread in these factors is so large that their separate influence cannot be established beyond statistical doubt. Nevertheless qualitative and semi-quantitative trends are clearly observed. Laboratories are not identified in the data but the calculations and standardization procedures allow the laboratories to identify their own counter data in the graphs. Each plot does not contain a complete compilation of all counters. This is either because of lack of or internally inconsistent information.

COUNTER TYPES

Forty-nine laboratories reported use of 174 gas counters, and 32 laboratories employed liquid scintillation spectrometry. We have identified different types of counters according to construction material and type of filling gas. A survey is presented in figure 1. Most of the ^{14}C laboratories appear to apply CO_2 counting in proportional counters made of copper.

The largest single counter is a Cu/CO_2 counter with a volume of 7.5L. The smallest are Qu/CO_2 microcounters of 5mL. There are 15 Oeschger-type counters. The counter set-ups obtaining the high precision of ca 1 ‰ (2 x 22 hours counting period) consist of sets of counters connected in parallel.

EFFECTIVE VOLUME AND NORMALIZED COUNTING RATE

In order to compare counter performances, certain characteristics such as the recent counting rate, background and meson counting rate, are normalized to a standard volume of 1L. In so doing, the dead volume (= volume of the counter

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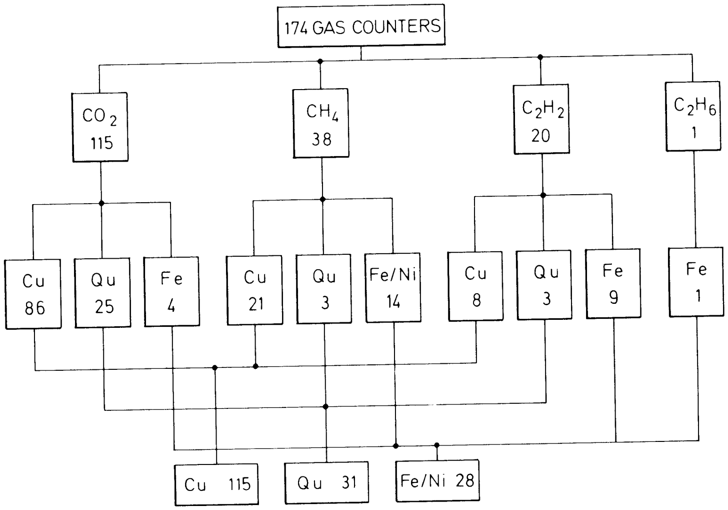

Fig 1. Survey of construction materials and counting gases used for ^{14}C proportional counters. This compilation represents > 80% of the ^{14}C counters in operation. Cu = copper, Qu = quartz, Fe = iron, Ni = nickel.

end pieces + tubing of the inlet system) has to be subtracted from the total volume. The resulting effective volume for each counter is more easily obtained by dividing the standard counting rate A_{ox} by 0.95 x the specific activity of NBS oxalic acid (= standard activity) and by the operating pressure of the counting gas, p. Further, we must consider the number of carbon atoms per molecule of counting gas, N_c. The normalized standard counting rate then is:

$$A_N = \frac{A_{ox}}{pN_c} \qquad (1)$$

and the effective volume:

$$V_e = \frac{A_N}{13.51} \times 2 \text{ (liters)} \qquad (2)$$

where 13.51 dpm is the standardized specific activity of 1g of recent carbon in 1981.

Comparing the effective volume with the counter volume as calculated from the dimensions reveals that the ratios observed for the large as well as the minicounters range between 0.65 and 0.95, with the majority between 0.85 and 0.95. This means that the wire connections in the counter can be made so that only minimal end effects occur.

MESON COUNTING RATE

To make an easy comparison between counters at ground surface and underground, the meson counting rate is normalized to the counting rate per cm^2 of effective horizontal counter cross-section, S_e. To obtain the latter, the effective length L_e, is multiplied by the inner diameter, D:

$$S_e = L_e . D = \frac{V_e}{\pi (D/2)^2} . D \tag{3}$$

Figure 2 shows the thus obtained standardized meson counting rates per cm^2 as a function of the effective counter size (equivalent to the normalized standard counting rate). For ground-level counters, the spread is in the range of 0.8 to $1 cpm/cm^2$, but within this range, it seems to hardly depend on the counter size.

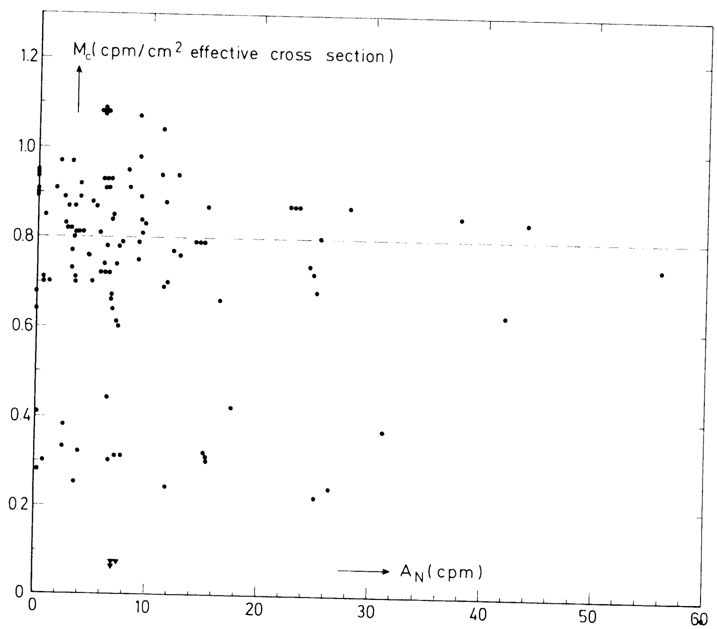

Fig 2. Meson counting rates of ^{14}C proportional counters per cm^2 of effective counter cross section (Eq 3) as a function of the effective counter size (normalized standard counting rate (Eq 1)). The triangular points are maximum counting rates.

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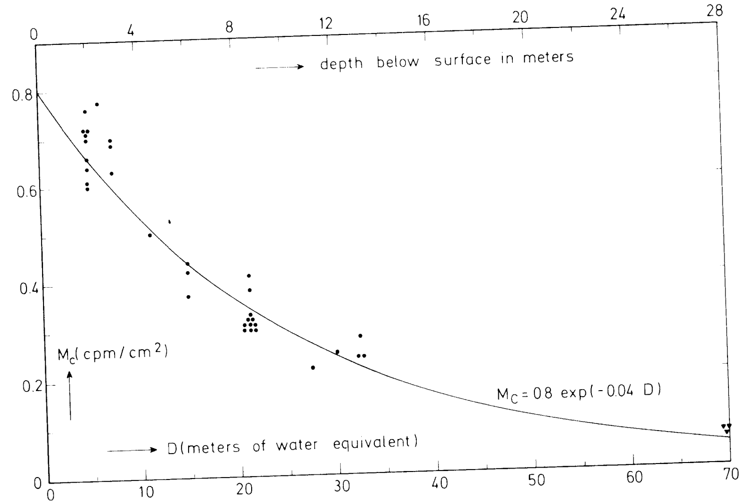

Fig 3. Meson counting rates of a number of ^{14}C proportional counters per cm^2 of effective counter cross-section as a function of the shielding. The shielding capacity of sand and concrete was transferred to meters of water equivalent by using an average specific gravity of 2.5g/cm^3. The shielding effect observed is 4%/m of water or ca 10%/m of depth (Eq 4).

In figure 3, the meson counting rate per cm^2 of effective counter cross-section is shown as a function of the amount of shielding. For a better comparison, the latter is given in meters of water equivalent, obtained by multiplying thickness by specific gravity. For earth and concrete we have used an average value of 2.5g/cm^3. This comparison only can be made on a limited number of counters because most are operated in basements with an unspecified shielding effect of the buildings above. Nevertheless, the results point to an absorption coefficient for water of 4%/m equivalent to ca 10%/m of sand or concrete:

$$M_C \simeq 0.8 \exp(-0.04\ D)\ (\text{cpm/cm}^2) \qquad (4)$$

where D is the depth in meters of water equivalent. The meson counting rate at the earth surface again is ca 0.8 cpm/cm^2.

COUNTER BACKGROUND

The background of the proportional ^{14}C counters originates from mesons and neutrons in cosmic radiation, from the radioactivity of the environment (building walls, paint, etc), and from the radioactivity incorporated in the materials of the counter itself and its surroundings (GM and anti-coincidence counters, shielding, etc). Shielding against the

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second component is simply obtained by some 15 to 30cm of lead or iron. The last component is minimized by selecting low-activity material as copper or quartz, and by including an inner shield of old lead or mercury. The meson/neutron component is dealt with by installing the counters as deep underground as possible, further shielding with heavy metals and a material of high hydrogen content such as paraffin or polyethylene for reducing neutron energies and boron (boric acid) for absorbing thermal neutrons. It is essential that an anti-coincidence shield of GM-counters or plastic scintillator surrounds the ^{14}C counter. Such shielding is also obtained by the concentric internal arrangement of anti-coincidence wires in Oeschger-type counters.

In order to study the shielding capacity of various counter set-ups, we will compare the background counting rate with the measured meson counting rate (fig 4). Distinction

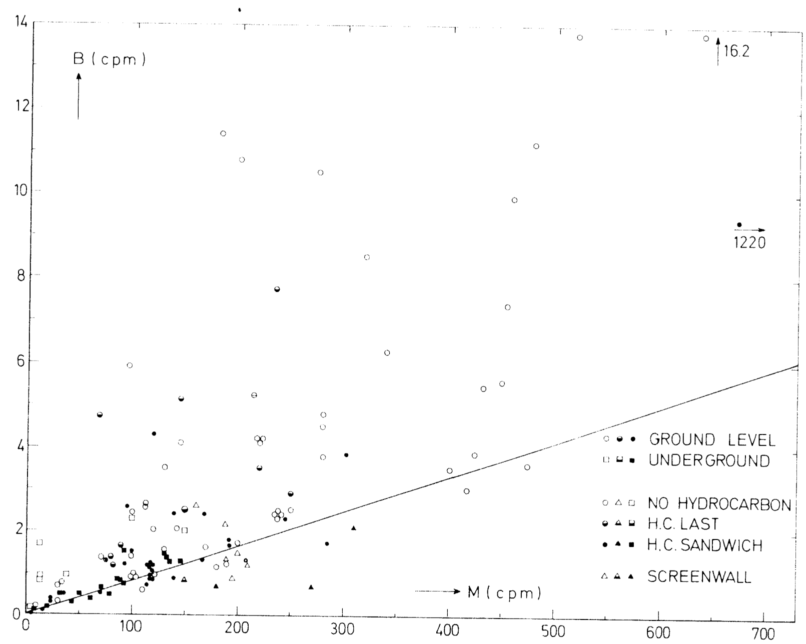

Fig 4. Background counting rate (B) as a function of meson counting rate (M). The underground laboratories (below 7m) are specifically indicated. Further identification was made for Oeschger-type counters and for the counter shielding: no neutron shielding by a layer of a hydrocarbon compound (HC), merely an inner layer of HC and a layer of HC between two layers of high specific density material (HC sandwich). The solid line indicates a favourable trend of B/M of 0.83% (2.5cpm/300cpm).

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has been made between surface or basement level laboratories and those deeper underground. We also indicate the type of shielding applied with a neutron shield consisting of any form of hydrocarbon. High backgrounds seem to coincide with the absence of this HC shield. Further, it is our impression that shielding with HC is further improved by applying a heavy material inside the HC shield for the absorption of gamma rays or protons, originating in the neutron shield ("HC sandwich").

It appears that a background/meson ratio of:

$$B/M \simeq 0.8cpm/100cpm \quad (5)$$

is a favourable trend, although a theoretical justification for this number cannot be given. This trend also applies to the underground laboratories. It does not imply that the background is only determined by cosmic radiation. The meson counting rate is also proportional to the counter size and thus, more or less, to the amount of material used in the counter construction.

Although not indicated in figure 4, no significant difference is found among different counter materials. In practice, copper and quartz appear to be equally suitable, as well as steel which is generally used in Oeschger-type counters.

QUALITY OF COUNTERS

We must emphasize that good quality ^{14}C counters are not only characterized by high precision, which is, in turn, determined by the choice of counter size, depending on the amount of sample generally available.

Again, for counter comparison we have to normalize for filling pressure, using the normalized standard counting rate (A_N) defined by Eq (2). The counter background is normalized for a standard filling pressure of the counting gas of 1 atm. From the data reported by a few laboratories, we obtained a pressure dependence of the background of ca 0.2‰/Torr (= (dB/dp)/B). The normalized background then is:

$$B_N = B\left[1 - (p - 760)\,\frac{dB/dp}{B}\right] \quad (6)$$

(cf Grootes, 1977)

where the filling pressure, p, is in Torr.

The normalized values of the standard counting rate and the background are compiled in figure 5. Identifications have been made of the position of the counter with respect to ground level and the type of shielding (cf fig 4). Again, the HC sandwich shielding appears to be the most favourable. We present the same set of data in figure 6 according to counter

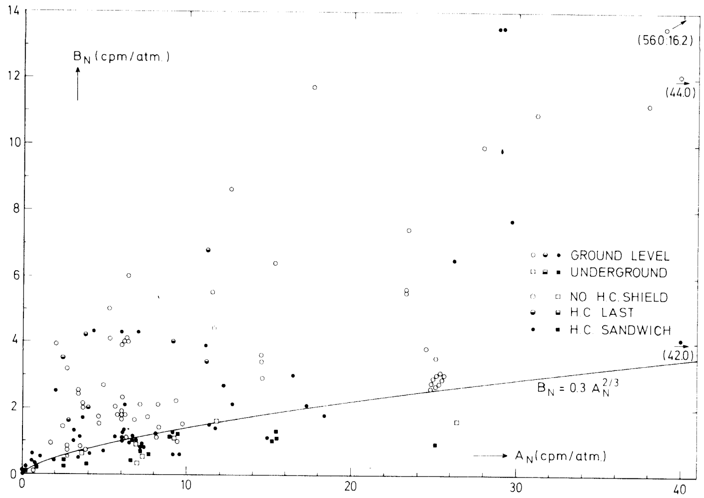

Fig 5. Relation between ^{14}C counter background, B_N, normalized to 1 atm (Eq 6) and the effective counter size, represented by the normalized standard activity, A_N (Eq 1). Identifications are made of the position of the counter with respect to ground level (underground = > 7m below ground level) and type of shielding.

material and filling gas. The plot does not show a generally lower background for one type of counter material. Comparing the two graphs of figures 5 and 6, however, reveals that a high background for a certain counter size is hardly observed for quartz counters that have been properly shielded by an HC sandwich.

A further striking point is that underground counters generally but not necessarily have a relatively low background. Also a proper neutron shield seems to reduce the background, which is confirmed by Stuiver et al (1979).

Assuming that the background is proportional to the counter surface, while the standard counting rate is proportional to the volume, a tentative favourable trend has been drawn in both figures 5 and 6 given by:

$$B_N = 0.3A_N^{2/3} \tag{7}$$

This is approximately equivalent to a ratio of

$$B_N/A_N \simeq 0.1 \text{cpm/cpm} \tag{8}$$

Finally, figure 7 represents a compilation of all data on factors determining the quality of a counter. The figure of merit represents the dating limit of a counter. If this is

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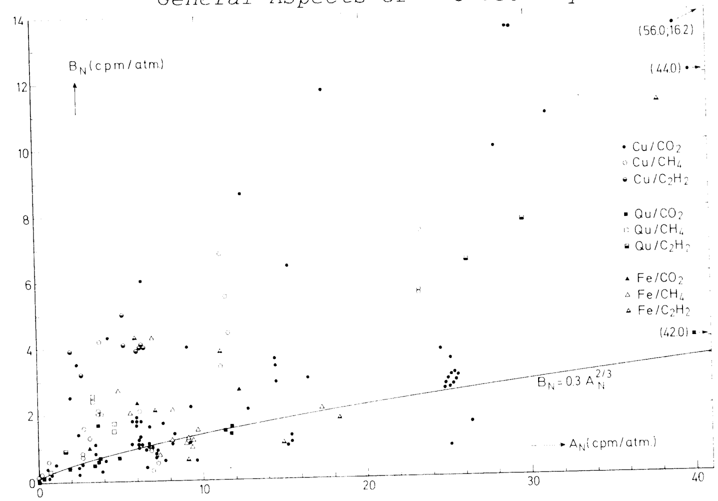

Fig 6. Relation between ^{14}C counter background and effective counter size, both normalized to 1 atm of filling pressure (cf fig 5). Identifications are made of the counter construction material and the type of filling gas. Fe refers to iron, nickel or steel. The same set of data is in figure 5.

conventionally defined as the measured activity, A_{min}, equal to twice the standard deviation in the background, σ_B, the age limit is:

$$T_{max} = -8033 \ln \frac{A_{min}}{A_{ox}} \tag{9}$$

where

$$A_{min} = 2\sigma_B = 2\sqrt{B/t_c} \tag{10}$$

and t_c is the counting time (taken as 2 x 22 hours = 2640min), resulting in:

$$T_{max} = 8033 \ln \left[\tfrac{1}{2}\sqrt{t_c} \cdot \frac{A_{ox}}{\sqrt{B}} \right] \tag{11}$$

The factor $A_{ox}/\sqrt{B}$ is the figure of merit.

The relative precision of the standard counting rate is:

$$\sigma_{A_{ox}}/A_{ox} = \sqrt{(A_{ox} + B)/t_c}/A_{ox} \tag{12}$$

To better understand this plot, a separate, similar graph is inserted showing the true standard counting rate, A_{ox}, and the measured background, B, deduced from chosen realistic values of the figure of merit and the standard precision.

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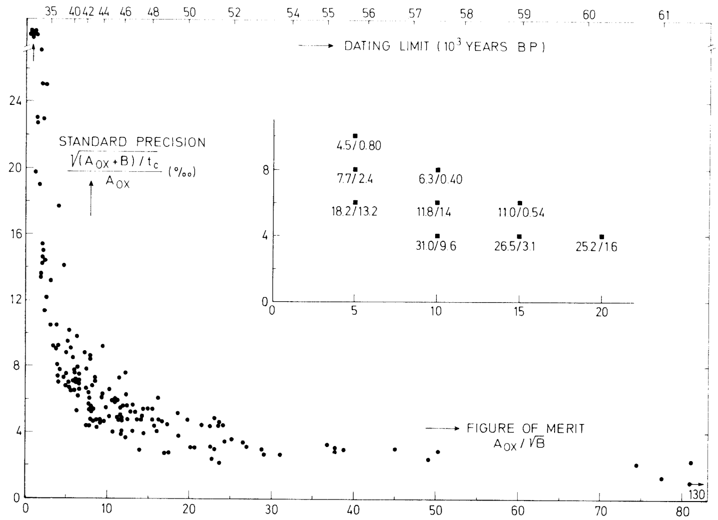

Fig 7. Compilation of the figure of merit and the precision of a two-day (2640 min) measurement of the standard activity in ^{14}C proportional gas counters. This plot merely presents a general view of the detection capacities of ^{14}C laboratories with regard to obtainable precision and dating limit (upper scale; Eq 11). The upper left hand corner shows that data on some mini- and microcounters with volumes below 100mL are beyond the scale. The similar, inserted graph shows A_{OX} and B values (given as A_{OX}/B) deduced from certain realistic values of the figure of merit and the standard precision.

These values are given as A_{OX}/B. The inner curvature of the assembly of data points consists of the relatively small counters with low background, the outer curvature of the large counters with relatively high background.

SUMMARY AND CONCLUSIONS

From the compilation of data on the performance of 174 proportional gas counters for ^{14}C detection the following conclusions can be drawn:

1) the large number of, and variability in parameters allow only qualitative or semi-quantitative conclusions.
2) from 111 ^{14}C laboratories recently listed in RADIOCARBON, 32 use liquid scintillation counting; 49 use 174 proportional gas counters.
3) ca 50% of these counters are made of copper and are used with CO_2 as a counting gas (fig 1).
4) the volumes of the counters range from 7.5L to 5mL.
5) the effective volume of the counters (= counter volume - dead volume) ranges from 65 to 95% and generally is 90 ± 5%.

6) the meson counting rate per cm^2 of effective cross section ranges from 0.1 to 1cpm, whereas the ground-level laboratories generally detect 0.8 to 0.9cpm/cm^2. The trend with increasing depth below ground level is a reduction of ca 4%/m of water, equivalent to roughly 10%/m of soil (figs 2 and 3).
7) in general, the lowest background is obtained if the shielding consists of a neutron shield containing a material with a high hydrogen density (paraffin + boric acid) between two layers of heavy metal. This also seems to apply to underground laboratories (figs 4 and 5).
8) generally, counters in underground laboratories (below 8m) have a relatively low background.
9) a favourable trend in the ratio between the background and the meson counting rate is ca 0.8% (= 0.8cpm/100cpm) (fig 4).
10) generally, copper, quartz, steel and nickel seem equally suitable for obtaining a low background although exceptions exist for copper and steel (fig 6). A favourable trend in the ratio between background, B_N, and standard activity, A_N, both normalized to 1 atm of filling pressure is observed of $B_N = 0.3\ A_N^{2/3}$ (figs 5 and 6).

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