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SOME PROBLEMS IN RADIOCARBON DATING

BY

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Introduction.

Since the discovery of natural radiocarbon[1), 2)] and its application to dating problems[3)] by LIBBY and coworkers, a number of papers have appeared describing the technique[4), 5), 6)] and the results obtained at Chicago[7), 8)] and elsewhere[9)]. In addition, the concordance of the results with geological and archaeological expectation has been discussed[10), 11), 12), 13)]. However, no extensive treatment of the limitations and errors of the method has been given although several isolated examples have been discussed[11), 14), 15), 16)]. It is the purpose of the present paper to present some general conclusions regarding the range of the method, its accuracy, possible sources of error other than that of provenience, and the rationale of the instrumentation. The problems are considered from the theoretical viewpoint and no new experimental data are given. Since the fundamentals of the method are well summarized in the literature and in LIBBY's book[17)], it is assumed that the reader is familiar with them.

Instrumentation.

Description of Screen Wall Counter.

For the measurement of natural radiocarbon, the screen wall counter[4)] was selected. The design of this instrument is based on two considerations, namely, the desire to obtain the maximum net count from a sample in a detector of a given size, and the necessity of establishing accurately a net count which may be small compared with the background of the counter. The carbon powder to be measured is mixed with water to form a thick paste and is spread evenly over the inside surface of a cylinder which is placed concentrically to the axis of the counter. In this way, all absorption losses other than self-absorption are eliminated, a geometry of essentially 50 per cent is obtained, and the sample fills the maximum solid angle around the counter. The cathode of the counter consists of an open grid or screen of wires. The screen does not define the radial extent of the sensitive volume of the counter which, in fact, extends to the surface of the sample. The purpose of the screen is simply to improve the electrical characteristics of the system. Through the use of a double sample cylinder, one half holding the sample, the other a blank, frequent alternation between the two is possible without affecting the counter filling. Thus, the effects of temporal changes in background are minimized.

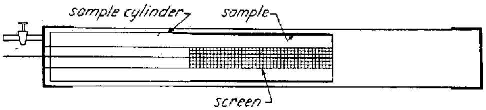

Fig. 1. Screen wall counter.

Comparison with Gas Sample Counter.

The fundamental problem in the selection of the proper detector unit is to obtain the maximum counting rate from a detector of minimum physical size. The physical size is an important parameter, since the background rate of the instrument is proportional to the size, and the effect to be determined is of the same order as or smaller than the background even under the most favourable circumstances.

For a gas sample counter, practically all the disintegrations occurring in the sample will be detected and, therefore, the efficiency defined as the fraction of the disintegrations detected is essentially unity. For the screen wall counter, using a thick sample of elemental carbon, the efficiency is only 5.46 % 4), 5)*.

* When this value for the efficiency is used in equation 1, the range ϱ must be taken as 20 mg/cm^2. If the more recent value for the range of 28 mg/cm^2 is used, the efficiency E_1 is 3.8 %. The quantity which has been determined experimentally is the product $E \cdot \varrho = 1.10$ mg/cm^2. Physically, this is the thickness of the layer which—in the absence of self-absorption—would give the same counting rate as the thick sample.

However, a screen wall counter of conventional size will contain some 20 times the amount of carbon that can be used in a gas sample counter having the same background. The two methods thus happen by chance to be similar with respect to this parameter and a more detailed comparison is in order.

This can be made as follows. The counting rate S_1 to be expected from a screen wall counter is

$$S_1 = E_1 \varrho A S, \tag{1}$$

where E_1 is the efficiency as defined above, ϱ is the range in mg/cm^2 of the C-14 β-particle, A is the sample area which is equal to $\pi d_1 l_1$, d_1 being the inside diameter and l_1 the length of the sample cylinder, S is the specific activity of the sample in disintegrations per minute and milligram.

For a spherical gas counter, assuming an efficiency of unity, we have

$$S_2 = M \frac{\pi}{6} d_2^3 S, \tag{2}$$

where M = mg of carbon per cm^3. If the filling gas contains one atom of carbon per molecule and has a pressure of one atmosphere, $M = 0.54$ mg/cm^3.

In order to consider counters having the same background we will assume the background to be proportional to the horizontal cross-sectional area, which is for a cylindrical screen wall $l_1 d_1$ and for a spherical gas counter $\frac{\pi}{4} d_2^2$. Therefore,

$$l_1 d_1 = \frac{\pi}{4} d_2^2 \tag{3}$$

is the condition for identical background rates. Since the sample area of a screen wall A is π times its background area, the screen wall sample area can also be expressed as π times the background area of the corresponding spherical gas counter, viz. $\frac{\pi^2}{4} d_2^2$. Combining equations (1) and (2), using the numerical values $E_1 \cdot \varrho = 1.10$ and $M = 0.54$, and setting $A_1 = \frac{\pi^2}{4} d_2^2$, we find

$$\frac{S_1}{S_2} = \frac{1.10 \frac{\pi^2}{4} d_2^2}{0.54 \frac{\pi}{6} d_2^3} = \frac{9.60}{d_2}. \tag{4}$$

This equation is plotted in Fig. 2, using the volume of the spherical counter as the independent variable.

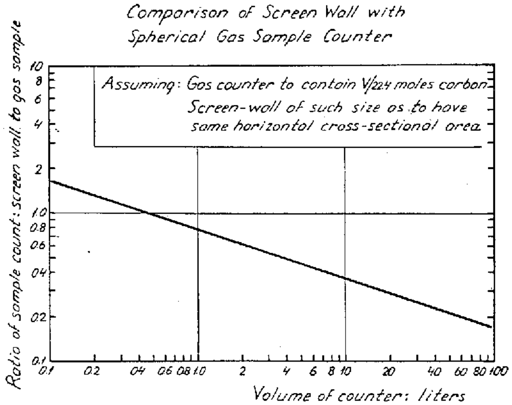

Fig. 2.

This figure offers a comparison of the sample count ratio of a screen wall counter to that of a gas counter as a function of the sample size.

It is clear that the screen wall counter is superior to the gas counter in the case of small detectors, while the gas counter is to be preferred when the detectors are large. Although it is always desirable for reasons of counting statistics to use as large a sample as possible, other considerations (such as difficulty of operation of large counters and the mass of the shield required) usually limit one to a volume of a few liters. It is seen from Fig. 2 that, in this region, the ratio of counting rates is not much different from unity and is slightly in favour of a gas sample counter. Because of the geometrical requirements of the anti-coincidence shielding method, however, the sphere is not the optimum shape. That is to say, a bundle of anti-coincidence counters of conventional design makes a cylindrical cavity for the sample counter. Therefore, from the viewpoint of the mass of shielding required as well as the electrical and the construction characteristics, the comparison should be made between a screen wall counter and a *cylindrical* gas sample counter of the same dimensions.

Such a comparison is given in Fig. 3, where the additional parameter a of the ratio of counter length to diameter has been introduced. The principles of good counter design will in general dictate that a be greater than 1. Although this shifts the curves in favour of the screen wall counter, it is apparent that the count ratio is still so close to unity that the choice will probably be made on the basis of considerations other than sample: background ratio. One such consideration is the amount of sample available. From this point of view the gas counter is to be preferred. But this advantage will in general be of considerably less importance than the ease of operation (stability, reliability). The gas counter assumed in the previous analysis requires operation in the proportional region with a filling pressure of 1 atm. This necessitates an operating voltage of the order of 5 kV and the detection of pulses of the order of millivolts. Such a system is inherently more difficult to operate than a Geiger counter at 1 kV, giving pulses of several volts.

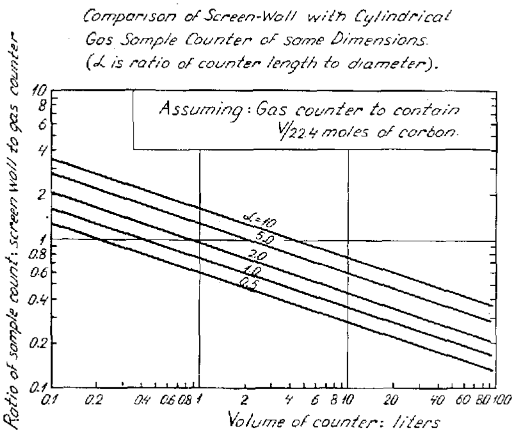

Fig. 3.

Statistical Considerations.

Effect of Background Rate.

To maximize the statistical precision obtainable in a given counting time it is desirable to maximize the ratio S^2/B, where S is the net sample count and B the background counting rate[18].

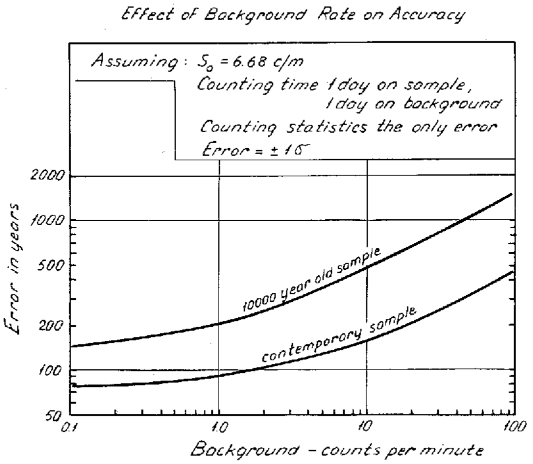

Fig. 4.

For a screen wall counter, where both the background and the sample counting rate are linear functions of counter size, the larger of two counters is always better from this point of view. As pointed out in the preceding section, the maximum possible size of the counter is determined by practical considerations and therefore the only possibility to improve the accuracy obtainable in a given counting-time is to reduce the background count. The extent to which this is necessary and profitable is indicated in Fig. 4, in which the error in years calculated merely on the basis of statistical considerations is plotted as a function of the background rate for a contemporary sample and for a 10,000 year old sample. The counting rate from a contemporary sample is assumed to be 6.68 counts/min.[4] and the counting time 24 hours on sample and on background. In practice, the background of the counter shielded with lead is found to be about 100 counts/min., and when shielded with anti-coincidence it becomes 4 to 6 counts a minute. It is clear from the curves that, while it is essential to make use of the gain from the anti-coincidence shielding, further reduction of the background gives no spectacular gain in accuracy except for very old samples. The assumption of equal times spent on sample and background is not optimum when the background rate becomes small compared to sample rate[18]. This effect will be noticeable in Fig. 4 only for fairly recent samples. It will make the curve somewhat steeper over the middle cycle of the abscissae and will make the asymptote a factor of $\sqrt{2}$ lower.

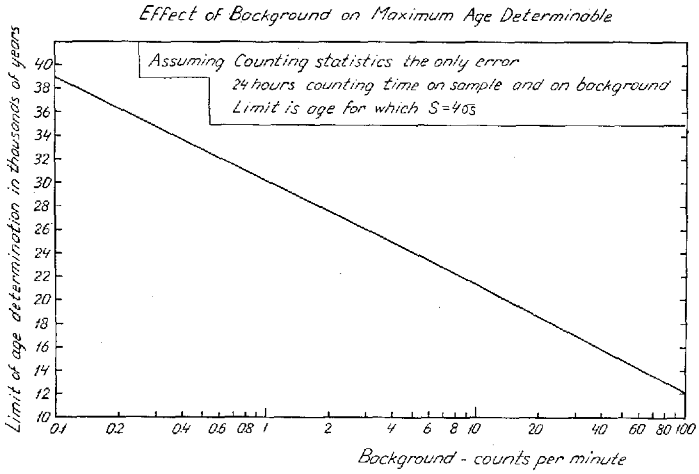

Fig. 5.

The principal gain from background reduction is the extension of the method to older samples. This effect is shown in Fig. 5 which represents the maximum age determinable (with 2 days' counting) as a function of background rate.

The limiting age is here arbitrarily defined as the age for which the net sample count is equal to four times its statistical standard deviation. It is necessary to be rather conservative in the choice of this limit since, as the difference between background and sample becomes very small, one is less certain that the statistical error is the limiting one. It is apparent from the curve that every time the background is reduced by a factor of two, 2500 years are added to the range of the method.

The extension of the method to older samples through the reduction of background is clearly a difficult process because of the logarithmic relationship between these quantities. Moreover, other sources of error which are not considered in this calculation, such as low level chance contamination, may become the limiting factors preventing the attainment of the limit given by the curve.

In the measurement of the contemporary assay and of the samples of known age, the limiting factor in the accuracy appears to be counting statistics.

Effect of Counting Time.

In Figs. 4 and 5 the counting time has been assumed to be 48 hours. This time has proved in practice to be a good compromise between the gain in accuracy with prolongation of counting times and the desire to measure more samples. The two days counting time is a convenient one in that this period fits in well with the time required to prepare a sample. The effect on the

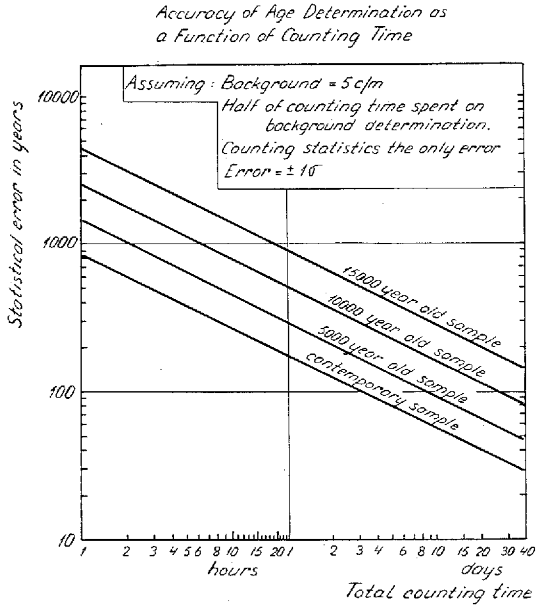

Fig. 6.

accuracy of changing the total counting time is presented in Fig. 6, where the statistical error is plotted against total counting time on a double logarithmic scale. It is obvious from this figure that the accuracy increases rapidly during the first day of counting and much more slowly thereafter. For example, after two hours of counting on a 5000 year old sample, the age is determined to ± 1000 years. After one day of counting the accuracy is ± 300 years, and after two days of counting it is ± 200 years. To reduce the error to 100 years would require counting for eight days.

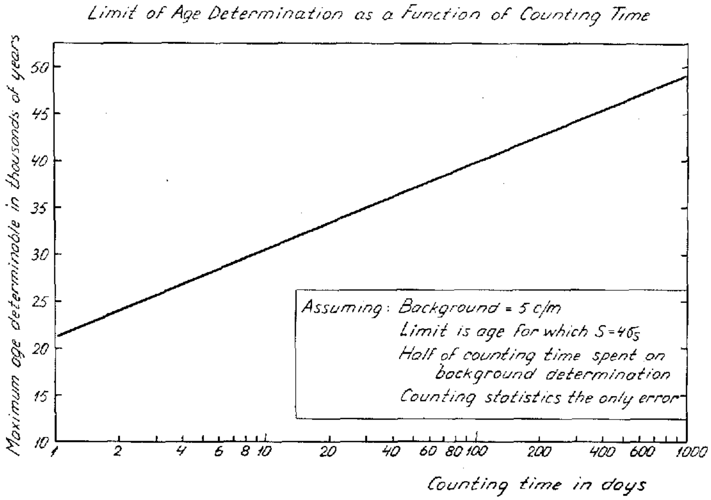

Fig. 7.

Because of the possibility of other sources of error, such as contamination of the sample during processing, it is to be preferred that greater statistical accuracy be obtained through the measurement of independently prepared samples rather than through the extended counting of a single sample.

The extreme difficulty encountered in an attempt to extend the method to very old samples by the extension of the counting time is illustrated by Fig. 7. Even in the *reductio ad absurdum* of counting 100 days with a background of 5 counts per minute, the limit of the method is seen to be 40.000 years. The maximum age so far reported by radiocarbon dating is 28.000 years[19].

Effect of Isotopic Enrichment.

The discovery of natural radiocarbon was made on samples which were isotopically enriched by methane thermal diffusion columns[1], [2], and a similar apparatus was constructed at the University of Chicago for possible use with very old samples. There are several practical difficulties associated with the use of this technique, such as the large amounts of sample necessary, the length of time required, the cost of the process, and the difficulty in establishing the exact value of the enrichment factor. This will of necessity limit the application of isotopic enrichment to a few samples of extreme importance. The results which can be accomplished by this technique are indicated in Fig. 8, in which the maximum age as previously defined is plotted against the isotopic enrichment factor for three different counting times. It seems unlikely that enrichment factors more than a few hundred can be obtained in practice, and it must be remembered that the sample size requirement will increase by a factor at least as large as the enrichment factor. Again, the gain in maximum age determinable is a function of the logarithm of the independent variable so that appreciable extension of the method is accomplished only with great difficulty.

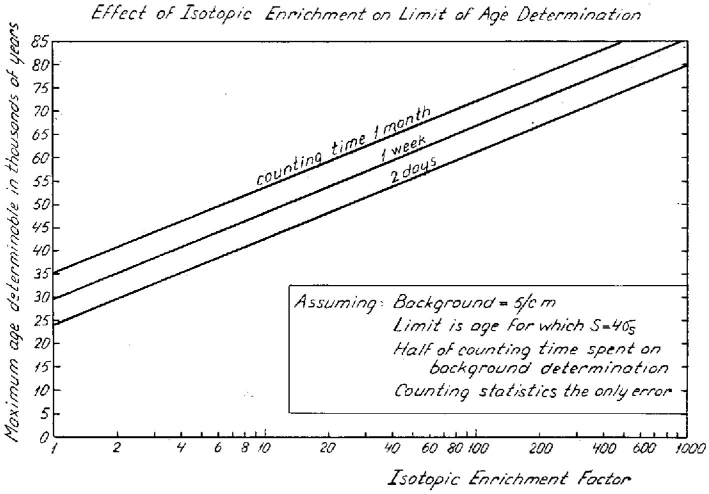

Fig. 8.

It is clear from a consideration of all the factors as represented on Figs. 5, 7, and 8 that, while the method can easily reach back to periods of a few tens of thousands of years, further extension can result only from a very considerable expenditure of effort.

This fundamental limitation of the method derives directly from the exponential decay law, and it can be said in general that any dating method based on radioactive decay will have a range of applicability of approximately $k \cdot t_{1/2}$, where k is of the order of 10.

Sources of Error.

Contamination by other Radionuclides.

Contamination errors can result from the intrusion into the sample of carbon of a different specific activity or of other radioactive species. One of the main purposes of the chemical treatment of the sample before counting is to separate radiocarbon from all chemically different activities. The widespread occurrence of radium and its decay products as well as the high specific activity of these nuclides make them the principal source of such contamination. Most rocks contain of the order of 10^{-12} parts of radium, and soil contains 10^{-13} parts of radium[20)]. Therefore, samples consisting of many small pieces of material of large surface area can carry along with them comparatively large quantities of such activities. Fortunately, chemical separation is particularly easy. When the sample is burnt to CO_2, radium and its solid decay products are left behind. However, the gaseous member of the decay chain (radon) is collected with the CO_2 and, if not eliminated, will give rise to active deposit on the carbon sample. If the CO_2 is precipitated as $CaCO_3$, and the CO_2 regenerated by acid treatment, contamination from this source can be eliminated.

Obviously, it is of prime importance that the reagents used during the preparation of elemental carbon from the sample be of extreme purity with respect to other radio-elements. The only criterion of adequate purity of all reagents used prior to mounting the sample is the attainment of a truly zero count from a dead sample such as coal or petroleum.

Because of the extremely high adsorptive properties of the elemental carbon samples prepared by magnesium reduction (surface areas of the order of 200 m^2/g) great care must be exercised to prevent recontamination. The principal danger is again radon, this time the radon which is present in the atmosphere to the extent of 10^{-16} curies/cc[20]. Exposure of the sample to the air, especially when the sample is dried, should be kept to an absolute minimum. It has been observed, for example, that a sample which had been carefully mounted and had given a count equal to background, may show an activity above background if simply removed from the sample cylinder in the dry state and remounted.

It seems possible that contamination from aerial radon together with low level contamination of reagents may be the limiting factors preventing the extension of the method to the ultimate ages which seem possible from a consideration of errors due to counting statistics, only. As the activity of the sample decreases, it will probably be found that the errors indicated by analyses of duplicate samples prove to exceed the errors calculated on the basis of counting statistics.

For this reason, one should be cautious in accepting as practicable the limits of age which have been calculated as being accessible to the method on a statistical basis.

Contamination by Carbon of Different Specific Activity.

Contamination by carbon of different specific activity (i. e. different age) is a more difficult problem, since chemical methods may be of little or no use in rectifying the situation. Processes of contamination may be separated in two groups, viz. mechanical inclusion and exchange or chemical reactions. In all cases, the contamination may be by carbon older or younger than the sample.

Under mechanical inclusion may be grouped such events as penetration of a sample by rootlets of plants, crystallization of carbonates or deposition of organic compounds (e. g. humic acids) from solution onto or within a porous sample, and stirring and mixing of strata of different ages by the action of natural forces or by human or animal activity.

Direct exchange of carbon atoms between chemical species, without a net chemical reaction, occurs, for example, in the following equilibrium

$$CO_3^{=(aq)} + C^*O_2^{(g)} \rightleftarrows C^*O_3^{=(aq)} + CO_2^{(g)}.$$

This type of reaction is improbable with organic compounds. The ease of exchange of $BaC^{14}O_3$ with atmospheric CO_2 is well known[22], but, on the other hand, Urey[23] found shell carbonate capable of resisting oxygen exchange with dissolved carbonate over geologic periods. The results of Kulp[9] indicate that finely divided carbonate in ocean sediments can maintain itself at a specific activity different from that of its environment for a period of at least 14,000 years (sample no. 107 B). Therefore, the situation looks encouraging for the use of shell carbonate as a dating material; however, the possibility of exchange under certain conditions remains, and research is planned to investigate this point by comparison of the specific activity of shell and wood from the same provenience and under different conditions of

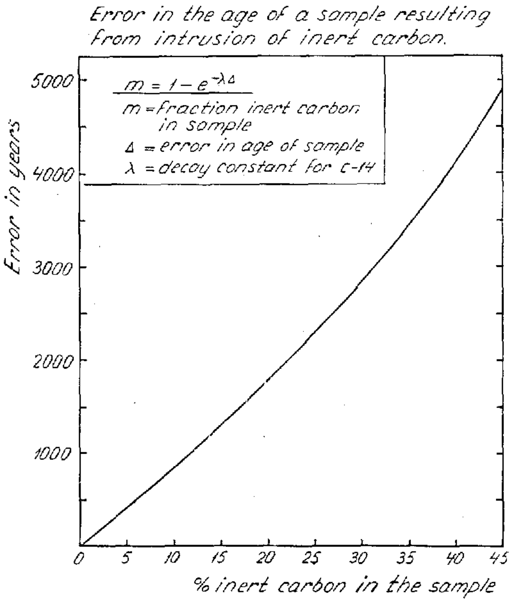

Fig. 9.

exposure. (The problem of the initially different specific activities of shell due to isotopic fractionation is discussed below.)

While the strongly bound carbon in organic molecules is not subject to direct exchange, such molecules can serve as a substrate for micro-organisms. For example, micro-organisms can break down cellulose and resynthesize other compounds. During this process, carbon of different specific activity, if present in the surrounding medium, can be incorporated into the new compounds and invalidate measurements made indiscriminately on the whole mass. For this reason, it may be necessary in some cases to isolate the unaffected cellulose or lignin for dating purposes.

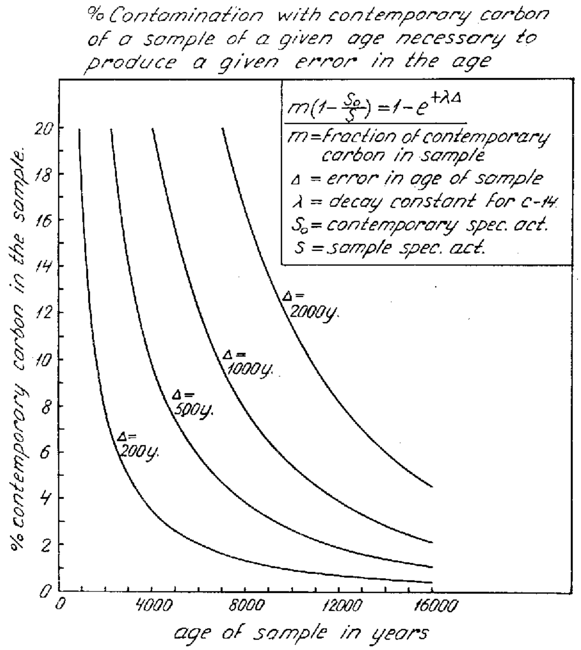

Fig. 10.

Another example of indirect exchange must be envisaged when aquatic plants grow in a hard water lake and have their specific activity displaced since the carbon they photosynthesize is derived partly from redissolved limestone[15].

The effect on the measured age caused by intrusion of inert carbon into a sample is shown in Fig. 9. It is clear that an error from this source of more than 1000 years can arise only if gross replacement (intrusion) of the carbon, i. e. amounting to more than 10 per cent, has occurred.

Fig. 10 shows the effect of contamination by contemporary carbon. In this case, the error is a function of the age of the sample. Each of the family of curves gives the degree of contamination by contemporary carbon necessary to produce a given error as a function of the age of the sample. Errors of 200, 500, 1000, and 2000 years are given. For example, to produce an error of 1000 years in the age of a 6000 year old sample, a contamination by 12 per cent with contemporary carbon is required. These curves may permit a decision as to the probability of a suspected error being due to this type of contamination.

Other Sources of Error.

Since the age of a sample is determined by the ratio of its activity to the activity of contemporary samples of the same material, it is unnecessary to make absolute specific activity determinations for age measurements. All that is required is the measurement of the counting rates of the unknown and of the reference sample under identical conditions. This is clear from the exponential decay law $S = S_0 e^{-\lambda T}$ which, when solved explicitely for sample age T, gives

$$T = 18{,}500 \log S_0/S,$$

S_0 and S being the activities of materials of age zero and T, respectively.

Obviously, the efficiency factor for the counter, which would appear in this equation if conversion were made to absolute disintegration rates, would appear in both numerator and denominator and, hence, cancel out. However, great care must be taken in the choice of the proper value of the contemporary activity, S_0.

Extensive measurements on contemporary samples[4), 9)] have shown, for example, that shell carbonate gives a counting rate 11 per cent higher than wood, so that S_0 for carbonate samples is 1.11 times S_0 for wood samples. A neglect of this difference would result in a dating error of some 900 years.

The analogous fractionation of carbon 13 has been known for some time[24)]. The ratio C-12/C-13 is 91.8 for wood and 89.2 for limestone, corresponding to a fractionation factor of 1.03 for C-13. While no further C-14 data are available, the C-13 measurements indicate the possible existence of a group of material with still a different isotopic composition. This group is rather poorly characterized due to the few data, (single measurements on each of seven samples) but seems to consist of such diverse materials as weeds, algae, spores, peat, linseed oil, chinawood oil, and rubber, which show a C-12/C-13 ratio of about 92.8.

C-14 measurements on contemporary samples from similar sources are highly desirable from the point of view of our understanding of the detailed chemistry of the exchange reservoir, and they are equally essential for the dating of such materials.

Any error in the half life of radio-carbon will appear as the corresponding percentage error in the measured age of the sample. Since the estimated uncertainty of the best value for the half life of C-14 is less than one per cent[4)], this error is small compared with other uncertainties of the method and need be considered only in those cases where the sample is measured to an unusually high accuracy.

Improvements in Instrumentation.

Double Screen Wall Counter.

Since the long counting time required for obtaining sufficient precision is at present a limiting factor, it is desirable to introduce improvements which would reduce the required counting time. This has been accomplished in Copenhagen through the use of the double screen wall counter[*]. In this instrument (Fig. 11), two independent detector units in a common envelope are used together with a triple sample cylinder. The sample is mounted

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[*] The assistance of R. L. SCHUCH of the Los Alamos Scientific Laboratory in the design and construction of the double screen wall counter is gratefully acknowledged.

on the middle cylinder, while the two end cylinders provide backgrounds. As in the conventional screen wall, the cylinder unit can be alternated between two positions. In one position, the sample is exposed to detector A (cf. Fig. 11) and a background to detector B, in the other position, the sample is exposed to B and a background to A. Two advantages are obtained by the use of this system. First, the counting time necessary to obtain a given statistical accuracy is reduced by $\frac{1}{2}$, and second, simultaneous background and sample counts are obtained, whereby the effect of any temporal changes in the background is eliminated. Considerably less than twice the conventional equipment is required for the double screen wall system. Duplication of the mixing and the recording stages of the anti-coincidence circuit is necessary, but only one set of anti-coincidence counters is needed. The shield dimensions need be increased only slightly, since the counter length increases by only $\frac{1}{3}$. Moreover, the necessity for frequent alternation between the two positions is considerably reduced in the case of the double screen wall counter so that the complexity of an automatic sample changer is avoided.

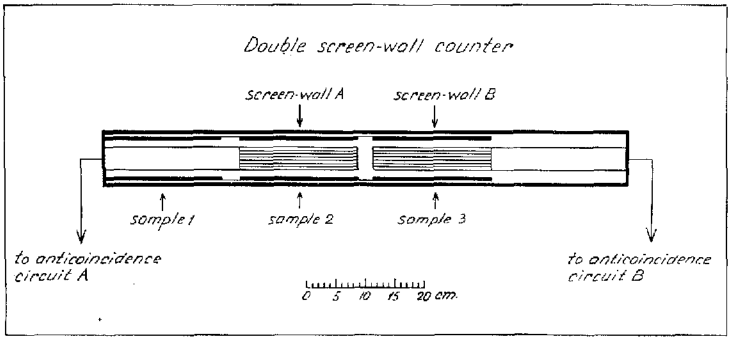

Fig. 11.

Solution Scintillation Counter.

An alternative system of measurement suggested by J. Arnold in 1948 is based on the scintillation counter method. It is proposed to convert the carbon of the sample into the chemical form of a suitable solvent in which some material can be dissolved to form a system with high efficiency for the detection of the C-14 β-particles. Some work has been done in this direction, but many technical difficulties must be solved before the method is proved to be practical. It is, however, worth considering the possibilities of this method in some detail, since it is difficult to imagine a system capable of giving a higher sample count combined with as low a background. The peculiar advantage of this system, which makes it so outstanding, is that it is able—theoretically at least—to use a sample in a condensed phase as a detector of nearly 100 per cent efficiency. Suppose, for example, that the carbon from the sample to be measured be converted to carbon disulfide or a similar organic liquid of a density about unity and containing of the order of 10 $^0/_0$ carbon. A small amount of a scintillator, e. g. terphenyl, is dissolved in this liquid sample and coincidence counts recorded from a pair of photomultiplier tubes triggered by scintillations from this system. On the assumption of 100 per cent counting efficiency, 80 cc of carbon disulfide would give a counting rate of 120 counts/min. In the form of a cube, this volume will have a cross-sectional area of about 18 cm^2 and, therefore, might be expected to have a background of 0.03 counts/min. if background could be reduced as efficiently as in the case of the screen wall counter.

With such an idealized instrument the maximum age determinable as defined above becomes 58,000 years.

The principal advantages of the system are its extreme compactness, thus eliminating the need for a massive shield, and the great economy of counting time obtained for samples of reasonable age. On the other hand, the gain in the maximum age determinable while significant is not spectacular.

The authors wish to express their gratitude to Professor P. Brandt Rehberg, head of the Zoophysiological Laboratory, where the dating apparatus is installed, for his hospitality and kind interest in the work.

The Copenhagen "dating project" was initiated jointly by members of the Danish National Museum, the Danish Geological Survey (D.G.U.), and the Zoophysiological Laboratory. It is supported financially by the Carlsberg Foundation whose sponsorship is gratefully acknowledged.

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Summary.

A detailed comparison of the screen wall counter with a gas sample counter with respect to the problems of radio-carbon dating is given. Curves are presented showing the accuracy and range of the method as functions of the background rate and the counting time. The errors due to intrusion of extraneous carbon are presented and discussed and certain improvements in the method are described.

Zoophysiological Laboratory, University of Copenhagen.

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