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MEASUREMENT OF RADIOACTIVE CONTAMINATION BY WHOLE BODY SPECTROMETRY

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Abstract—This paper describes a whole body counter which has been used to measure contamination levels of persons over the past 3 years. The results from 2,600 measurements are discussed. The evolution of the mean caesium-137 body burden activity, from 1959 to date, is reported. The first results of a study on iodine-131 metabolism in order to evaluate the hazards of iodine contaminations are given. For continuous exposure the amounts of iodine-131 activity in the thyroid and in the rest of the body are in the ratio 86 : 14 implying a maximum permissible body burden of 0.16 μc iodine-131.

The workers of nuclear centres and of radioisotope laboratories are exposed to two types of irradiations—those due to external sources located near the personnel and those due to external or internal contaminations. The latter result from radioelements ingested or inhaled chronically or during an incident.

To determine the internal contamination of the personnel of the Centre d'Etude de l'Energie Nucléaire at Mol, a cell for the measurement of total internal gamma activity of men was installed in 1958. Approximately 2600 measurements have been carried out since this time and the data thus obtained enabled us to evaluate the possibilities and the importance of this device.

THE TOTAL BODY COUNTER

Basically the technique of the "tilting-chair" described by Marinelli in 1956[1,2] has been adopted. The man to be measured sits in a chair which allows a reproducible position in relation to the detector. Most of the body forms an arc with 35 cm radius around the crystal (Fig. 1). The detector consists of a large NaI(Tl) crystal of 20.3 cm diameter and 10.2 cm height. The gamma spectrum is recorded by a 256-channel pulse height analyzer with magnetic memory. The person to be measured and the detector are placed in a room shielded by 20 cm steel (inner dimensions 1.50 × 2.10 × 1.50 m; weight ~40 tons).

MEASURING AND CALIBRATION METHOD

The gamma spectrum is recorded on a diagram, and the contaminating radioelements are quantitatively determined by planimetric measurements of the number of pulses accumulated under a characteristic photoelectric peak. Results are interpreted in terms of the efficiency of the crystal for the radioactive gamma source, assuming a uniform distribution in the body. The calibration method generally adopted consists of simulating the human body by a dummy or by containers filled with a radioactive solution; this procedure applies to a particular radioelement.

This method has been used to obtain the photoelectric efficiency of our apparatus as a function of γ-ray energy in order to overcome the problem of estimating rapidly the quantity of a contaminating radioelement when an accident occurs. The importance of this curve has been shown by the wide variety of radioelements encountered during measurements made in the nuclear centre and also on personnel from various industries and university laboratories.

The yield curve has been established *in vitro* by means of isotopes of known activities whose gamma energies range from 140 keV to 2.75 MeV. These isotopes are diluted in a 70-l. solution simulating the body under the crystal

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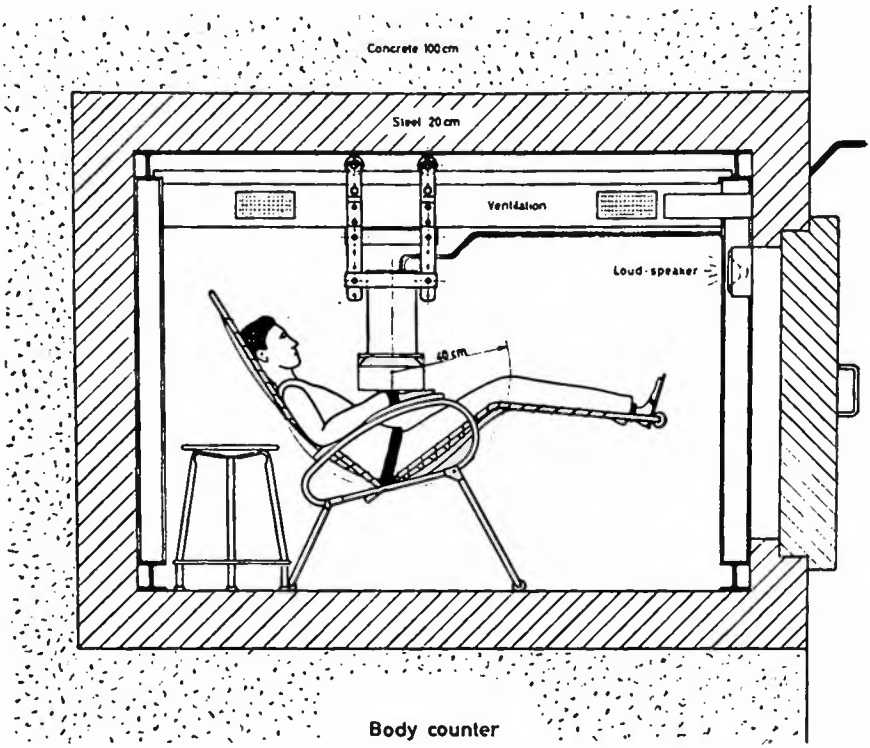

Fig. 1

(Fig. 2). The curve obtained $R(E)$ allows the determination of the activity present in the body if the probability of the gamma transition in the disintegration scheme is known. However it must be pointed out that the assumption that radioactive substances are non-uniformly distributed in the body may lead to erroneous results.

Calibrations were also carried out *in vivo* by means of ingested potassium-42 (1.52 MeV) and iodine-131 (364 keV).

Through this procedure the calibration *in vitro* was checked and the variations of the estimate due to individual differences were observed. In the case of potassium-42 a standard deviation of 5.8 per cent has been found. In the case of iodine-131, measurements were carried out $1\frac{1}{2}$ hr after ingestion, supposing that only negligible traces of radionuclide were still localised in the stomach at this moment. A lead shield removed the influence of the thyroid gland, the amount of iodine already present in this organ being determined by directional scintillation counting of thyroid uptake. For seven different individuals, the standard deviation was about 8 per cent.

Cesium-132 (670 keV; $T_{\frac{1}{2}} = 6.2$ d) would be another valuable isotope for such measurements.[3]

SENSITIVITY OF THE APPARATUS

The lower limit of detection has been set by the minimum spectrum surface to be measured as photoelectric peak, taking into account the

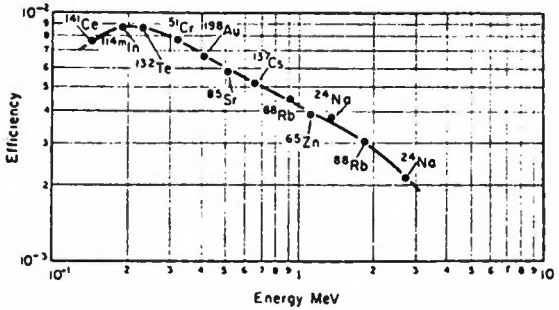

FIG. 2. Body-crystal efficiency of the human counter

Table 1

Limit of detection for different isotopes as a percentage of the maximum permissible body burden.

Radionuclide	Energy	Smallest detectable activity	
		μc	percentage of MPBB
Na^{24}	2.75 MeV	10^{-3}	0.01
Cr^{51}	323 keV	$1.5\ 10^{-2}$	0.002
Fe^{59}	1.10 MeV	$2.5\ 10^{-3}$	0.01
Co^{60}	1.17 MeV	$1.5\ 10^{-3}$	0.02
Zn^{65}	1.11 MeV	$3\ 10^{-3}$	0.005
Sr^{85}	513 keV	$1.5\ 10^{-3}$	0.0025
Zr^{95}	750 keV	$1\ 10^{-3}$	0.003
Sb^{124}	1.69 MeV	$1\ 10^{-3}$	0.01
Cs^{134}	797 keV	$1\ 10^{-3}$	0.005
Cs^{137}	662 keV	$1\ 10^{-3}$	0.003
La^{140}	1.60 MeV	$5\ 10^{-3}$	0.05
Au^{198}	411 keV	$1.5\ 10^{-3}$	0.008
Ra^{226}	(see Bi^{214})	$7.5\ 10^{-3}$*	7.5
(Bi^{214})	1.12 MeV	$2.5\ 10^{-3}$	—
	1.72 MeV	$2.5\ 10^{-3}$	
Th^{228}	(see Tl^{208})	$1.5\ 10^{-3}$†	7.5
(Tl^{208})	2.62 MeV	$2.5\ 10^{-4}$	—
$Sr^{90} - Y^{90}$	100–200 keV "bremsstrahlung"	$4\ 10^{-4}$	20

* Ra^{226}: 30 per cent of the decay products are assumed to remain in the body.
† Th^{228}: 50 per cent of the decay products are assumed to remain in the body.

natural activity of potassium-40 in the body and the background of the counter.

These limits, for 50-min measurements, are listed in Table 1. According to I.C.R.P. rules, they are generally a few hundredths of a per cent of the maximum chronic permissible radioactivity level for the human body except for thorium-232 and radium-226, owing to the very low tolerance levels for these radioelements. For strontium-90 the sensitivity is very low owing to the fact that it can only be detected through its "bremsstrahlung" spectrum.

RESULTS OF TOTAL BODY MEASUREMENTS

During the past 3 years, 2588 examinations were performed:

(1) as routine checks (R), the periodicity depending on the work of the person.
(2) after an exposure (E) resulting from a particularly hazardous operation with a high possibility of contamination and
(3) after an incident (I) involving the dispersion of radioelements.

Tables 2 and 3 summarize data of the measurements performed on the members of the various departments of C.E.N. or other institutions whose personnel have been measured in our shielded room. Among the C.E.N. personnel, several positive results were observed even for the routine checks where no ingestion of radioactive products could normally be assumed. The section "Radioisotope production" lists the greatest amount of contaminations; this is due to the high activities manipulated, the type of treatment applied to the radioactive products, the chemical operations and the various conditionings. Nevertheless most of the contaminations are lower than 1/10 of the maximum permissible level. For the whole C.E.N. personnel, out of 2010 examinations, 303 (i.e. 15 per cent) reveal contaminations of less than $\frac{1}{10}$ of the tolerance level and 25 (i.e. 1.25 per cent) are of a higher value. In the case of workers of other institutions, an even greater percentage of positive results has been observed —174 out of 578 (i.e. 30 per cent.) However, these measurements are generally requested after an incident.

In Tables 4 and 5 are listed the frequencies of occurrence of different radioelements. Iodine-131 is detected most frequently. Large amounts of this isotope are being produced at the C.E.N. and the high volatility of most of its compounds makes its manipulation most hazardous. Some recent incidents which could not be reported in the tables gave rise to contaminations of 30 times the maximum permissible level of iodine-131. Zinc-65 appeared frequently in the case of workers employed near the reactor BR-1; this was particularly observed with all persons exposed during the annealing of the reactor BR-1.* The zinc is probably carried into the reactor as a consequence of the erosion of plates in the ventilation ducts. Moreover, it seems worth while to notice also some contaminations by radium-226 whose tolerance level will be readily reached since it is fairly low (0.1 μc Ra^{226}). This proves the importance of a regular control of the personnel employed in laboratories where this radioelement is being handled.

NATURAL ACTIVITY AND RADIOACTIVE FALL OUT

A mean activity of 0.12 μc of K–40, the natural radioactive isotope, is found in the human body without any contamination. Moreover atomic explosions cause a general contamination of the population by cesium-137. The mean amount of this isotope reached a maximum in 1959 but considerably decreased afterwards owing to the cancelling of nuclear weapon tests; now it is increasing again. However, contaminations of local populations can reach mean values 10 to 50 times that observed in other regions.[6,7]

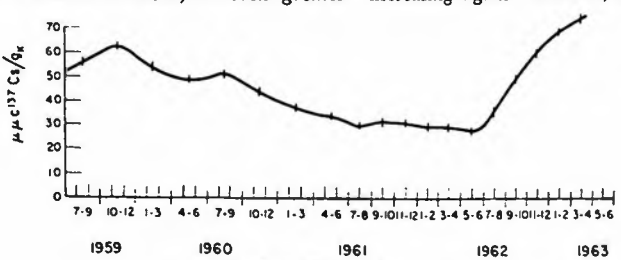

FIG. 3. Caesium-137 body burden due to fallout.

Another fission product, zirconium-95, has been detected recently in the lungs of various

* This phenomenon has been observed also by S. H. COHN *et al.*[5]

Table 2

Data from routine (R), after incident (I) or after exposures (E) examinations for C.E.N. employees showing observed contamination.

C.E.N Departments		Performed examinations	Positive examinations	Number of examinations in which radionuclides were detected at indicated level as percentage of the maximum permissible body burden.				Observed isotopes
				<1%	1 to 10%	10 to 100%	100 to 700%	
Radioisotope production	R	105	62	51	23	6(^{131}I)	4(^{131}I)	^{24}Na ^{56}Mn ^{60}Co ^{65}Zn ^{95}Zr — Nb ^{103}Ru ^{131}I ^{132}I ^{132}Te ^{137}Cs ^{140}Ba — La ^{198}Au ^{239}Np
	I	16	15	4	4	5(^{131}I)	5(^{131}I)	
	E	6	6	3	3	2(^{131}I)	—	
Chemistry	R	229	32	39	7	—	—	^{60}Co ^{65}Zn ^{95}Zr + Nb ^{103}Ru ^{131}I ^{132}I ^{132}Te ^{137}Cs ^{239}Np
	I	11	7	7	—	—	—	
	E	—	—	—	—	—	—	
Research reactor	R	129	29	35	—	—	—	^{59}Fe ^{60}Co ^{65}Zn ^{95}Zr — Nb ^{103}Ru ^{110m}Ag ^{122}Sb ^{124}Sb ^{131}I
	I	14	14	20	3	—	—	
Neutron physics	R	70	9	11	—	—	—	^{24}Na ^{57}Co ^{60}Co ^{65}Zn ^{95}Zr — Nb ^{115m}Cd ^{122}Sb ^{131}I
	I	2	1	2	—	—	—	
	E	2	2	2	—	—	—	
Metallurgy	R	187	12	16	—	—	—	^{60}Co ^{65}Zn ^{85}Sr ^{95}Zr — Nb ^{103}Ru ^{124}Sb ^{131}I ^{137}Cs
	I	7	1	5	—	—	—	
Measurements and radiation monitoring	R	257	32	36	4	2(^{131}I)	1(^{226}Ra)	^{51}Cr ^{60}Co ^{65}Zn ^{95}Zr + Nb ^{110m}Ag ^{131}I ^{137}Cs ^{203}Hg ^{201}Tl ^{226}Ra
	I	9	7	6	4	—	—	
	E	8	7	7	—	—	—	
Electromechanics	R	273	35	42	7	—	—	^{60}Co ^{65}Zn ^{95}Zr — Nb ^{103}Ru ^{124}Sb ^{131}I ^{137}Cs ^{198}Au
	I	9	6	5	2	—	—	
	E	24	20	22	—	—	—	
Solid state physics	R	81	3	3	—	—	—	^{65}Zn ^{95}Zr + Nb
	I	2	—	—	—	—	—	
Technology	R	104	6	5	1	—	—	^{51}Cr ^{60}Co ^{65}Zn ^{131}I
Mathematics	R	61	9	10	1	—	—	^{65}Zn ^{103}Ru ^{131}I
Medical	R	72	2	2	—	—	—	^{60}Co ^{65}Zn
Administration	R	253	5	6	—	—	—	^{60}Co ^{65}Zn ^{85}Sr ^{95}Zr–Nb ^{103}Ru ^{131}I
	I	1	1	1	—	—	—	
	E	1	—	—	—	—	—	
Reactor BR3	R	19	1	2	—	—	—	^{95}Zr–Nb ^{103}Ru
Radiobiology	R	54	—	—	—	—	—	^{131}I
	E	4	4	4	—	—	—	

Table 3

Examinations performed on employees of external establishments.

Establishment	Performed examinations	Positive examinations	Number of examinations in which radionuclides were detected at indicated level as percentage of the maximum permissible body burden				Observed isotopes
			<1%	1–10%	10–100%	100–900%	
Universities	60	28	20	10	13 (^{226}Th)	—	^{60}Co ^{65}Zn ^{131}I ^{137}Cs ^{230}Th
Industries	110	72	70	30	2(^{226}Ra) 16(^{131}I)	5(^{226}Ra)	^{54}Mn ^{56}Co ^{60}Co ^{65}Zn ^{82}Br ^{95}Zr + Nb ^{103}Ru ^{110m}Ag ^{131}I ^{137}Cs ^{192}Ir ^{198}Au ^{203}Bi ^{206}Bi ^{226}Ra
Nuclear centres and laboratories	387	73	69	13	1(^{232}Th) 1(^{131}I)	2(^{231}I)	^{60}Co ^{65}Zn ^{95}Zr + Nb ^{103}Ru ^{131}I ^{134}Cs ^{137}Cs ^{139}Ce ^{226}Ra ^{232}Th
Other	21	1	1	—	1(^{226}Ra)	—	^{137}Cs ^{226}Ra

persons who were not connected in any way with radioactive laboratories.

STANDARDS FOR IODINE-131

Contaminations by this radioactive element are obviously as frequent among the C.E.N. employees as among the staff of other institutions. Moreover its presence in the fall-out of nuclear explosions or during a reactor incident (Windscale, 1957) urged us to study the metabolism of this radioelement which concentrates in the thyroid gland.

A theoretical model of iodine metabolism has been proposed by BROWNELL (Fig. 4).[9] The basic assumption is that the daily uptake by the thyroid gland is a constant amount of 70 μg of iodide and its elaboration into iodinated hormones; moreover, it has been accepted that the exchanges between the different compartments occur at constant rates.

The mathematical analysis of these processes leads to a set of four differential equations which has been established and applied to the case of a single or chronic contamination by iodine-131, assuming a daily food intake of 100–300 μg of iodide. These equations have been solved on a digital computer and the results were plotted on a graph. (Fig. 5).

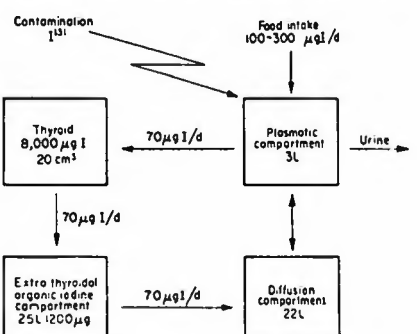

FIG. 4. Four-compartments model for iodine metabolism.

The urinary excretion of iodide causes a fast decrease of the body radioactivity whereas the maximum activity is reached in the thyroid gland some 24 hr after the ingestion. After some days an increase of the internal radioactivity, due to the release of iodine-131 labelled thyoxine by the thyroid gland is observed.

In the case of chronic contamination by iodine-131, the calculations show that the distribution of the radioactivity is as follows:

(1) 86 per cent in the thyroid gland,

Table 4

Radionuclides observed during examinations performed on C.E.N. employees.

Radionuclides	Number of investigations in which a radionuclide was detected at indicated level as a percentage of the maximum permissible body burden				
	Total	<1%	1–10%	10–100%	100–900%
^{24}Na	2	2	—	—	—
^{51}Cr	2	2	—	—	—
^{56}Mn	1	1	—	—	—
^{59}Fe	2	2	—	—	—
^{57}Co	1	1	—	—	—
^{60}Co	46	45	1	—	—
^{65}Zn	124	124	—	—	—
^{85}Sr	2	2	—	—	—
^{95}Zr–Nb	34	34	—	—	—
^{103}Ru	27	27	—	—	—
^{110m}Ag	3	3	—	—	—
^{115m}Cd	1	1	—	—	—
^{122}Sb	2	2	—	—	—
^{124}Sb	12	11	1	—	—
^{132}Te	4	3	1	—	—
^{131}I	144	68	52	15	9
^{132}I	5	2	3	—	—
^{134}Cs	3	3	—	—	—
^{137}Cs	15	14	1	—	—
^{140}Ba–La	1	1	—	—	—
^{198}Au	4	4	—	—	—
^{203}Hg	1	1	—	—	—
^{204}Tl	1	1	—	—	—
^{226}Ra	1	—	—	—	1
^{239}Np	3	3	—	—	—

(2) 14 per cent in the body (for a food intake of 150 μg daily).

For the calculations developed by the I.C.R.P committee, iodine was assumed to have a biological half-life of 138 days, both in the thyroid gland and in the body, but only the thyroid organic iodine has this half-life and a considerable fraction in the form of iodide is eliminated by urinary excretion with a biological half-life of a few hr. Consequently, if a maximum dose of 8 rems per 13 weeks is admitted for the thyroid, the maximum permissible amount permanently allowed in the body is 0.16 μc (0.14 μc in the thyroid gland) and not 0.7 μc as proposed by the I.C.R.P.

The validity of our theoretical results has been confirmed by experiments on eight male volunteers weighing 59–80 kg and 23–38 years old; 0.5 μc of iodine-131 as carrier free sodium iodide in 20 cm^3 $Na_2S_2O_3.10^{-3}$ M solution were ingested by each individual, fasting for 12 hr. Results of two typical measurements of the radioactivity in the thyroid gland and in the body are shown in Fig. 6. They agree fairly well with the calculated curve if one assumes a daily food intake of 100 μg of iodide. The curve observed in a slightly hyperthyroid subject clearly illustrates the release of iodinated hormones. A complete discussion of these results will be published shortly.

Thus the proposed model appears valid and the conclusions drawn concerning the maximum permissible radioactivity level lead to a revision of I.C.R.P. values for maximum permissible iodine body burden.

CONCLUSION

Owing to its sensitiveness and its selectivity, a total body counter is essential for the control

Table 5

Radionuclides observed in investigations performed on employees of external establishments.

Radionuclides	Number of investigations in which a radionuclide was detected at indicated level as a percentage of the maximum permissible body burden				
	Total	<1%	1–10%	10–100%	100–900%
^{54}Mn	1	1	—	—	—
^{58}Co	3	3	—	—	—
^{60}Co	25	23	2	—	—
^{65}Zn	13	13	—	—	—
^{82}Br	1	1	—	—	—
^{95}Zr–Nb	1	1	—	—	—
^{103}Ru	2	2	—	—	—
^{110m}Ag	1	1	—	—	—
^{131}I	96	42	35	17	2
^{134}Cs	11	11	—	—	—
^{137}Cs	57	56	1	—	—
^{139}Ce	3	3	—	—	—
^{192}Ir	4	4	—	—	—
^{198}Au	5	4	1	—	—
^{203}Bi	2	1	1	—	—
^{206}Bi	2	1	1	—	—
^{226}Ra	10	—	2	3	5
^{228}Th	23	—	10	13	—
^{232}Th	1	—	—	1	—

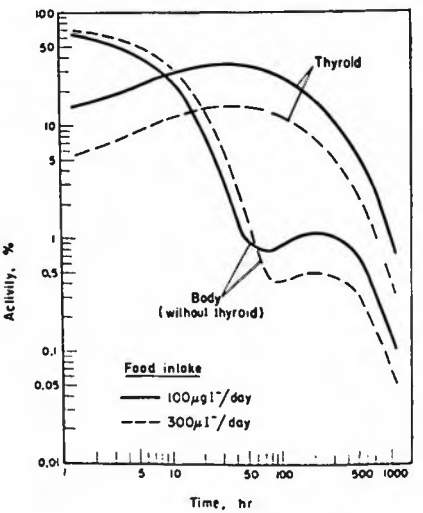

FIG. 5. Iodine-131 activity (theoretical curves).

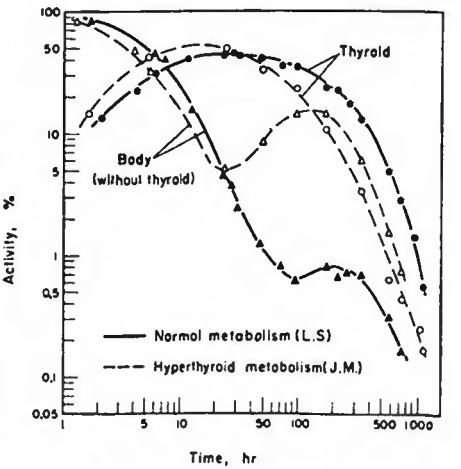

FIG. 6. Iodine-131 activity (experimental curves).

of the personnel in institutions where unsealed sources of radioactive products are being handled, not only in the case of an accident but also as a preventative by means of routine checks.

Such a device was successfully used to study the iodine metabolism and to check the validity of a model proposed for its turn-over.

Results obtained lead us to propose new standards for MPL of iodine-131.

REFERENCES

1. L. D. Marinelli, *Brit. J. Radiol. Suppl.* 7 (1956).
2. C. E. Miller, Argonne National Laboratory, A.N.L. 5596 (1956).
3. R. A. Dudley, IAEA Circular Sc/751–3, Vienna (1961).
4. R. R. Boulenger, J. F. Colard and J. Henry, *Whole Body Counting*, p. 310–320. Proceedings Series IAEA, Vienna (1962).
5. S. H. Cohn, R. A. Love and E. A. Gusmano, *Science* **133**, 1362 (1960).
6. K. Liden and Y. Naverstenn, Second Symposium on Radioactivity in Man, Chicago 1962 (unpublished).
7. H. E. Palmer, W. Hanson and B. I. Griffin, *Ibid.* (unpublished).
8. J. Rundo, *Ibid* (unpublished).
9. G. L. Brownell, *J. Clin. Endocrinol.* **11**, 1095 (1951).
10. D. S. Riggs, *Pharm. Rev.* **4**, 285 (1952).