Fallout gamma-emitting radionuclides in air, precipitation, and the human body up to spring 1967

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ABSTRACT

Results from measurements of some radionuclides (especially cesium-137) in stratospheric air, ground level air, precipitation, and the human body are discussed. First during 1967, four years after the cessation of large scale atmospheric testing in late 1962, the supply of stratospheric cesium-137 to the ground will be balanced by decay of the activity already deposited. Average cesium-137 body burdens have since the maximum values occurred during 1964 been halved in every 1.5 year. Internal and external gamma-doses have been calculated. Measured ratios between some other long-lived radionuclides and cesium-137 are discussed. Some beryllium-7 concentration values are reported.

Introduction

The testing of nuclear devices, which was very extensive especially during 1958, 1961 and 1962, has introduced large amounts of radioactive debris at various heights in the atmosphere. National networks for routine sampling and analyses of the debris have been set up in many countries. In Sweden sampling of air and precipitation is mainly carried out by the Research Institute of National Defence. Data from radioactivity measurements Oct. 1961 to Dec. 1966 (in some cases June 1967) are discussed below. Earlier data are given by Edvarson & Löw (1960) and Lindblom (1962).

It has been of special interest to follow the appearance and transport of the radionuclide cesium-137, which together with iodine-131 and strontium-90 is of significant biological importance. After the large-scale atmospheric test series of 1961 and 1962, cesium-137 fallout became a health-physics problem in some areas, where certain conditions favor concentration processes. Several extensive studies have thus been carried out in northern Scandinavia, where reindeer meat is an important diet component in a small population group. This situation is followed in Sweden especially at the University of Lund (Lidén, 1967). Body burdens of cesium-137 are measured with whole-body counting techniques at several places. Data up to Sept. 1967 for one of the control groups measured at this institute are discussed below. Special investigations of the atmospheric transport of fallout-radioactivity and the radioactive characteristics of debris from single nuclear explosions have also been undertaken (Lindblom, 1961; Persson, 1966, 1967).

Cesium-137 in air and precipitation

Sampling

Air at ground level: two sampling devices with capacity of about 9×10^3 m³/day each are since Apr. 1964 in operation in the Stockholm region. The sampling is carried out with glass-fibre filters which are changed 3 times a week. First gamma-spectrometric measurement follows 3–4 days after changing, and pooled monthly samples are measured first time 15–30 days and then about 6 months after the end of the sampling month.

Precipitation is sampled in large heated funnels (2 m diameter) at 6 sampling stations in

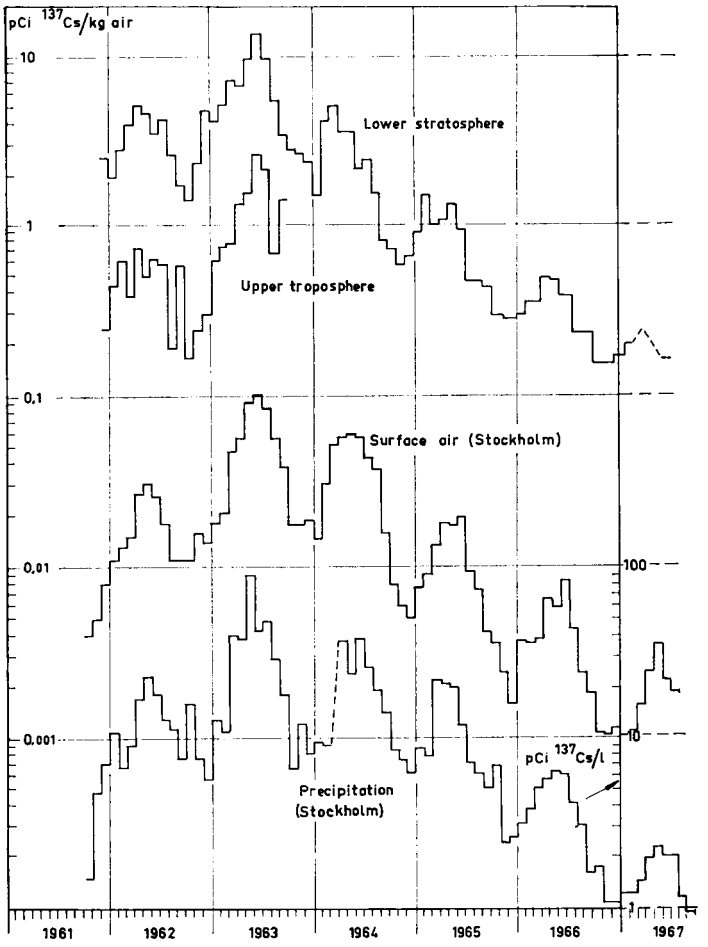

Fig. 1. Cesium-137 concentrations in air and precipitation (decay corrected to Feb. 1964).

different parts of the country. The rain water is filtrated with ion-exchanger technique.

High altitude air sampling is carried out in co-operation with the Swedish Air Force. An improved installation on the aircrafts now makes it possible to filter about 400 kg of air at each of the two sampling altitudes (1 km below and 1 km above actual tropopause level) at every sampling occasion. The mixed monthly samples normally represent about 1100 kg of air; the total sampling-time varies from 2 to 8 hours.

The measurements have been carried out with highly standardized methods (gamma-scintillation technique). The error of measurement and analysis is lower than the error introduced by the uncertainties in determination of sampled volumes which can be estimated to about ± 10 %.

Concentration data

Results are presented in Fig. 1 and Table 1. Note that the data in Fig. 1 are corrected for decay to Feb. 1964. The 1967 data (ground level air, precipitation) are supplied by B. Bernström at this institute.

Discussion

When the first massive injections of radioactive debris occurred in 1958 very little was known about the storage qualities and transport mechanisms in different parts of the atmosphere. The debris turned out to be a valuable tracer, and many detailed interpretations of the

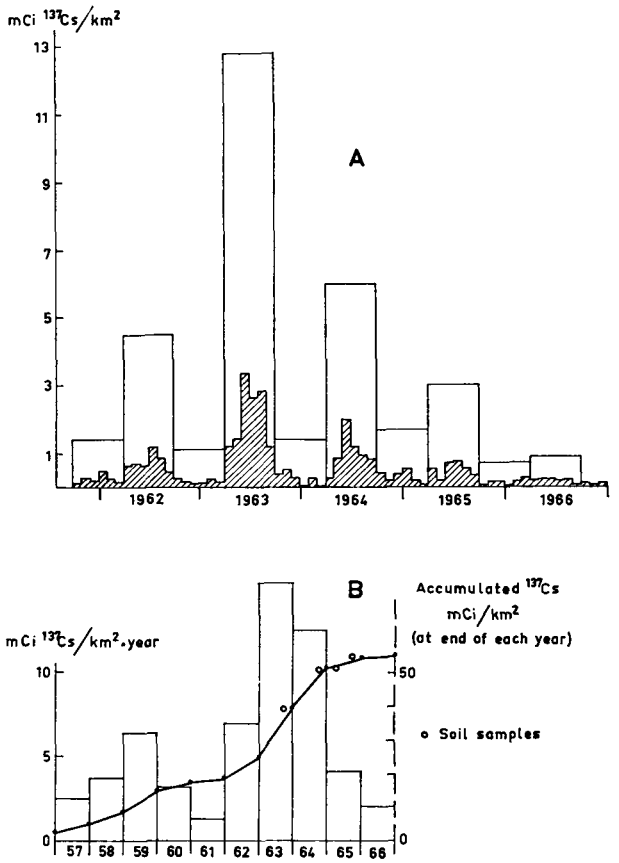

Fig. 2. (*A*) Monthly cesium-137 deposition in Stockholm (decay corrected to Feb. 1964); (*B*) yearly average cesium-137 deposition in Sweden.

increasing amount of reliable data have been presented (e.g. Gustafson, 1964; Peirson & Cambray, 1965). An evaluation of the stratospheric transport problem in the light of strontium-90 data from the U.S. High Altitude Balloon Sampling Program has recently been published (Krey, 1967).

The lower stratospheric air-sampling in Sweden is carried out at latitudes about 50–68° N and near the tropopause level. The monthly mean values show peak activity during the early months of the years 1964–1966 followed by the well-known spring maximum in ground level air.

This seasonal trend is somewhat different from what is reported by Krey for higher altitudes in the stratosphere, 23–32 km. His data suggest that vertical mean motion is the most important mechanism of tracer transport at these altitudes. The concentration variations in the lowest part of the stratosphere are however not in disagreement with this theory, as the maximum concentration layer occur at about 18 km, and an average sinking of air during winter then will cause higher activity-values in the air at lower levels. The intensified turbulent exchange of air etc during late winter and early spring is also likely to play an important role in the region of the tropopause.

For all levels in the atmosphere radioactivity data 1964–66 show that the time required for a 50 % reduction of the concentration is about 8 months which means a yearly decrease with a factor 3.3. If there is no additional significant injection of debris an extrapolation of data suggests that the 1968 peak cesium-137 concentration in ground level air will be in the order of 1 percent of the 1963 maximum values.

Table 1. *Cesium-137 in air and precipitation*

Activity values corrected for decay to mid month

Month		Cesium-137 concentrations: Ground level air sampling: Stockholm, 5×10^5 m³/month (pCi/1000 m³)	Precipitation: Average of 6 stations (pCi/l)	Total monthly sample (l)	Cs-137 deposition: Average of 6 stations (mCi/km²)
1964	10	10.3	7.5	995	0.40
	11	7.8	5.5	635	0.27
	12	6.5	6	875	0.18
1965	1	8.2	8	680	0.29
	2	10.2	6	395	0.12
	3	15.1	6.5	435	0.15
	4	20.4	15	415	0.33
	5	19.8	14	490	0.36
	6	21.8	11	1040	0.60
	7	10.6	7.5	1680	0.65
	8	8.2	6.5	1360	0.45
	9	4.6	4.5	1130	0.26
	10	4.0	4	285	0.06
	11	2.6	2	925	0.11
	12	1.7	2	925	0.09
1966	1	4.0	2.5	245	0.03
	2	3.9	4	585	0.12
	3	4.2	5	830	0.22
	4	7.0	5.5	570	0.16
	5	6.3	6	600	0.20
	6	8.9	5.5	720	0.21
	7	4.5	3.5	1120	0.22
	8	2.5	3	1220	0.20
	9	1.9	1.5	750	0.06
	10	1.0	1	810	0.05
	11	1.0	1	980	0.04
	12	1.1	1	1130	0.06

In Fig. 1 are also shown average monthly concentration values of cesium-137 in precipitation. A computation of the so called wash-out factor (pCi per l/pCi per kg air, not labelled on figure) gives no trend 1964–66, indicating that a change in effectivity of washout (which could be attributed to e g a systematical variation of particle sizes) seems not to have occurred during this period.

Deposition data

Results in Fig. 2 and Table 1. Note that the data in Fig. 2*A* are corrected for decay to Feb. 1964. The data in Fig. 2*B* including accumulation are decay-corrected to the end of each year.

Discussion

The seasonal variation is well established also in the deposition data. The deposition rate is however to a certain degree dependent on the actual precipitation pattern, and thus its maximum occurs later in the year (June–July) than the corresponding concentration data. Volchok (1967) reports an earlier maximum of the deposition rate (April) in New York City at 41° N. This difference may reflect a latitudinal effect or is just caused by a different time-distribution of the rain.

The relation between average deposition amounts during summer (Apr.–Sept.) and the following winter has been remarkably constant during the whole period since the resumption of large-scale testing in autumn 1961, the quotient being about 5. This is not necessarily a significant relationship but may be the result of chance combinations of concentration and precipitation data. It gives however an indication of how the summer half-year dominates the deposition picture.

Yearly averages of cesium-137 deposition 1957–1966 in Sweden have been estimated from average concentrations and climatologically defined mean precipitation amounts. The resulting accumulated cesium-137 level reaches 55 mCi/km² at the end of 1966 (Fig. 2 *B*), radioactive decay included. A number of soil samples have been measured to test the reliability of the precipitation sampling. The agreement indicates that the funnel method is effective in sampling also the dry component of the total fallout.

If there is no further significant injection of radioactivity into the atmosphere, decay and supply of cesium-137 to the ground will nearly balance each other during 1967. Thereafter the cumulative deposit will begin to decrease for the first time since testing of high-yield nuclear devices began in 1954.

The area of Sweden is 450,000 km² or 0.17 per cent of the area of the hemisphere. The total Swedish cesium-137 fallout thus can be estimated to about 0.025 MCi. The globally—mostly in the northern hemisphere—deposited amount can in turn be estimated to about 19 MCi from reported 12 MCi world-wide strontium-90 deposition (Volchok, 1967) and the experimentally found average cesium-137/strontium-90 ratio 1.6 (Scott Russell, 1966). The figure 19 MCi is also in good agreement with an estimated production of 0.16 MCi/Mton fission. The ratio of the Swedish cesium-137 fallout to the hemispheric thus nearly equals the ratio of the areas, which seems reasonable taking into consideration the geographical and climatological situation of the country.

Results from measurements of cesium-137 in the Baltic sea have been reported from Finland (Salo, 1967). The total amount has been estimated to about 0.021 MCi, which is about 50 mCi/km². To this should be added the activity of the bottom sediments. Current measurements in Sweden (Hannerz & Schelin, 1968) suggest that the additional activity is of the order of 10 mCi/km². Though these figures are very approximate they suggest that no greater differences exist between deposition on ground and deposition on sea surfaces in Scandinavia.

Other long-lived gamma-emitting nuclides in air and precipitation

The concentrations of cerium-144, antimony-125, rhodium/ruthenium-106 and manganese-54 have been determined in air and precipitation since 1962. The ratios of these nuclides to cesium-137 are shown in Figs. 3–4. All data are corrected for decay to Feb. 1964. Extensive evaluations of similar data have been carried out by e.g. Gustafson *et al.* (1964) and Peirson & Cambray (1965). Here will only be presented some suggestions of possible interpretations.

The Ce/Cs-ratio of 1964–1965 in ground level air (Fig. 3) gives a mean production month of July 1962. The slightly increasing tendencies of Sb/Cs, Ce/Cs and Ru/Cs during 1965–1966 may indicate an increased domination of debris from the test series of late 1962. Mn is said to have been produced mainly in connection with high-yield shots during the USSR 1961 tests (Gustafson, 1964). The Mn/Cs ratio is remarkably constant during 1964–1966 suggesting that the mixing of 1961 and 1962 debris was completed already in 1964. The high Mn/Cs ratios during the second half of 1963 indicate that the debris from the 1961 high yield tests at very high latitudes (74° N) not until 2 years after injection reached ground-level air in considerable quantities. The Mn/Cs-ratio at time of production can be estimated to about 5:1 from the 1964–1965 data which is not unreasonable.

Studies of fractionation effects in globally distributed debris (Edvarson, 1959) show that larger particles (> 1.0 μm) usually are improverished in Ru. A possible explanation of the comparatively high Ru/Cs ratios during the second half of 1963 therefore is that a larger amount of the 1961 debris with small average particle sizes was introduced at that time. This is in concordance with the interpretation of the Mn/Cs ratios mentioned above.

Some interesting differences between the data of Figs. 3 (ground level air) and 4 (precipitation) remain to be explained. The reason for the lower Sb/Cs and Ce/Cs ratios in precipitation during 1962 and 1963 (Sb/Cs) may be that the spectrum of particle sizes is wider during and immediately after test periods. The conclusion regarding the Ru/Cs ratios in ground level air could also be applied on the very low Ru/Cs ratios which are occasionally measured in precipitation at test periods, suggesting a domination of large particles in wet fallout. Studies of the washout and rainout processes show that a small shift in average particle diameter may have great influence on the effectivity of the processes. The interpretation of these and other

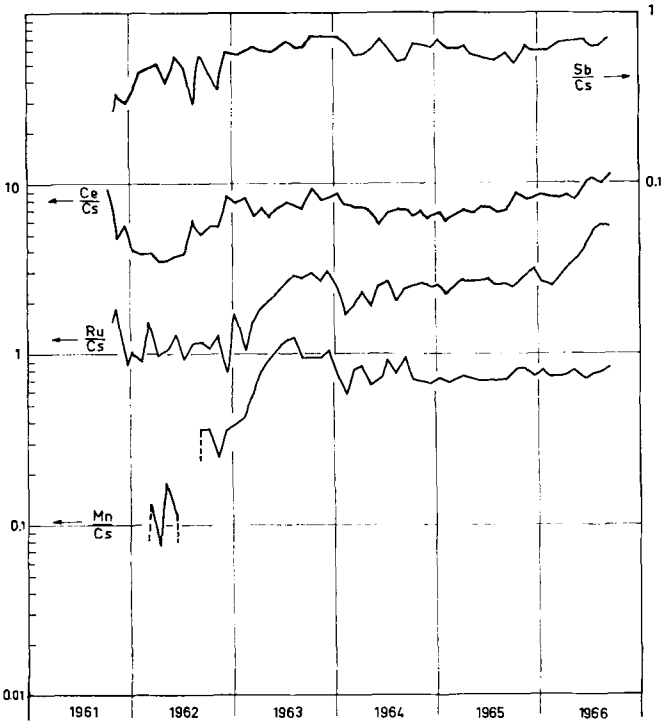

Fig. 3. Ratios of antimony-125, cerium-144, ruthenium-106 and manganese-54 to cesium-137 in ground level air. Decay corrected to Feb. 1964.

data of the same kind (cf. Dingle & Gatz, 1966) is however very complex.

The deposited amount of radionuclides in soil samples from the Stockholm region have also been determined (Table 2). The yearly precipitation amount is about 500 mm.

Detection of gamma-emitting nuclides shortly after nuclear tests

The additional deposition of radionuclides with shorter half-lives resulting from isolated nuclear tests in the atmosphere 1963–1966 has been determined weekly or monthly. The third Chinese explosion in May 1966 thus caused an additional fallout during May–Aug. as shown in Table 3 together with data from the cosmic-ray produced beryllium-7.

Radioactivity with a composition characteristic of debris from a vented underground nuclear explosion was detected in ground level air and precipitation during the latter part of Dec. 1966. Gamma-spectrometric and radiochemical examination showed an enrichment of strontium-89, strontium-90, cesium-137 and barium/lanthanum-140 (Persson, 1967). The maximum concentrations occurred in Finland (Kauranen *et al.*, 1967).

Beryllium-7 concentrations in air and precipitation

The concentration of beryllium-7 has been determined in the monthly samples 1960–1961 (Lindblom, 1962) and 1964–1966 (Fig. 5). The values are ranging from 0.02 to 0.08 pCi/kg of ground level air (monthly averages) and from 10 to 60 pCi/l precipitation. Total deposition was in the order of 1.5 mCi/km²/month.

A determination of the beryllium-7 concentration in the lower stratospheric sample of June 1967 gives about 2.5 pCi/kg. The ratio between concentrations in stratospheric and ground level air is thus about the same as the corresponding value for cesium-137. Beryllium-7 was produced also during the nuclear tests of 1961 and 1962

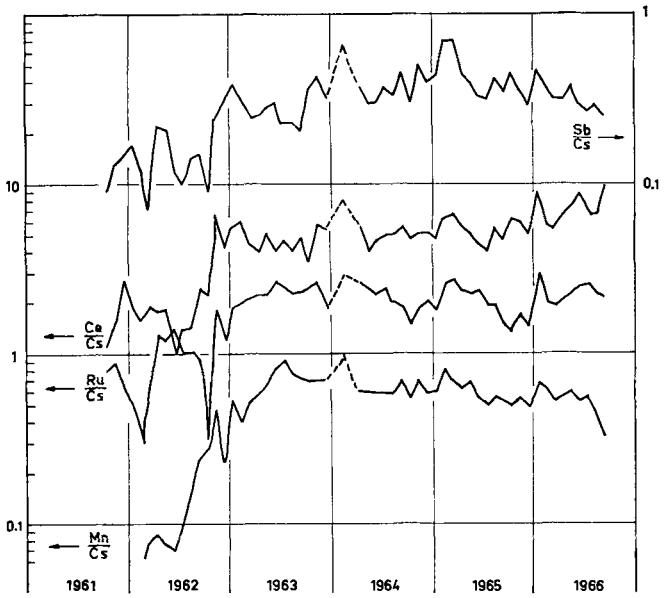

Fig. 4. Ratios of antimony-125, cerium-144, ruthenium-106 and manganese-54 to cesium-137 in precipitation. Decay corrected to Feb. 1964.

(Bleichrodt & Abkonde, 1963). As its physical half-life is 53 days the contribution from these tests became negligible during 1964.

Cesium-137 in the human body

The first whole-body counter with Na(Tl)I-detection system in Scandinavia (HUGO) was built in 1956 by the Research Institute of National Defence and the Atomic Energy Company in co-operation. The Company moved in 1963 large parts of the research work to Studsvik where a new body counter was constructed (HUGO II).

The detection equipment of the original HUGO has since then become modernized. An experiment with a controlled intake of cesium-137 in a male person was carried out in 1964 in connection with the recalibration of the body counter (Lindblom, 1965). The biological half-life turned out to be 115 days. This is considerably longer than indicated by results from other Scandinavian measurements of "normal" body burdens and dietary intake. The experiment also pointed out some difficulties involved in drawing conclusions about body burdens and biological half-lives from urinary analysis.

A small control group from the Stockholm region was at that time set up and has been measured 3–4 times a year. Five male persons with no extreme dietary habits were specially

Table 2. *Soil samples*

Month	Activity, mCi/km² (corrected for decay to sampling date)				
	^{144}Ce	^{125}Sb	^{106}Ru	^{137}Cs	^{54}Mn
Sep. 1963	95	19	34	39	11
Oct. 1964	113	18	42	50	15
Apr. 1965	72	19	30	51	10
Oct. 1965	63	16	22	54	7

Table 3. *Deposition of gamma-emitting radionuclides, May–Aug. 1966*

Nuclides	Average deposition, mCi/km² (corrected for decay to mid month)			
	May	June	July	Aug.
^{144}Ce, ^{125}Sb, ^{106}Ru ^{137}Cs, ^{54}Mn	0.6	0.6	0.5	0.4
^{141}Ce, ^{103}Ru, ^{95}Zr	~0.5	1.6	0.6	0.3
^{7}Be	1.2	1.9	1.4	1.4

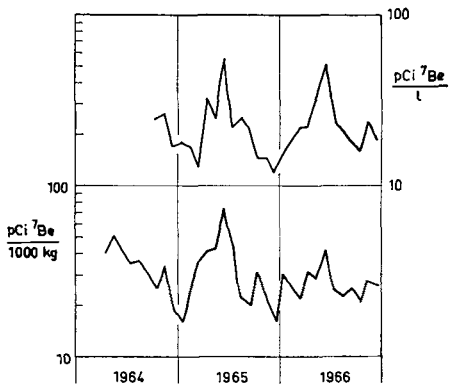

Fig. 5. Concentrations of beryllium-7 in ground level air and precipitation.

selected, and their body content of cesium-137 have been followed in detail.

The results of the body counting are shown in Fig. 6. In the figure is also inserted data from the Atomic Energy Company control group; before 1964 measured at HUGO, from 1964 at HUGO II. The decrease of average body burden 1964–1967 has been 50 per cent every 1.5 year.

Relations between deposition, diet and body content of cesium-137 have been calculated by Lindell & Magi (1965). They found the following formula applicable for the Stockholm region:

$$Q(i) = 0.80\,[F(i) + F(i-1) + F(i-2)],$$

where $Q(i)$ is the average body burden in nCi for the year i and $F(i)$ the total annual deposition in mCi/km² for the year i. An introduction of the deposition data of this report into the formula gives good agreement up to 1966.

If one assumes that the total average supply of cesium-137 to the body is proportional to the contribution from milk as measured by the National Institute of Radiation Protection (Snihs *et al.*, 1967), it turns out that an average biological half-life of 90 days corresponds to the observed effective decrease of body burdens 1965–1967. A curve representing a half-life of 90 days (0.25 year) starting mid 1967 is shown in Fig. 6 together with an extrapolation of the experimentally found 50 per cent decrease every 1.5 year for the male control group. The future "normal" population levels would lie somewhere between which is also suggested by the Lindell-Magi relation. The mean genetic cesium-137 dose for the control group was at maximum year 1964 less than 2 mrad/year, which can be compared to the average background dose 125 mrad/year. It can also be compared to the average yearly body potassium-40 dose, 17 mrads.

External gamma-doses from deposited fallout can of course also be estimated. A detailed description of the procedure is given by Beck (1966). Introducing the data of this report in his calculations gives a total open field fallout dose rate of about 0.7 μR/hr at the end of 1964. The

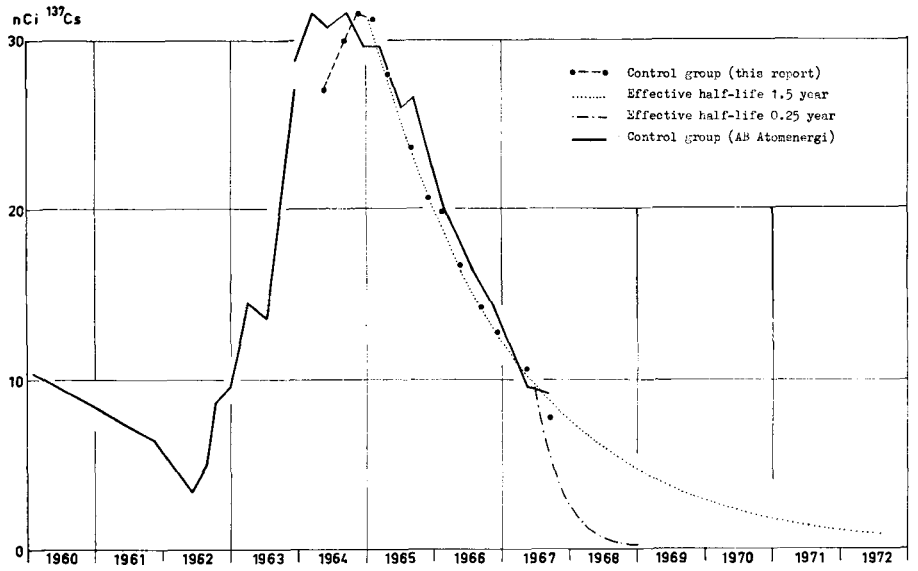

Fig. 6. Whole-body measurements of cesium-137.

total open field gamma dose can be estimated to 6 mrad for 1965 and somewhat less for 1966. It can be noted that during June 1966 dose rates from fresh fallout from the third Chinese nuclear test dominated over dose rates from additional deposited long-lived fallout by a factor of 3, maximum contribution coming from zirconium-95. This has however no influence on average yearly dose-estimations.

The above calculations can be compared with data from the National Institute of Radiation Protection which reports direct measurements of gamma-radiation from the ground (Snihs *et al.* 1967). The average natural background dose rate is measured to about 10 μR/hr and the additional average exposure from fallout is calculated to 5 mR during 1966.

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УМЕНЬШЕНИЕ СОДЕРЖАНИЯ РАДИОАКТИВНЫХ ЯДЕР В ВОЗДУХЕ, ОСАДКОВ И В ЧЕЛОВЕЧЕСКОМ ТЕЛЕ К ВЕСНЕ 1967 г.

Обсуждаются результаты измерений концентрации некоторых радиоактивных ядер (особенно цезия-137) в стратосфере, на уровне земли, в осадках и в теле человека. В течение 1967 г., четыре года спустя после прекращения больших атмосферных испытаний в конце 1962 г., приток стратосферного цезия-137 к поверхности земли балансировался распадом радиоактивности, уже содержащейся на поверхности. Среднее содержание цезия-137 в человеческом теле после максимума в 1964 г. уменьшается вдвое каждые 1,5 года. Вычислены внутренние и внешние дозы гамма-облучения. Обсуждаются также величины измеренных отношений между концентрациями некоторых других долгоживущих радиоактивных ядер и цезия-137. Сообщаются некоторые величины измеренных концентраций бериллия-7.