

MEASUREMENTS OBTAINED WITH THE 2π LARGE VOLUME COUNTER
(GENCO) AND THEIR COMPARISON WITH THE RESULTS OBTAINED
FROM OTHER PHYSICAL OR CHEMICAL METHODS

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External counting with large volume detectors of the liquid scintillation type offers mainly two advantages over similar devices using other detection techniques:

1. Less geometrical error introduced by the shape and the size of the object being counted, and
2. Relatively short counting times (100 sec.).

These factors are appreciated in routine as well as in diagnostic application of large volume counters.

In contrary to a so-called 4π counter now in use for several years in the U.S.A., we have a 2π counter available for our experiments. The outstanding feature of this type is its low price as compared to a 4π detector. But there is a disadvantage due to its incomplete geometry. Therefore the first topic of this report deals with geometrical problems in a 2π detector.

The second topic to be discussed is inherent in all γ ray spectroscopy. If the γ ray energies of an unknown

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mixture of radionuclides are the same or close together there is in most cases no other way for identification but conventional radiochemical procedures. By chance only a few fission products are absorbed to a broad extent by biological systems. Therefore large volume detectors of the liquid scintillation type can be used to determine the Cs-137 and K-40 content in human beings or in milk. But the Cs-137 contamination is not restricted to these objects alone. We thought it would be worthwhile to see how far a large volume liquid scintillation detector could be used for Cs-137 measurements in animal foodstuffs in which other fission products could interfere in Cs-137 and K-40 determinations.

Large volume counters were originally designed for low level counting. Therefore it is no wonder that strange effects can be observed if people contaminated with activities about $1 \mu\text{c}$ shall be counted. From this reason the last topic of this report deals with phenomena observed if activities higher than 10^{-7} c Cs-137 were introduced in the large volume detector.

GEOMETRICAL PROBLEMS INVOLVED IN A 2π LARGE VOLUME LIQUID SCINTILLATION DETECTOR

A suggestion was made by Onstead (previous article) to introduce a calibration system for the 2π counter based on weight and height of the object being counted. The calibration curves obtained by such an ideal model of natural objects (for example human beings) should be checked for its reliability with a natural object which could be analyzed easily by other methods.

We decided to use hams for this purpose. The reason was threefold:

1. Hams have irregular external shape and enough lean meat to get sufficient counts due their K-40 content.

2. Size and weight of hams are not too great. They are easy to handle, complete ashing is no problem and the ash can be estimated conveniently for its K-content by a flame photometer.

3. If our model proved to be reliable we could compare our results with those reported by Kulwich, Feinestein and Anderson (1) who found a correlation between the fat-free lean of the ham and its K-40 content. Furthermore we could learn how far a simple calibration system would work if a 2π detector for K-40 studies on living animals should be designed.

Ten hams, five fat and five lean at random were selected. Table 1 shows the different fat hams as F-1 to F-5 and the lean ones as M-1 to M-5 and gives the approximate dimensions, the weight and the percentage of the different parts of the ham related to the total weight of each ham. The hams were counted in the middle position of the detector, then cut into 33 pieces (skin, bones, external fat and lean) and each part was counted again, and weights of these parts were determined. The internal fat of the lean meat was estimated and from this the fat-free lean was calculated. All parts were ashed at 400°C. The K content of ash of the different parts as well as the total ham ash was estimated by flame photometry. The error of the counting and the flame photometric procedures could be estimated to be not more than 5% in all measurements.

Table 2 shows the total K content and the percentage of K in different parts of the various hams obtained by flame photometric measurements. The values for the external fat seem to be relatively high. But this might be due to the fact that it is very difficult to remove all the meat from the fat.

In Table 3 the results of K obtained by external counting, using the calibration diagram (Fig. 1) for the calculations of the K content, are compared with the results obtained by flame photometry.

The values of the total K obtained by analytical method fairly agree with the values obtained by radioactivity measurements. Therefore the calibration system, by taking weight and height into consideration, is proved to be reliable. If the calibration had to be done only on the weight basis, there would be clear discrepancy between the counted and the analyzed values.

TABLE 1. DIMENSIONS, WEIGHT AND PARTS OF HAM IN PERCENT OF TOTAL WEIGHT.

| Ham No. | Dimensions cm | Weight kg | Parts of Ham in % of Total Weight | | | | | Fat ext. % | total % | Lean Fat-Free % |
|---------|------------------|--------------|-----------------------------------|-----------|-----------|------------------|------------|------------------|------------|-----------------------|
| | | | Bones % | Skin % | int. % | Fat ext. % | total % | | | |
| F-1 | 46x29x14 | 9.67 | 10.25 | 5.89 | 5.48 | 31.71 | 37.19 | 46.67 | | |
| F-2 | 40x34x15 | 11.64 | 8.97 | 6.62 | 4.28 | 32.75 | 37.03 | 47.37 | | |
| F-3 | 45x27x15 | 9.74 | 9.11 | 4.94 | 3.67 | 39.33 | 43.00 | 42.95 | | |
| F-4 | 51x30x18 | 10.18 | 8.84 | 4.36 | 3.97 | 37.51 | 41.48 | 45.33 | | |
| F-5 | 51x32x17 | 10.56 | 9.69 | 6.15 | 2.27 | 37.38 | 39.65 | 44.51 | | |
| M-1 | 45x30x16 | 12.11 | 8.09 | 4.95 | 4.85 | 29.95 | 34.80 | 52.18 | | |
| M-2 | 44x27x17 | 11.84 | 8.31 | 4.51 | 5.07 | 35.46 | 40.53 | 46.65 | | |
| M-3 | 49x29x18 | 12.94 | 8.66 | 5.12 | 3.95 | 32.92 | 36.87 | 49.74 | | |
| M-4 | 43x26x17 | 10.90 | 6.32 | 4.27 | 2.52 | 35.21 | 37.73 | 51.68 | | |
| M-5 | 45x30x18 | 12.47 | 6.39 | 4.57 | 3.43 | 39.19 | 42.62 | 46.42 | | |

TABLE 2. TOTAL K IN HAM AND PERCENTAGE OF K IN VARIOUS PARTS OF THE HAM
OBTAINED BY FLAME PHOTOMETRIC MEASUREMENTS.

| Ham No. | Total K in Ham (analyzed) g | % of K in Various Parts of the Ham | | | | | Fat ext. % | total % | Fat-Free-Lean % |
|-------------|-----------------------------------|------------------------------------|-----------|-----------|------------------|--------------------|------------------|------------|--------------------|
| | | Bones % | Skin % | int. % | Fat ext. % | Fat-Free-Lean % | | | |
| F-1 | 17.78 | 3.45 | 1.95 | 2.31 | 13.39 | 15.70 | 78.90 | | |
| F-2 | 20.56 | 3.93 | 2.92 | 1.42 | 15.29 | 16.71 | 76.68 | | |
| F-3 | 18.12 | 3.16 | 1.61 | 1.60 | 16.94 | 18.54 | 76.70 | | |
| F-4 | 18.49 | 2.89 | 1.22 | 1.42 | 13.20 | 14.62 | 81.27 | | |
| F-5 | 17.41 | 3.76 | 2.95 | 1.00 | 16.48 | 17.48 | 75.82 | | |
| M-1 | 22.96 | 2.88 | 1.87 | 1.39 | 8.59 | 9.98 | 85.26 | | |
| M-2 | 20.24 | 3.00 | 1.72 | 1.45 | 10.12 | 11.57 | 83.71 | | |
| M-3 | 24.20 | 2.89 | 1.90 | 1.67 | 13.93 | 15.60 | 79.50 | | |
| M-4 | 22.02 | 2.04 | 1.29 | 0.84 | 11.74 | 12.59 | 84.08 | | |
| M-5 | 25.33 | 2.19 | 1.51 | 1.52 | 15.05 | 16.37 | 80.00 | | |
| Mean values | | 3.03 | 1.91 | (1.44) | (13.47) | 14.92 | 80.19 | | |

TABLE 3. TOTAL K IN HAM AND IN LEAN PORTION OF THE HAM OBTAINED BY EXTERNAL COUNTING AND BY FLAME PHOTOMETRIC MEASUREMENTS.

| Ham No. | Total K in Ham | | K in Lean Portion of Ham | | | |
|---------|----------------|-----------|--------------------------|--------------|---------------------|-----------------------|
| | analyzed g | counted g | analyzed g/kg | counted g/kg | counted single g/kg | counted together g/kg |
| F-1 | 17.78 | 17.49 | 2.86 | 2.91 | 3.05 | 3.27 |
| F-2 | 20.56 | 21.48 | 2.88 | 2.87 | | |
| F-3 | 18.12 | 16.04 | 3.12 | 3.10 | | |
| F-4 | 18.49 | 17.62 | 3.05 | 3.13 | | |
| F-5 | 17.41 | 18.27 | 2.88 | 3.19 | | |
| M-1 | 22.96 | 24.06 | 2.88 | 3.29 | 3.14 | 3.29 |
| M-2 | 20.24 | 22.88 | 2.82 | 3.29 | | |
| M-3 | 24.20 | 24.96 | 2.83 | 3.22 | | |
| M-4 | 22.02 | 22.11 | 3.16 | 3.27 | | |
| M-5 | 23.33 | 22.67 | 3.04 | 2.93 | | |

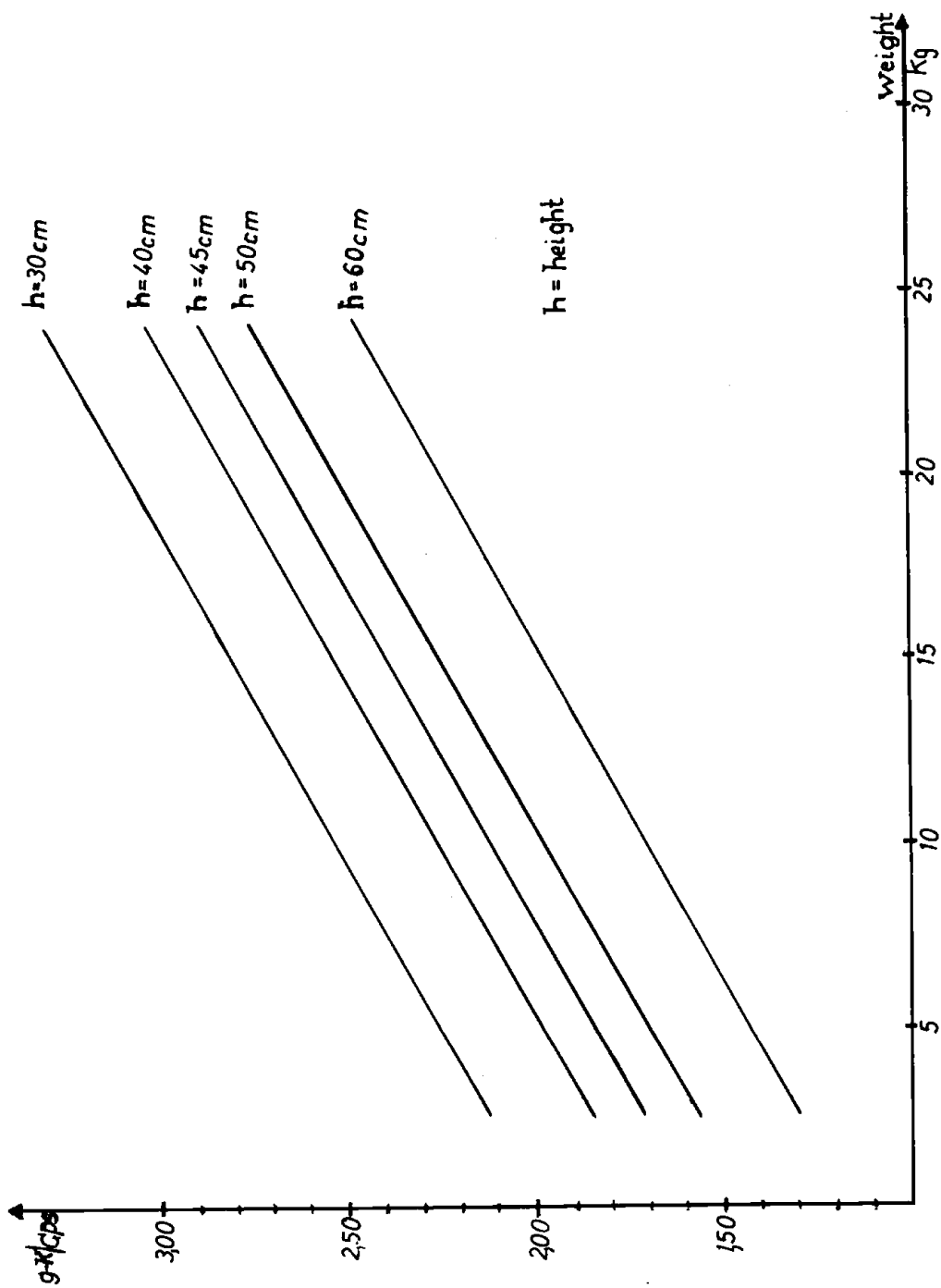


Figure 1. Calibration diagram for K in k-40 channel. Grams of K per net counting rate against weight (density $\sim 1 \text{ g/cm}^3$) with the height of the object as parameter.

Table 4 shows a comparison between the gravimetric determinations of the fat-free lean of the hams in percent

TABLE 4. FAT-FREE LEAN OF THE HAM IN % OF THE TOTAL WEIGHT AND GRAMS OF K RELATED TO THE TOTAL WEIGHT OF THE HAM OBTAINED BY GRAVIMETRIC AND K-40 MEASUREMENTS.

| Ham No. | Fat-Free Lean of Ham in % of Total Weight % | Grams of K Related to Total Weight of Ham (counted) g/kg |
|---------|---|--|
| F-1 | 46.67 | 1.81 |
| F-2 | 47.37 | 1.85 |
| F-3 | 42.95 | 1.65 |
| F-4 | 45.33 | 1.73 |
| F-5 | 44.51 | 1.73 |
| M-1 | 52.18 | 1.97 |
| M-2 | 46.65 | 1.93 |
| M-3 | 49.74 | 1.93 |
| M-4 | 51.68 | 2.03 |
| M-5 | 46.42 | 1.81 |

of the total weight and the content of K in grams as related to the total weight of the hams obtained by K-40 measurements. The values indicate a correlation for the fat-free lean and the K content related to the total weight of the hams. But the nondestructive method of the K-40 determination is too coarse to show slight variations in the fat-free lean of the hams. Even to get the radiometric results of Table 4 a counting time of 33 min. for each ham was necessary without taking into consideration that the same amount of time had to be spent on the background counts too.

Therefore in our opinion the nondestructive method of determining the lean portion of the ham by its K-40 content has no practical value because in this case the K-40 yield in ham is too small and for this reason the counting time

for significant results is too long. The situation is improved by using a whole animal instead of a single part.

In Table 5 the specific K content was calculated on the basis of our ham results for the different parts of a swine. In a rough estimation one should expect at least as many counts as necessary to operate a 2π counter by normal counting times (100 sec.) used for measuring human beings.

TABLE 5. EXPECTED TOTAL NET COUNTING RATE FOR A 110 kg. SWINE IN A 2π COUNTER.

| Parts of Swine | Weight (mean values of 305 animals 208 days old) kg | Mean Values of K g/kg | Expected Net-Counting Rate in 2π counter cps |
|----------------|---|-----------------------|--|
| Lean Port. | 36.54 | 3.01 | 37 |
| Fat | 32.90 | 0.695 | 8 |
| Bones | 11.29 | 0.648 | 2 |
| Skin | 5.57 | 0.658 | 1 |
| Other Parts | 23.71 | ? | ? |
| | 110.00 kg Total Weight | | 48 cps Total Net-Counting Rate |
| | | | (1 g \cong 0.3 cps) |

At the moment it is uncertain if this K-40 determination would be of any value to a slaughter house. We think more investigation on the biological side of the problem has to be done until this external K-40 determination as an indicator of internal lean percentage of an animal can be introduced in practice. Especially more experimental data of the K-content of the different parts of the body of the animal must be obtained and the K content as function of the age of the animal must be studied. But besides this,

the experiments done, showed the reliability of the calibration system based on weight and height of the object being counted in a 2π detector.

PROBLEMS INVOLVED IN Cs-137 MEASUREMENTS IN OTHER OBJECTS
THAN IN MAN AND MILK

The energy resolution (half-width at half-height) for the K-40 peak in a liquid scintillation detector was reported to be 30% for a 4π counter and 18% for a 2π counter (2). The equivalent value for a NaI crystal of maximum dimension is about 3-5%.

The γ ray energy discrimination for a NaI crystal still remains a problem. It becomes obviously more complicated if one uses a liquid scintillation detector. There are no difficulties if the radionuclides are known like in medical diagnosis with the human or animal bodies' natural discrimination against certain radionuclides. Therefore Cs-137 measurements in man and milk can be done without any interference of other radionuclides. But if one wants to use the same nondestructive method for other objects, for example animal foodstuff, to get experimental data for the chain of contamination--animal foodstuff--> domestic animals --> animal products --> man--one must be aware that other gamma emitting fission products are present which show higher Cs-137 or K-40 activities than is really present.

The bomb tests were stopped in autumn 1958, and in autumn 1959 we tried to measure the Cs-137 content in animal foodstuffs grown at the field station of Mariensee near Hannover, Germany, by external measurements in the 2π detector.

About three metric tons of foodstuff could easily be measured in two days. The results were reported as activities in the Cs-137 channel and as activities in the K-40 channel for the different foodstuffs (3).

The activities, especially for all products grown above the soil, turned out to be rather high in both the

Cs-137 channel and K-40 channel, and the potassium values were not in agreement with the values obtained by flame photometric measurements. Better agreement in the K values could be found for all products grown protected by outer shells like corn, rye, etc. All fruits grown underneath the soil like turnips or potatoes showed nearly the same values for both K measurements.

From this we concluded that differences between the K values from both methods could be used as an indicator for the presence of fission products other than Cs-137, but only in a very rough qualitative manner.

If one keeps in mind that the bomb tests were stopped in autumn 1958 and if it is so that radionuclides produced as induced activity during a nuclear explosion can be neglected because of abundance of total activity of other long living fission products which is 10 to 100 times more than induced activity, then there are only two fission products Zr-Nb-95 and Ru-Rh-106 which must be taken into consideration for their interference in Cs-137 and K-40 measurements with a 2π liquid scintillation counter.

That these radionuclides are really present can be seen from Figure 2 which gives a γ ray spectrum of three representative foods mentioned above. The gamma spectra were made with a NaI crystal and a 256 channel analyzer. Besides the expected energy peaks at 0.66 Mev and 1.46 Mev due to Cs-137 and K-40 there are peaks at 0.51 Mev and between 0.72 and 0.77 Mev for hay, a slight indication at 0.51 Mev for rye, but there is no additional peak for fodder beets. All three results are in agreement as predicted on the discrepancies of the K measurements.

Though an abnormally high K-yield as a result of the K-40 measurements may be used as a quantitative indicator for additional fission products contamination, from this there is only a rough estimation possible to get the true value of the Cs-137 contamination. If one looks at the decay scheme of Ru-Rh-106, there are energy peaks at 0.51 Mev with 11% gammas per disintegration, at 1.55 Mev with 3% gammas per disintegration and at 1.13 Mev with 12% gammas per disintegration, while Zr-Nb-95 has gamma energy lines at 0.75 Mev with 49% gammas per disintegration, at 0.72 Mev with 49% gammas per disintegration and at 0.77 Mev with 100% gammas per disintegration which could interfere in Cs-137 or K-40 measurements.

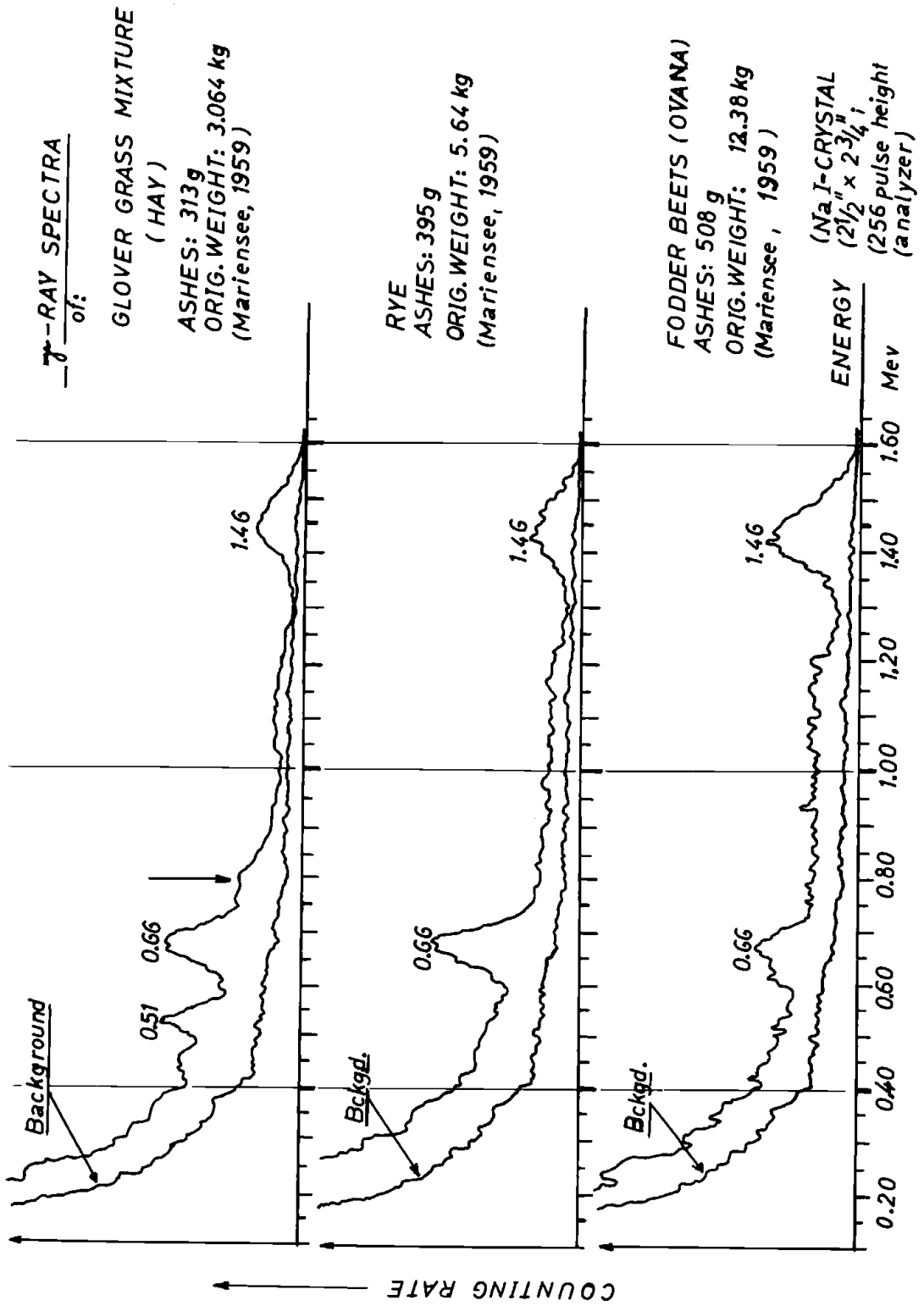


Figure 2. Gamma ray spectra of hay, rye and fodder beets grown 1959 at the fieldstation of Mariensee nearby Hannover, Germany

Gammas from Zr-Nb-95 do not appear in the K-40 channel of the 2π liquid scintillation counter. Therefore there is no way in estimating their contribution to the Cs-137 channel. Gammas from Ru-Rh-106 should appear in both the K-40 and the Cs-137 channel. The ratio of their appearance due to the percentage per one disintegration is expected to be about 1:4 for the ratio of K-40 to Cs-137 channel. Therefore if the ratio of the K radio-measurement and the K flame photometric determination is multiplied by 4 and then the activity of the foodstuff obtained in the Cs-137 channel is divided by this factor a better approximation for the Cs-137 activity should be obtained.

In Table 6 the Cs-137 activities in the Cs-137 channel, the K yield obtained in the K-40 channel, the Cs-137 activities found by chemical analysis and the flame photometric K estimations are listed. In the last column the estimated Cs-137 content calculated by the way described above is given.

It is true of course that in case of the foodstuffs the 2π liquid scintillation counter shows only true Cs-137 activity if by biological discrimination turnips and potatoes, in our case, have no measurable other fission products absorbed. But using a large volume counter for routine gamma ray activity determinations of foodstuffs it is worthwhile to know that the potassium value sometimes can be used, for products grown above the ground, as an indicator of additional contaminations. If by other means the presence of other long living gamma ray emitting fission products can be determined, which interfere both in Cs-137 and in K-40 channel, a rough calculation can be made by the K-ratio of the counted and analyzed values and the ratio of the contributions of the interfering radionuclides in both channels.

BEHAVIOR OF THE 2π LANDSTUHL COUNTER WITH ACTIVITIES HIGHER THAN 10^{-7} CURIES Cs-137

If activities of Cs-137 and K-40 in the Landstuhl 2π counter are not higher than 10^{-7} c the spectra of point and extended sources look like those shown in Fig. 3. It can

TABLE 6. COMPARISON OF THE Cs-137 AND K-40 RESULTS OBTAINED BY DIFFERENT METHODS.

| Foodstuff From Marien- see, 1959. | Activity in Cs-137 channel $10^{-10}c/kg$ | K measured in K-40 channel g/kg | Radio- chemical Cs-137 Ana- lysis $10^{-10}c/kg$ | Flame photometric K Analysis g/kg | Approximate Cs-137 yield (calculated) $10^{-10}c/kg$ |
|---|--|--|--|--|---|
| Clover grass mixture (Hay) | 72.7 | 34.6 | 3.45 | 14.8 | 7.2 |
| Rye | 3.94 | 4.08 | 1.23 | 4.24 | -- |
| Turnips (Ovana) | 0.16 | 3.85 | 0.22 | 3.75 | -- |

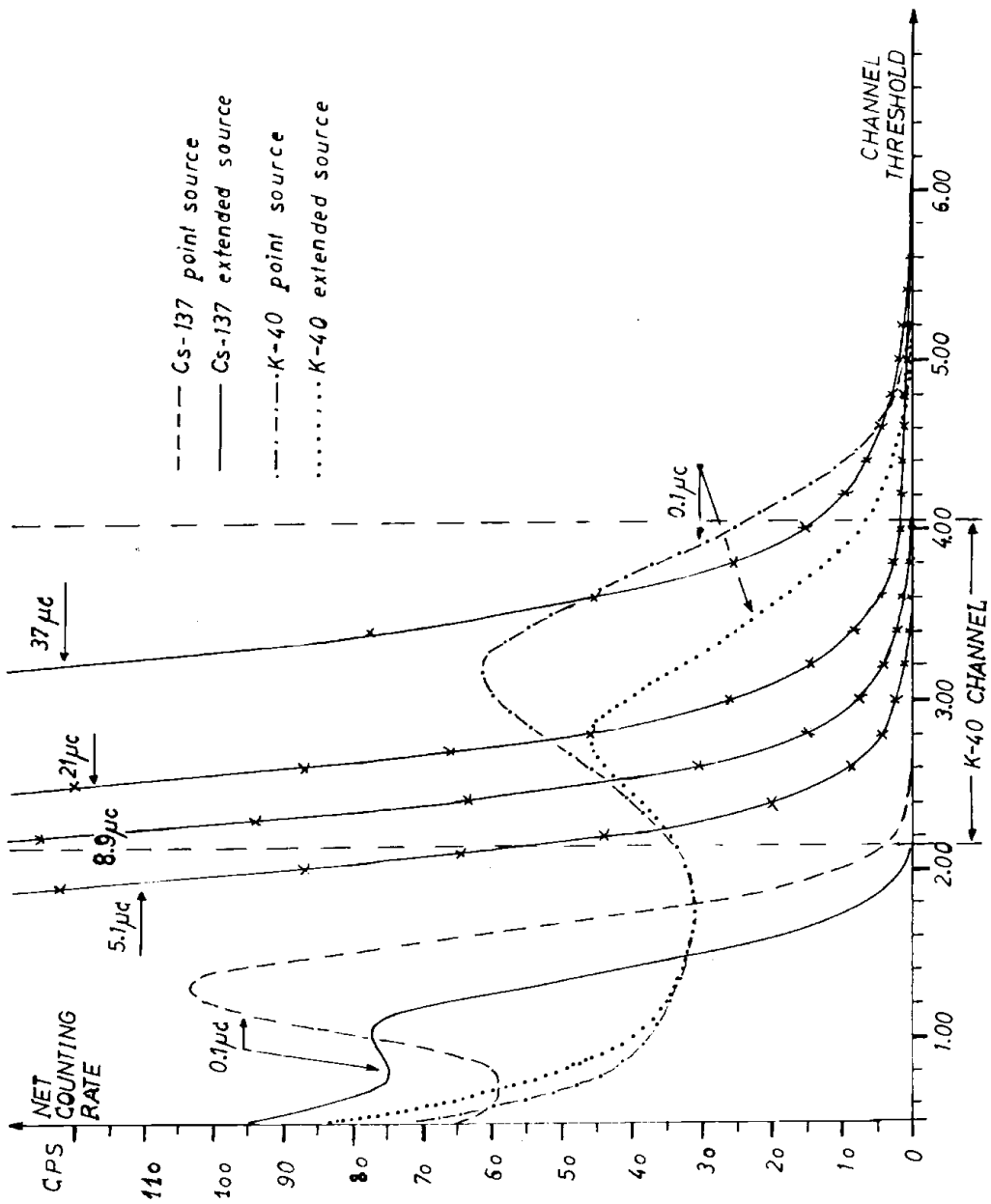


Figure 3. Gamma ray spectra of Cs-137 and K-40 from point and extended sources. For comparison purpose the normal spectra are related to the same activity. The channel width for the K-40 measurements is indicated.

clearly be seen that for some activities the maxima for point sources in comparison to extended sources are in both cases (Cs-137 and K-40) higher and shifted to higher energies. This effect must be kept in mind if a large volume counter has to be calibrated for the settings of the Cs-137 and the K-40 channel of the discriminator threshold and channel width. If the activity, for example in Fig. 3 for Cs-137, is increased using an extended source not only the maximum of the peak will be increased but also the bases of the curves. That means the spectrum is widened in such a way that it looks like a shift of the whole spectrum of Cs-137 to higher energies. The more activity is used the more counts can be observed in the K-40 channel until the complete activity due to K-40 is covered by the counts due to Cs-137 (Fig. 3).

The efficiency (cps/dps) for Cs-137 counts in the K-40 channel does not remain constant any longer. It is as one can see from Fig. 4, a function of the activity (net counting rate in Fig. 4) of the source used.

If this efficiency versus net counting rate curve, which seems to be a straight line in the range covered by Fig. 4, is watched for a few weeks it can be noticed that the calibration points fluctuate, but all points lie on straight lines (Fig. 4). Because counting rates of Cs-137 activities higher than 10^{-7} c will block the mechanical register of the scaler of the Cs-137 channel, the result of the Cs-137 measurements in the K-40 channel represented in Fig. 4 could be of help in estimating the body burden of a person who was contaminated by Cs-137 in the range of 10^{-6} c.

The person and the calibration sources were measured at the same intervals (Fig. 4). In the meantime urine and fecal excretion measurements of the same person were done. Due to the fluctuation of the calibration curve after 28 days when the second whole body counting was made the first measurement had to be corrected.

The results of all the measurements are shown on the right side of Fig. 4. From this table it can be seen that the agreement between the whole body counting and the excretion measurements is pretty good. The limiting factor using this method for higher activities is set to be the mechanical register in the scaler.

Though the effect of the shifting of the spectra of

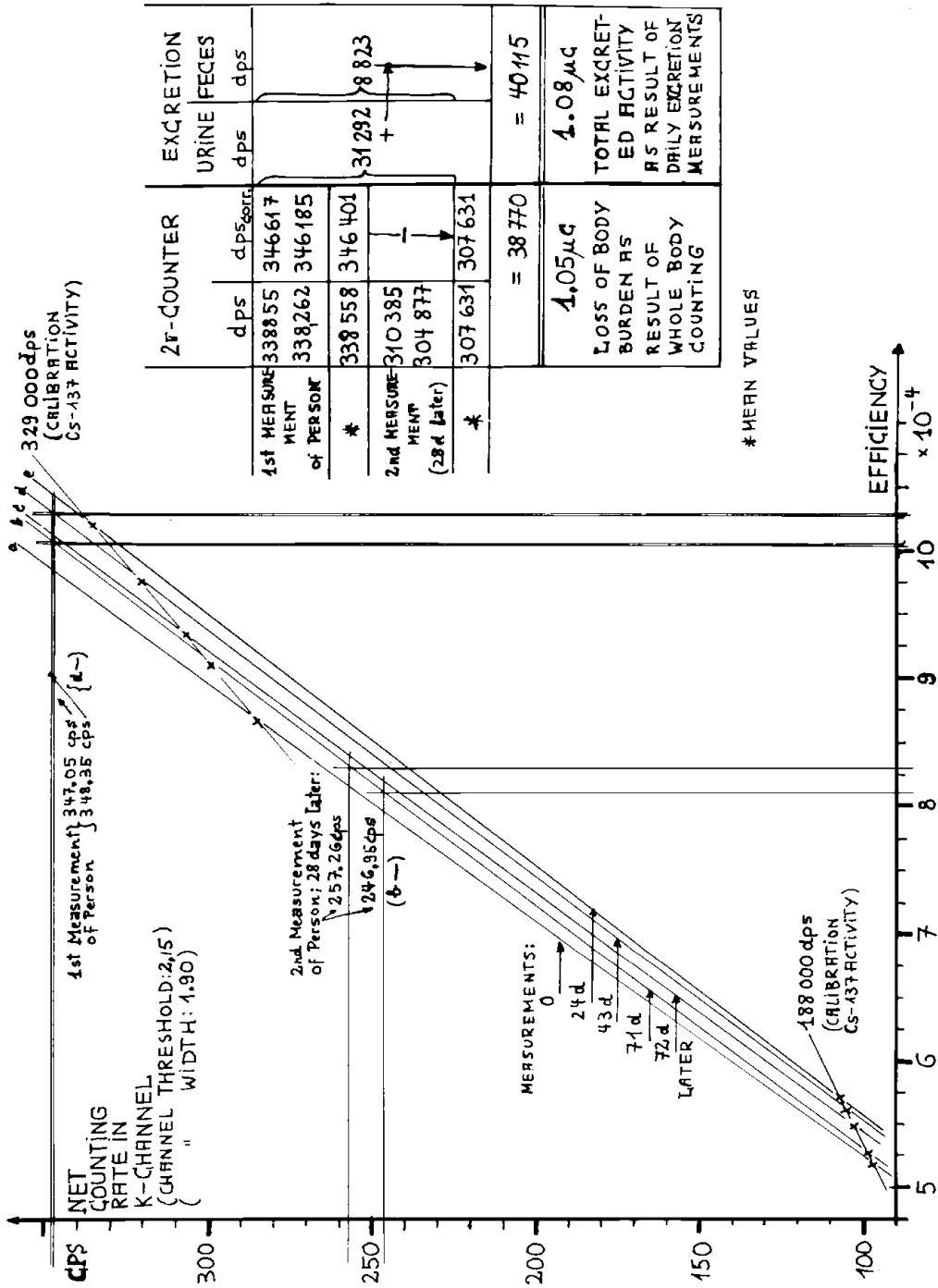


Figure 4. Net counting rate against efficiency for Cs-137 in the K-40 channel. On the right side of the diagram the values for the whole body and excretion measurement for Cs-137 are given.

Cs-137 for higher activities is not quite clearly understood at the moment, it must be stated for future designing of large volume counters of the liquid scintillation type for measuring human bodies, to improve the electronic part of the machine in such a manner that the activity range from 10^{-10} c to 10^{-5} c can easily be covered, avoiding such effects described above.

SUMMARY

To summarize our experiments and experience with the 2π Landstuhl large volume liquid scintillation counter (abbreviated GENCO) we would say the incomplete geometry is no handicap in getting significant results if a calibration system as described by Onstead is used. The reasonable price in comparison to a 4π counter recommends the counter for routine work itself. The measurements of different objects must be of course restricted to such items from which the kind of the radionuclide to be measured is known or can be learned by other methods. In this connection it would be good to combine the 2π counter with a NaI crystal device in one unit. In doing so the vertical arrangement used in Landstuhl must be changed back to the horizontal version like in use for 4π counters.

The electronic part of the unit should by future designers be improved in such a way that the upper limit should be at least 10^{-5} c to cover the important range of clinical application of a whole body counter and to give a chance to measure human body contaminated in the level around $1 \mu\text{c}$ without any complicated calibration system as described.

Using large volume counters for lean meat determinations on the basis of K-40 measurements should be of no practical value for single parts of an animal (ham, for example) because the K-40 content is too low, and therefore the counting time too long for significant results. Things will improve if whole animals can be measured. For these studies a horizontal 2π counter or better two 2π counters which are movable against each other would be versatile

instruments. The correlation between fat-free weight and the potassium yield in ham found by other investigators could be confirmed. But to decide if this method can ever be used in practice for living animals is still a problem for biological research.

ACKNOWLEDGMENT

Our thanks are due to Mrs. Beate Pahl for skilled technical assistance and to Mr. D. G. Padval for carrying our chemical estimations.

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