The Determination of Doubly Labeled Artificial Soil in Detergency Studies

B. E. GORDON, W. T. SHEBS and R. U. BONNAR,
Shell Development Company, Emeryville, California

Abstract

A liquid scintillation method for the determination of carbon-14 and tritium labeled soil on white fabric has been developed. The method involves the use of an external standard to determine the counting efficiency for each isotope. The requirement that these determinations be made on swatches of fabric complicates the analysis because of their variable antiquenching effect. The method has been applied to cotton, Dacron, nylon, and Dacron/cotton.

A computer program has been written which accepts the raw counting data and performs all necessary calculations, printing out the data in a convenient form. The great convenience of the method has encouraged its application to large scale screening studies without sacrificing the precision, accuracy, or selectivity normally associated with small model systems.

Introduction

Before embarking upon an extensive study of detergency of various formulations under a variety of conditions, an analytical method was required to measure the effectiveness of the various detergents. Detergency studies are generally performed in a Terg-o-tometer (U.S. Testing Co., Hoboken, N.J.) on swatches of test fabric about 10 cm square. During the course of an 8-hr day the Terg-o-tometer, which is simply a battery of four beakers with stirrers in which the soiled test fabrics and the washing solution are placed, can generate several hundred samples. This sample load requires simplicity in sample preparation and automation in sample and data processing. Radiotracer methods coupled to liquid scintillation counting are ideal in this respect, as has been described previously (1).

Artificial soil, the material used to soil the test fabrics, has been the study of numerous investigations (2–4). Generally the compositions of these soils have not resembled closely the composition of natural sebum (5). Spangler (6) therefore employed a multicomponent soil which somewhat resembled natural sebum and in our work a similar soil was employed. Its composition was dictated to some extent by the availability of labeled compounds. The composition is shown in Table I. The tagged lubricating oil, representative of hydrocarbon in sebum, was prepared in this laboratory by exchange (7). Three passes over activated silica gel using hexane-benzene as eluant removed all surface-active by-products. All the other labeled compounds were obtained commercially and purified by gas-liquid chromatography (GLC) and tested for radiochemical purity by gas and thin-layer chromatography (TLC). The problem of radiochemical purity has been discussed before (1).

TABLE I

Composition of Artificial Soil

Composition	%W	Label
Lubricating oil: Shellflex-371-N	25	^{3}H
Tristearin	10	^{3}H
Peanut oil	20	Unlabeled
Stearic acid	15	^{14}C
Oleic acid	15	^{14}C
Octadecanol	8	^{14}C
Cholesterol	7	^{14}C

Doubly labeled soil was used in this work to study not only total soil removal by various detergent formulations, but selectivity of removal of the various components. The use of two radioisotopes would reduce the number of runs, an important consideration when about one hundred samples are generated per run.

Choice of Standardization Method

Liquid scintillation counting with its great sample capacity, completely automatic operation, and compatibility with modern computers was the obvious choice for radioassay. To reduce counting data to concentration one must obtain the disintegration rate (or a normalized counting rate) of the sample. This inevitably requires a standardization step to obtain the counting efficiency. For liquid scintillation counting there are three methods available, the pulse height shift method (PHSM), the internal standard method (ISM), and the external standard method (ESM).

The PHSM has been successfully applied to the determination of singly labled anionic surfactants on cotton fabric (1). This method involves dividing the beta ray spectrum of the isotope into two energy channels and measuring the ratio of count rates in the two channels. The change of this ratio with efficiency is obtained over a useful range and thus provides a calibration plot or function from which the efficiency is computed. For optimum performance two channels are required per isotope. Because our instrument (Packard Model 3003, Packard Instrument Co., Downers Grove, Ill.) has three channels, the PHSM was not considered further.

The ISM is the most widely used standardization method. It involves the addition of a spike of a standard to the sample after the initial count, and a recount. For doubly labeled samples two additions and three counts per sample are required: an unduly tedious procedure for a program generating hundreds of samples per week.

The ESM is the most recent and most promising of the three methods for double label work. The principle of the method is simple. Scintillations induced in a liquid scintillator by the Compton electrons from the gamma rays of a gamma emitting nuclide are quenched in a manner similar to the quenching of scintillations due to beta rays from the sample. Thus, if one locates a gamma emitter in a position where some of the radiation traverses the counting vial, one should obtain an incremental count whose magnitude is proportional to the efficiency. This approach has been described by Kaufman (8) who used a ^{57}Co needle and by others using ^{137}Cs, ^{144}Ce, ^{90}Sr (9), and ^{60}Co (10).

These reports demonstrated that, by proper adjustment of the energy channels of a liquid scintillation counter, one could indeed obtain a monotonic calibra-

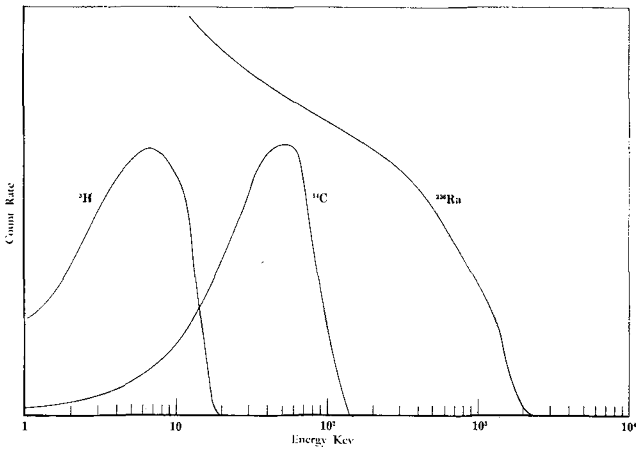

FIG. 1. Spectra of ^3H, ^{14}C and ^{226}Ra.

tion curve of external standard counts versus carbon-14 efficiency. This feature is now standard equipment on the modern liquid scintillation counter.

When counting a gamma emitter via its Compton electrons, one must bear in mind that a continuum of electron energies up to the gamma ray energy is present. Thus, if one is counting soft beta emitters such as tritium and carbon-14, there will be considerable interference by the external standard. This can be avoided by making two counts; one without followed by one with the external standard. Commercial modern liquid scintillation counters perform this operation automatically. Furthermore, by choosing a relatively energetic gamma emitter ($>$ 0.5 Mev) one can set up an energy channel for the external standard which suffers no interference from the lower energy betas. The energy relationship is shown in Fig. 1. Commercial liquid scintillation counters use ^{60}Co, ^{137}Cs, or ^{226}Ra as their gamma emitters.

Operation of the ESM

The sequence of steps in our counter is as follows: The sample is counted for a preset time and the data printed out. A 4 μc pellet of ^{226}Ra is then injected into a position close to the counting vial, an additional one minute count is made, the data again printed out, the pellet is ejected into a shielded container, and the sample is changed. A schematic of the external standard transfer system is shown in Fig. 2. In the work-up of the raw data, one ignores data in the external standard channel from the first count and data in the two sample channels from the second count.

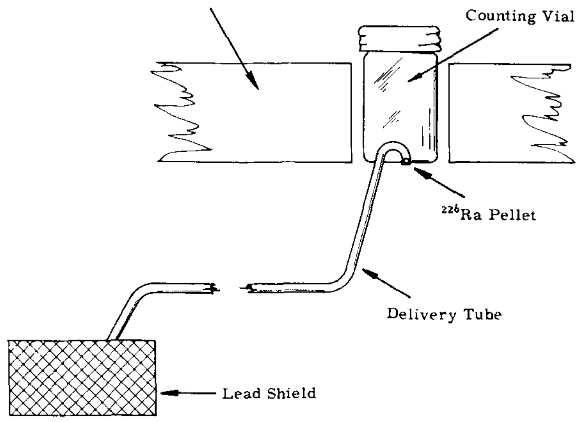

FIG. 2. External standard delivery system.

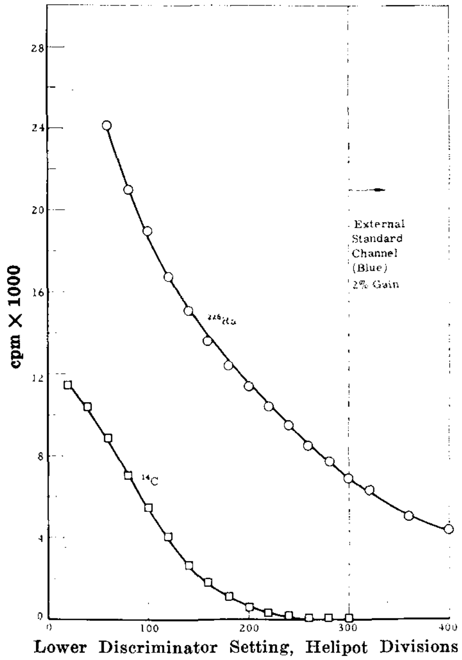

FIG. 3. ^{14}C and ^{226}Ra spectra in external standard channel.

Fig. 1 indicates that discrimination among the three isotopes should be fairly simple and that the only significant interference would be carbon-14 activity in the tritium channel. To determine precisely the proper location of the three channels, i.e., the amplifier gain and discriminator settings, linear plots of count rate versus discriminator setting were made of the three isotopes of interest at amplifier settings which would place significant portions of the spectra in the appropriate channel. In each of these plots only those two isotopes are included which might undergo mutual interference. Toluene-based liquid scintillator was used (6 g, PPO, 0.1 g dimethyl POPOP per liter A.R. grade toluene). Thus, in Fig. 3 the plot is of ^{226}Ra and carbon-14 at 2% amplifier gain, the setting for the ^{226}Ra. As can be seen, a discriminator setting of 300-∞ allows the ^{226}Ra to be counted with no contribution from carbon-14. Similarly, Fig. 4 shows

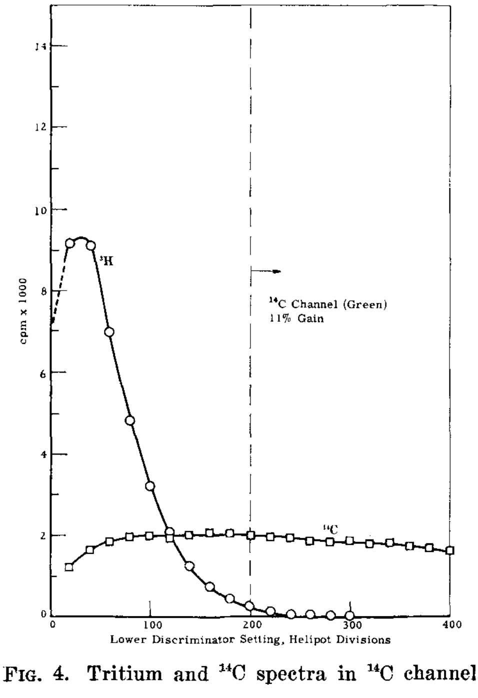

FIG. 4. Tritium and ^{14}C spectra in ^{14}C channel.

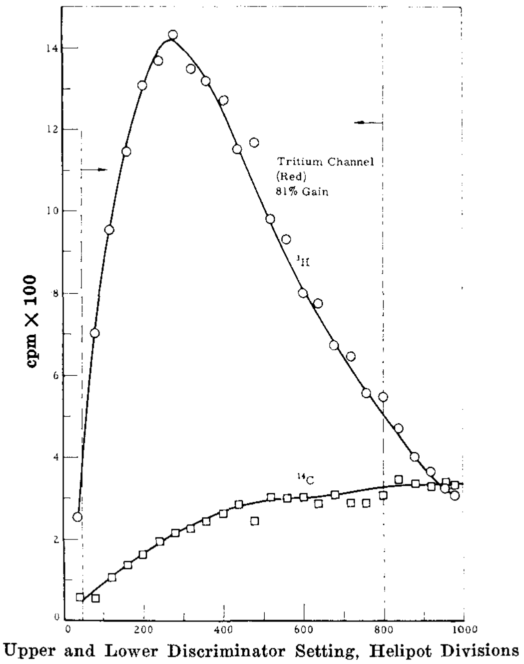

FIG. 5. Tritium and ^{14}C spectra in tritium channel.

tritium and carbon-14 in the carbon-14 channel at a gain of 11%. A lower discriminator setting of 200 effectively eliminates most of the tritium counts.

Fig. 5 demonstrates the spectra in the tritium channel where the gain is 81%. Here one must be more arbitrary in one's choice. For this purpose we desired a minimum tritium efficiency (unquenched) of 25% while keeping the carbon-14 overlap to a minimum. To satisfy this an upper discriminator setting in the tritium channel of 800 was selected. The final settings are presented in Table II.

The settings in Table II were used throughout this study. Periodically, the counter was recalibrated with a series of quenched carbon-14 and tritium standard samples. Over a period of four months, changes in the efficiencies vs ES count rate were less than 1%.

The above settings were made using clear solutions of toluene scintillator. The method, however, was to be applied to a heterogeneous system, swatches of white fabric immersed in the scintillator (1). It was, therefore, important to know whether the presence of fabric would affect significantly the counting efficiencies for all three isotopes. It was not unreasonable to expect some loss of efficiency with fabric present. Standard solutions of tritiated toluene and carbon-14 toluene were used.

The effect is surprising and large. The white fabric increases photon collection thereby increasing pulse height. The decrease in efficiency for carbon-14 in the tritium channel from 13.2 to 10.9%, results from the same effect, i.e., a shift of pulses from the low energy tritium channel into the higher energy carbon-14 channel (Fig. 4). The effect has been observed and discussed by Schwerdtl (11).

The increase in ^{226}Ra count rate suggests that since this reflects the enhanced efficiency for the two isotopes, one might possibly use the clear solutions as standards and obtain the efficiency by extrapolating the ES count rate vs efficiency curves. This does not prove to be the case as is demonstrated below (Fig. 7 and 8). Thus, the data in Table III and Fig. 7 and 8 established that standards for the analyses would have to contain fabric.

TABLE II
Instrument Setting for Double Label-ESM

Isotope	Channel	Amplifier gain, %	Window, V	Eff., %[a]	Interference
^{226}Ra	Blue	2	300–∞	....	None
^{14}C	Green	11	200–1000	58	0.2% of ^{3}H
^{3}H	Red	81	50–800	27	15% of ^{14}C

[a] In toluene scintillator.

TABLE III
Effect of Cotton Fabric[a] on Efficiency

Isotope	Fabric	Count rate in blue (external standard) channel	Efficiency in red (^{3}H) Channel, %	Efficiency in green (^{14}C) Channel, %
^{3}H	No	....	27.4	0.19
^{3}H	Yes	....	30.3	0.42
^{14}C	No	....	13.2	57.4
^{14}C	Yes	....	10.9	60.2
^{226}Ra	No	102,000	......	....
^{226}Ra	Yes	118,000	......	....

[a] About 0.5 g in 20 ml of scintillation solution.

Method Development

Soil of the composition shown in Table I should be completely soluble in the liquid scintillator. Failure to dissolve most, if not all, of the residual soil in the test swatch might result in low values due to self-absorption of the beta rays by the fabric. Several commonly used scintillators were tested for their solvency for soil at two temperatures. The data are in Table IV.

Toluene and ethylbenzene gave comparable count rates, with toluene slightly but not significantly lower. The others were significantly lower. Moreover, the expected ratio in count rates (^{14}C/^{3}H, basis known activity present) of 0.90 was observed with toluene and ethylbenzene. The nonhydrocarbon solvents showed lower values indicating some loss of the carbon-14 components. Temperature had only a minor effect. Toluene was selected because of its low cost and ready availability in high purity, a requirement for liquid scintillation counting.

The second point to be investigated was the ability of toluene scintillator to account quantitatively for soil present on the four fabrics to be used in the detergency studies. The fabrics: cotton, Style 405; nylon, Style 354A; Dacron, Style 754AW; Dacron/cotton, Style 7406—(Testfabrics, 55 Vandam St., New York, N.Y.). The ISM was used to avoid any variable effect of fabric type on efficiency. The ISM corrects for variations in each sample vial. Therefore, specimens of the four test fabrics (of equal area) were impregnated with known volumes of a standard benzene solution of soil, dried, immersed in 20 ml of toluene scintillator and analyzed by the ISM. The results are expressed in Table V.

The excellent agreement between added and recovered soil shows that the direct immersion of the fabric containing soil in scintillator is sufficient for

TABLE IV
Effect of Scintillation Solvent on Fabric Count Rate

Solvent[a]	Room Temp, cpm/mg		60°C for 10 min, cpm/mg	
	^{3}H	^{14}C	^{3}H	^{14}C
Toluene	1893	1708	1918	1732
Ethylbenzene	1929	1741	1956	1773
Dioxane (10% water)	1415	1246	1428	1223
Ethanol/Toluene (25/75)	1359	1103	1418	1173

[a] 1) Toluene scintillator composition, see above. 2) Ethylbenzene scintillator composition, same as toluene. 3) Dioxane scintillator composition, 80 g naphthalene, 6 g PPO, 0.1 g dimethyl POPOP per liter of 90% dioxane/10% water. 4) Ethanol/Toluene scintillator, same as toluene scintillator but having 25% v ethanol.

TABLE V

Recovery of Added Soil

Sample	mg Soil Added	mg Soil Found	Specific act., dpm/mg	
			^3H Fraction	^{14}C Fraction
Cotton, 0.65 g	2.04	2.00	5552	1200
	5.10	5.04	5643	1205
	10.20	9.85	5389	1208
	20.40	20.09	5497	1234
Nylon, 0.50 g	2.04	2.02	5491	1250
	5.10	5.02	5643	1198
	10.20	10.00	5448	1228
	20.40	20.26	5724	1204
Dacron, 0.40 g	2.04	2.03	5514	1249
	5.10	5.12	5648	1245
	10.20	9.95	5369	1233
	20.40	20.44	5657	1240
Dacron/Cotton, 0.30 g	2.04	1.96	5241	1224
	5.10	5.08	5782	1195
	10.20	9.95	5449	1218
	20.40	19.92	5583	1192

quantitative analysis. It also establishes that the ISM is an excellent referee method for the ESM. Calculations of precision from the data in the last two columns of Table V yield relative standard deviations for a single determination of 0.8% and 1.6% for tritium and carbon-14, respectively.

With the scintillation solvent and method established it was next desirable to ascertain whether the method would be affected by variations in fabric weight. This arose from the desire to use a single die to stamp out all samples. Because the different fabrics have different weave densities, swatches of the same size would have different weights. Using standard carbon-14 labeled toluene in toluene scintillator, increasing weights of the four fabrics were immersed in separate vials and counted. The efficiencies were then plotted versus weight with the results shown in Fig. 6.

Note there is clearly a significant effect of fabric type and weight on efficiency. Similar curves were found with tritium. However, one can obtain from Fig. 6 the probable errors introduced into the analysis if one were to use in the preparation of calibration standards a single fabric at a fixed weight. Thus, if one used 0.50 g of cotton or Dacron/cotton as the fabric component of the standard, the following errors would be introduced when analyzing weights of fabric stamped out by a single die:

Cotton	~0.5 g	0% Error
Dacron	~0.4 g	0%
Nylon	~0.4 g	−3%
Dacron/cotton	~0.3 g	+2%

The errors thus introduced were regarded as acceptable in view of the much greater variations introduced in the detergency studies themselves. The padding step, impregnating *large sections* of each fabric, showed a standard deviation range of 5–10%; and the washing step (4 replicates per cloth) showed a relative standard deviation range of about 5–35% over several hundred runs. Therefore, the set of permanent calibration standards used in this analysis contained 0.5 g swatches of Dacron/cotton.

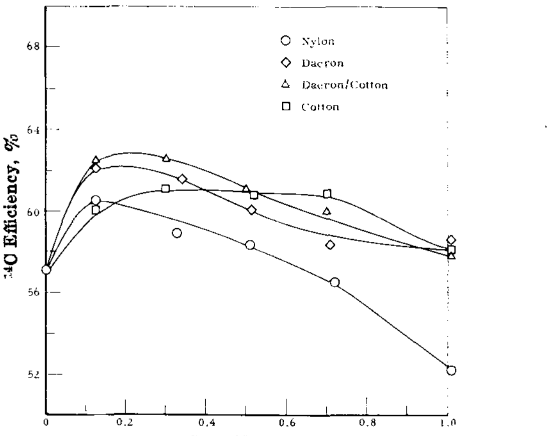

FIG. 6. Effect of cloth weight on efficiency.

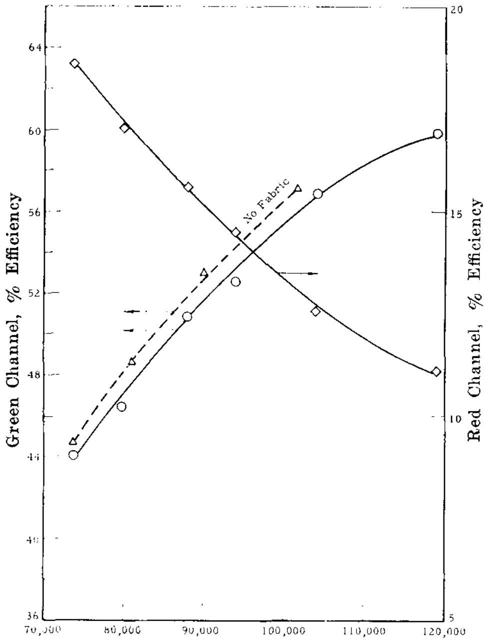

FIG. 7. ^{14}C standard quenching curves.

The Computer Based ESM

The ESM is simply a means of obtaining the counting efficiency, E, of each isotope in each channel. To compute the disintegration rate the simultaneous equations:

$$R = TE_{t,r} + CE_{c,r}$$
$$G = TE_{t,g} + CE_{c,g}$$

must be solved where *R* indicates observed count rate in red channel; *G*, observed count rate in green channel; *T*, tritium dpm; *C*, carbon-14 dpm; *E*, efficiency;

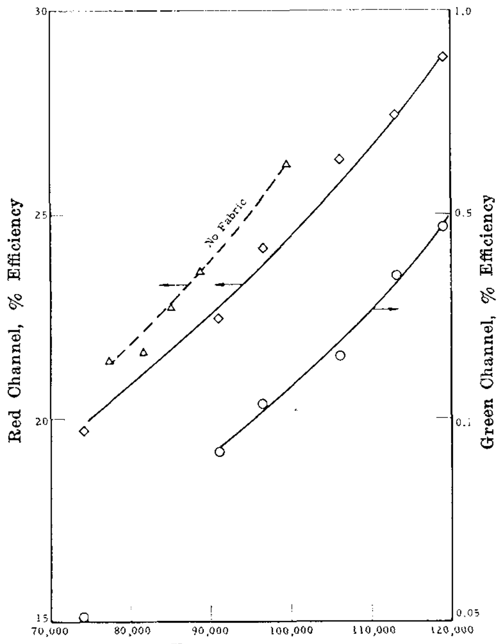

FIG. 8. Tritium standard quenching curves.

```
June 29, 1966
UNKNOWN SAMPLES
SAMPLE NO. 20341 SUB NO. = -0, SLOT NO. = 131, LABEL = *3112, SAMPLE WEIGHT = 0.390900
TRITIUM STD. DPM. = 21420000.0, C-14 STD. DPM. = 8250000.0

                 Net Counts Per Min.                       Efficiencies                      Disint./Min.         Del. Dpm.
Line  Time     Red      Green    Blue        G,C       G,H       R,C       R,H          Tritium     C-14     Tritium   C-14

  1   5.000  13099.4  6667.2  122555.0   0.600995  0.005376  0.107518  0.293371     40719.1   10729.4    -677.6   -18.6
  2   5.000  13453.0  6682.4  121792.0   0.600301  0.005211  0.108191  0.292059     42074.3   10766.5     677.6    18.6
   Means     13276.2  6674.8  122173.5   0.600648  0.005294  0.107855  0.292715     41396.7   10747.9
 Std. Devs.    250.0    10.7     539.5   0.000490  0.000117  0.000476  0.000928
MG. SEBUM/GM. *3112, TRITIUM, = 4.944
                        C-14, = 3.333

SAMPLE NO. 20341 SUB NO. = -0, SLOT NO. = 132, LABEL = *3212, SAMPLE WEIGHT = 0.400200
TRITIUM STD. DPM. = 21420000.0, C-14 STD. DPM. = 8250000.0

                 Net Counts Per Min.                       Efficiencies                      Disint./Min.         Del. Dpm.
Line  Time     Red      Green    Blue        G,C       G,H       R,C       R,H          Tritium     C-14     Tritium   C-14

  1   5.000  13102.8  6282.2  118409.0   0.596480  0.004515  0.111382  0.286075     41824.6   10215.5    1775.0   -96.2
  2   5.000  12583.6  6526.8  126832.0   0.603737  0.006353  0.104066  0.300473     38274.7   10407.9   -1775.0    96.2
   Means     12843.2  6404.5  122620.5   0.600108  0.005434  0.107724  0.293274     40049.7   10311.7
 Std. Devs.    367.1   173.0    5956.0   0.005131  0.001299  0.005173  0.010181
MG. SEBUM/GM. *3212, TRITIUM, = 4.672
                        C-14, = 3.123

SAMPLE NO. 20341 SUB NO. = -0, SLOT NO. = 133, LABEL = *3312, SAMPLE WEIGHT = 0.394100
TRITIUM STD. DPM. = 21420000.0, C-14 STD. DPM. = 8250000.0

                 Net Counts Per Min.                       Efficiencies                      Disint./Min.         Del. Dpm.
Line  Time     Red      Green    Blue        G,C       G,H       R,C       R,H          Tritium     C-14     Tritium   C-14

  1   5.000  12778.4  6429.4  122253.0   0.600728  0.005311  0.107783  0.292853     39824.7   10350.6     107.0     7.0
  2   5.000  12718.8  6421.2  122331.0   0.600798  0.005327  0.107714  0.292987     39610.7   10336.6    -107.0    -7.0
   Means     12748.6  6425.3  122292.0   0.600763  0.005319  0.107748  0.292920     39717.7   10343.6
 Std. Devs.     42.1     5.8      55.2   0.000049  0.000012  0.000048  0.000095
MG. SEBUM/GM. *3312, TRITIUM, = 4.705
                        C-14, = 3.181
```

FIG. 9. Example of output for unknown samples.

t,c refer to tritium and carbon-14; and *r,g* refer to the red and green channels. In practice one measures *R* and *G* and solves for *T* and *C*. Thus, we must obtain $E_{t,r}$, $E_{c,r}$, $E_{t,g}$, $E_{c,g}$ from the external standard. To this end two sets of sealed standards were prepared: one containing known amounts of carbon-14, the other known amounts of tritium. All had 0.5 g Dacron/cotton. Each set had increasing amounts of quenching agent (CCl_4) in order to define an efficiency versus external standard count curve over a range of efficiencies. Conventional, screw cap counting vials were used and the caps were lined with Teflon gaskets and sealed with epoxy cement. Both sets were then counted at the instrument settings in Table II using the automatic external standard feature. The data are plotted in Fig. 7 and 8.

Also included are plots when using clear solutions (dashed lines). The deviation from the solid calibration curves (with fabric) is significant particularly at the higher efficiencies and establishes the need for fabric-containing standards when analyzing fabric unknowns. The negative slope of carbon-14 in the red channel is due to the fact that as the pulse height is decreased (quenching) ^{14}C counts leave the higher energy (green) channel and enter the low energy (red) channel.

It was found that all four curves were adequately represented by a quadratic function of the form

$$E = a_0 + a_1x + a_2x^2$$

where E is the efficiency and a_0, a_1, a_2 are empirically determined constants and x = count rate in the blue channel × 10^{-4}. A computer program was written in Fortran IV to handle all computations required to reduce the counting data.

The computer program developed for this analysis first generates a four card calibration deck containing the constants for each calibration curve for each isotope in each channel. The calibration deck is then combined with the sample deck generated during the counting of unknowns, and the program proceeds to solve the simultaneous equations above. Additional computations are performed by the program to obtain data in the form desired, namely, mg soil/gram fabric. This, of course, requires additional data to be entered (e.g., fabric weight, specific activity of the ^3H and the ^{14}C fractions). Along with these data an additional punched card is generated for each sample. These cards are then further processed by a separate

TABLE VI
Comparison of ESM and ISM

Sample		mg Soil/g Fabric					
		^{14}C Fraction			^3H Fraction		
		ESM	ISM	Error, %	ESM	ISM	Error, %
Cotton	F–11	7.12	7.17	—0.7	7.94	8.01	—0.9
	–12	6.97	6.99	—0.3	7.81	7.59	+2.9
	–13	6.69	6.71	—0.3	7.70	7.51	+2.5
	–14	6.81	6.76	+0.7	7.81	7.56	+3.3
Nylon	F–15	8.65	8.90	—2.8	9.49	9.94	—4.6
	–16	8.46	8.62	—2.9	9.17	9.07	+1.1
	–17	8.57	8.70	—1.5	9.17	9.57	—4.2
	– 8	9.04	9.23	—2.1	9.44	9.71	—2.8
Dacron	F–19	8.21	8.15	+0.7	8.74	8.75	—0.1
	–20	8.04	8.09	—0.6	8.49	8.39	+1.2
	G–91	9.37	9.33	+0.5	9.98	9.83	+1.5
	–92	9.58	9.43	+1.5	10.19	10.13	+0.6

TABLE VII
Precision of ESM

Fabric	Tritium fraction, mg/g	Carbon-14 fraction, mg/g
Cotton	13.28, 13.23, 13.10, 12.99, 13.16 12.90, 12.98, 12.94, 13.28, 13.37	10.35, 10.29, 10.42, 10.38, 10.45 10.43, 10.56, 10.46, 10.52, 10.56
Mean	13.12	10.44
σ[a]	1.2%	0.9%
Nylon	13.13, 12.65, 12.35, 12.36, 12.42 12.37, 12.81, 12.51, 12.96, 12.78	10.32, 10.19, 10.27, 10.31, 10.36 10.30, 10.39, 10.35, 10.47, 10.48
Mean	12.63	10.34
σ	2.2%	0.9%
Dacron	13.08, 13.06, 12.90, 12.74, 12.78 12.76, 12.95, 12.09, 13.18, 13.16	10.39, 10.44, 10.50, 10.52, 10.47 10.46, 10.50, 10.50, 10.59, 10.61
Mean	12.97	10.50
σ	1.3%	0.6%
Dacron/cotton	13.13, 13.26, 12.99, 13.16, 12.81 12.74, 13.00, 13.14, 13.32, 13.33	10.26, 10.41, 10.34, 10.31, 10.54 10.46, 10.50, 10.50, 10.65, 10.64
Mean	13.09	10.44
σ	1.5%	1.1%

[a] Relative standard deviation, single determination.

program which expresses the data as percent soil removed and the errors associated with each value.

Comparison of the ESM and ISM

To assess the reliability of the computer-assisted ESM for routine samples it was compared to the ISM for the analysis of a number of samples. About one hundred such duplicates were run; Table VI presents typical results.

Agreement between the ESM and ISM is satisfactory, averaging about 2–3%. The error column presumes that the ISM is accurate (Table V) and so indicates the accuracy of the ESM. It is interesting to note that nylon shows the largest errors, mainly negative, an observation in accord with the predictions based on Fig. 6. However, errors of 4–5% are acceptable for reasons advanced previously.

To determine the precision of the method, samples of the four fabrics were impregnated with approximately the same amounts of doubly labeled soil, carefully dried, and analyzed by the ESM-computer method. Ten replicates of each fabric were so prepared and analyzed. The results are in Table VII.

The precision is good, about ±2% for tritium and ±1% for carbon-14 at this level. The precision, and of course, the accuracy would fall off as the concentration becomes very small or when a small amount of tritium is to be determined in the presence of a large amount of carbon-14. However, the requirements of the detergency studies are easily met by the precision and accuracy of the analysis. The method permits one to carry on extensive evaluation programs with a minimum of sample or data handling once the sample has been washed and cut. Double labeling allows one both versatility and economy in studying detergency using complex multicomponent soils.

REFERENCES

1. Gordon, B. E., W. T. Shebs, D. H. Lee, and R. U. Bonnar, JAOCS *43*, 525 (1966).
2. Hensley, J. W., M. G. Kramer, R. D. Ring, and H. R. Suter, JAOCS *32*, 138 (1955).
3. Hensley, J. W., and C. G. Inks, ASTM Special Tech. Bulletin No. *268*, 27 (1959).
4. Harris, J. C., R. E. Kamp and W. H. Yanko, ASTM Bulletin No. *170*, 82 (1950).
5. Warth, A. H., "The Chemistry and Technology of Waxes," Reinhold Publishing Co., New York, p. 138 (1956).
6. Spangler, W. G., JAOCS *41*, 300 (1964).
7. Gordon, B. E., and J. A. Van Klavern, Intern. J. Appl. Radiation Isotopes *13*, 103 (1962).
8. Kaufman, W. J., A. Nir, G. Parks and R. M. Hours, Proc. Univ. New Mexico Conf. on Organic Scintillation Detectors TID, 7612, 251 (1960).
9. Higashimura, T., O. Yamada, N. Nohara and T. Shidei, Intern. J. Appl. Radiation Isotopes *13*, 308 (1962).
10. Fleishman, G., and V. V. Glazunov, Pribory i Tekhn. Eksperim. *3*, 55 (1962).
11. Schwerdtl, E., Interm. J. Appl. Radiation Isotopes, *17*, 479 (1966).

[Received May 26, 1967]