ASSESSMENT OF THE RADIATION DOSE DUE TO FALLOUT*

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The testing of nuclear weapons during the past decade has resulted in environmental radioactive contamination of world-wide proportions. The degree of contamination varies with the geographical location, depending primarily upon its position relative to the testing sites, amount of rainfall, and the latitude (1). The variation of fallout deposition with latitude is illustrated in Figure 1. This distribution pattern holds true for fallout coming from the stratosphere, which is the primary source of world-wide deposition.

Of the many radioelements produced in the fission process, Sr^{90} and Cs^{137} have been examined most thoroughly because of their high fission yield, long radioactive half lives, and chemical properties which readily permit entry into the biosphere, including man himself. Many factors relevant to the processes whereby these radioisotopes enter the food chain and are deposited in the human body have been qualitatively assessed (2,3). The body burdens of Sr^{90} and Cs^{137} have been measured as a function of time, and thus the radiation doses arising from such internal deposition may also be determined (4,5). A second source of radiation also exists, namely that which is due to gamma-emitting fission products external to man, yet capable of delivering to man, a dose often manyfold greater than that due to Sr^{90} and Cs^{137} combined. This presentation will be concerned with this external dose, its measurement, variation with time, and how it compares with the dose from natural background radiation.

Measurements of gamma-ray emitting fission products have been made periodically on soils collected at the Argonne National Laboratory site since the spring of 1957. The measuring technique employed was that of gamma-ray spectrometry of soil samples of finite size (1-2 kg) using a 5" by 4" NaI crystal and an Argonne Type 256 channel analyzer. The soil was geometrically arranged by means of a stainless steel container into a one inch layer around and over the crystal (6). Background reduction, essential for the detection of the quantities of activity involved (approximately 10^{-10} curie of Cs^{137}/kg of soil), was effected through the use of thick steel shielding around the sample and detector. Soil sampling was done to a depth of 6 inches so that essentially all fission products present over a given area were included. Two gamma-ray spectra of soil taken with an NaI crystal are shown in Figure 2. The one from deep soil is free of any fission activity and contains only Th, U, and K^{40}. The other, from soil collected in July 1959, indicates the presence of two peaks not evident in the first case. The one peak is due to Zr^{95}-Nb^{95} and the other to $Ru^{103,106}$.

The concentration of fission and natural radioactivity in soil was determined by the use of sources of known activity incorporated in mock soil (Na_3PO_4). The method presupposes knowledge of the various isotopes present in the soil. The standard reference spectra, one for each isotope, permit setting up the fraction of the total counting rate occurring in each spectral region, there being as many regions

as there are isotopes. Thus the solution of the appropriate number of simultaneous equations yields the concentration of each radionuclide in the soil being analyzed. The errors involved in such a procedure were estimated by preparing and counting a mock sample consisting of known amounts of the various isotopes, and then solving the pertinent equations. Comparison of the analytical results with the amounts known to be present provides a measure of the errors involved in such a method. The error in determining Cs^{137}, for example, was found to be on the order of 15%, with similar errors for the other fission products.

Table I indicates the activity in mc/mi^2 for Zr^{95}-Nb^{95}, Cs^{137}, Ru^{103}, Ru^{106}, Ce^{141}, and Ce^{144} at various times since May 1957. These data indicate the continuing accumulation of Cs^{137}, doubling between October 1957 and September 1958 and again almost doubling by the spring of 1959. Because of its 27 year half life, Cs^{137} is a reliable indicator of the total fallout accumulated at any given locality. No figures appear for Sr^{90} since it is not a gamma-emitting isotope. However, work done in colaboration with the Health and Safety Laboratory of the AEC reveals that there is a fairly constant ratio of 1.6 ± 0.2 between total Cs^{137} and total Sr^{90} activity in soil (7). Therefore one would expect the Sr^{90} activity to be roughly 0.6 that of Cs^{137}.

The concentration of fission activity is thus measureable, and the distribution in depth within the soil may also be determined empirically. The dose arising from these various fission products may be calculated under the assumption that the activity is spread

uniformily on an infinite plane according to the procedure of G. M. Dunning (8). Absorption and build-up factors have been included for each isotope, and the resulting dose rates are shown in Table II. It should be emphasized that these dose rates in μrad/hour are for an unshielded point one meter above the ground.

The doses thus calculated were compared with the dose measured with an appropriate ionization chamber at various times during 1959. In order to compare calculated and empirical dose rates one must bear in mind that the measured dose conteins a component due to natural radioactivity and to cosmic radiation. The dose from U, Th, and K^{40} may be calculated from Hultqvist's equations using the concentrations of these radioelements determined from gamma-ray spectrometry (9). The dose from cosmic radiation was taken from the literature and expressed in μrad/hour. Table III shows the calculated dose for April 1959, totaling 19.2 μrad/hour, contrasted with a measured dose rate of 18.5 μrad/hour. Agreement of a similar order was observed during the succeeding spring and summer months, lending support to both the method of calculation and to the method of measuring fission product concentration.

As previously stated, these dose rates pertain to the unshielded situation. The actual dose to which an individual is exposed must take into account shielding, time spent out-of-doors, and the like. Again use was made of an ionization chamber to determine the dose rate indoors and outside. Within the accuracy of measurement, a shielding factor of 5 was found for fallout radiation (i.e. the dose due to

fallout prevailing indoors was 1/5 that found outside). Such a shielding factor is in agreement with that proposed by the UN as an average for Western type structures (10). An additional factor has been suggested by this body to account for weathering which reduces the dose because of leaching, washoff, and other mechanisms which reduce the activity present. Over the period 1957 to 1959 no direct evidence has been found at this laboratory for a downward movement of Cs^{137} in soil, nor has it been found for any of the other isotopes mentioned. Therefore, for this period, no factor has been included for weathering.

A unique situation exists at the present time for the study of fallout mechanisms. Testing of nuclear weapons has stopped, and there is no further injection of fission debris into the stratospheric reservoir, so that it should now be possible to obtain values for fallout rate and stratospheric storage time. Furthermore, since no new activity is being produced, there is some justification for hoping that future levels of radioactivity and radiation may be predicted within reasonable limits. To this end monthly soil samples have been collected at Argonne since March 1959, and from these samples fallout rates for each radioisotope mentioned have been determined. Interpretation is complicated by the fact that a portion of the fallout at Argonne comes from tests conducted by the US and UK in the Pacific area and a portion from Russian tests carried out in the far North. The stratospheric storage times and consequently the fallout rates appear to be substantially different for debris coming initially from these two locations (11,12).

The best fit to our data is obtained upon assuming that 60-75% of the fallout is due to Russian tests having a storage time of 6 to 9 months with the remainder coming from Pacific tests with a storage time of 4 years. This proportionation appeared to continue throughout the summer of 1959. On this basis it is possible to compute the initial amount of any isotope in the reservoir at the time of detonation. For example, the Cs^{137} content of the reservoir according to these data was 6.0 megacuries in October 1958. For this same date, air and rain sampling indicated 4 megacuries of Sr^{90} in the reservoir (13). Using the Cs/Sr ratio of 1.6, one obtains 6.4 megacuries of Cs^{137}. The initial reservoir content of the other fission products measured are in agreement with that of Cs^{137} according to the fission yields of these isotopes.

Encouraged by the consistancy of these findings, estimates of the future levels of external radiation and integral dose from such radiation were made under the assumption that testing is not resumed. It is to be emphasized that these figures actually pertain only to the Argonne site and its immediate surroundings, although the similarity in Sr^{90} deposition at Argonne and at other localities throughout the North Central and Northeastern United States implies that comparable fallout levels exist over this area, which contains roughly one half the population of the country. Figure 3 indicates the variation of gamma dose rate from all fission products, as well as that due to Cs^{137} alone, starting in 1957. Extrapolation to future levels is done

on the basis of no further testing. The maximum dose rate from all fission material occurred in May 1959, amounting to 8.46 μrad/hour unshielded, or 1.69 μrad/hour for indoor exposure, contrasting with 11.12 μrad/hour due to natural background. Concurrently the internal whole body dose from Cs^{137} was 0.2 μrad/hour and that to bone from Sr^{90} was 0.3 μrad/hour. The total internal dose being on the order of one third that arising from external fission products. As expected, after a few years the bulk of the external dose is due to Cs^{137}.

The integral 30-year and 70-year whole body dose to man from fallout radioactivity were calculated for the present situation in which there has been no further testing since the fall of 1958. These doses have been termed the genetically significant dose and the lifetime dose respectively. The whole body dose is composed of an external component, attenuated by the proper shielding factor, and an internal component arising from Cs^{137}. An additional source of radiation to bone exists due to the presence of Sr^{90}. The external dose was computed using two assumptions; (A) a shielding factor of 5 for all radionuclides, and (B) a shielding factor of 5 and a weathering factor of 2 for Cs^{137}. The internal dose from Cs^{137} also involved two assumptions; (C) the Cs^{137} body burden is proportional to fallout rate, and (D) the Cs^{137} body burden is proportional to the accumulated Cs^{137} deposition. Depending upon the assumptions considered, the 30 year dose lies between 42 and 113 mrads and the 70 year dose between 48 and 170 mrads. Investigations at Argonne have shown little weathering

effect up to mid-1959, tending to support assumption (A). Studies of the concentration of Cs^{137} in milk and other foodstuffs have shown primarily a dependence upon fallout rate, which in turn implies that the body burden will be rate dependent according to assumption (C) (2). The most probable value for the 30-year dose (assumptions (A) plus (C)) is 52 mrads and 62 mrads for the 70-year dose. These compare with 30 and 70 year doses of 3.75 and 8.75 rads respectively due to natural radiation which is assumed to amount to 125 mrads/year in the Chicago area. Thus fallout radioactivity results in a dose increment of 1.4 and 0.7 percent for the 30 and 70 year doses respectively over and above that due to natural sources. A summary of the values obtained using the various assumptions along with those due to natural sources is given in Table IV. Estimates of the dose to bone from Sr^{90} for these time intervals is also included in this table (14).

Consideration has also been given to the whole body fallout dose in the case where testing continues for 5 years beyond 1958 at the 1954-58 testing rate and for the case in which testing continues indefinitely at this rate. The genetic and lifetime doses under these circumstances are presented in Table V with the dose from natural sources shown for comparison. Continuous testing will result in an increase in whole body irradiation amount to some 6% over that coming from natural background.

It will be noted that no evaluation has been made of the dose due to I^{131}, C^{14}, or other radionuclides known to be present *in vivo* even very briefly as occurs in the case of non-assimilated elements passing through the GI tract. As far as whole body irradiation is concerned these radioisotopes are of secondary importance compared with ground fallout and internal Cs^{137} and Sr^{90}.

The methodology described and utilized in this work has made it possible to obtain a measure of the dose arising from gamma-emitting fission activity on the ground. Evaluation of the whole body dose to man from such sources has been made through the use of shielding factors applied to this gamma radiation. The present situation in which nuclear weapons testing is no longer being carried out will lead to a genetic dose of 52 mrads and a lifetime dose of 62 mrads to roughly one-half of the population of the United States, an increase of approximately 1% over that arising from natural radiation. The continuous testing of nuclear weapons at the rate occuring between 1954 and 1959 would result in an increase of the whole body dose of some 6% over that occurring naturally.

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TABLE I

Gamma-Ray Emitting Fallout Radioactivity on the Ground at Argonne National Laboratory in mc/mi^2

Isotope	1957 May	1957 July*	1957 October	1958 April	1958 July	1958 Sept.	1959 March	1959 April	1959 May	1959 June	1959 July	1959 August
Zr^{95}-Nb^{95}**	180	195	230	200	95	315	731	980	1043	917	756	
Cs^{137}	35	34	37	45	60	82	128	135	141	147	152	
Ru^{106}	175	180	180	170	150	210	624	812	881	1173	1202	
Ru^{103}	175	200	240	130	35	80	331	230	186	120	88	
Ce^{141}	110	120	140	60	15	50	580	329	268	182	113	
Ce^{144}	240	260	275	250	215	270	1127	1450	1533	1704	1812	
Totals***	1095	1184	1332	1055	665	1322	4252	4916	5095	5160	4879	

* Soil collected 40 miles from the Argonne site gave essentially the same results as the ANL soils, indicating that any contamination from the reactors at Argonne is of secondary importance in this study.

** Assuming Zr^{95}-Nb^{95} to be in equilibrium.

*** Totals include activity due to both Zr^{95} and Nb^{95}, i.e. May 1957 360 mc/mi^2 from these two isotopes.

TABLE II

Dose Rate due to Fallout at Argonne National Laboratory in μrad/hour

	1957			1958			1959					
Isotope	May	July	October	April	July	Sept.	March	April	May	June	July	August
Zr^{95}-Nb^{95}	1.14	1.23	1.46	1.27	0.60	2.00	4.64	6.22	6.62	5.82	4.80	
Cs^{137}	0.09	0.08	0.09	0.12	0.15	0.21	0.32	0.34	0.36	0.37	0.38	
Ru^{106}	0.15	0.15	0.15	0.14	0.13	0.18	0.53	0.69	0.75	1.00	1.03	
Ru^{103}	0.35	0.41	0.49	0.27	0.07	0.17	0.69	0.48	0.39	0.25	0.18	
Ce^{141}	0.01	0.02	0.02	0.01	0.01	0.01	0.13	0.07	0.06	0.04	0.03	
Ce^{144}	0.04	0.05	0.05	0.04	0.04	0.05	0.21	0.27	0.28	0.31	0.33	
Total	1.78	1.94	2.26	1.85	1.00	2.62	6.52	8.07	8.46	7.79	6.75	

Natural Background = 11.12 μrad/hour

TABLE III

Calculated and Measured Gamma-Ray Dose Rate One Meter above Ground at Argonne National Laboratory April 1959

	Calculated		Measured	
Thorium	2.53	µrad/hour		
Uranium	2.65			
K^{40}	2.77			
Cosmic Radiation	3.17			
Fallout	8.07			
Total	19.19	µrad/hour	18.48	µrad/hour

TABLE IV

Integral Dose for the Case in which Testing Stopped in Fall 1958
in mrad

	Whole Body						Bone
	External		Internal-Cs^{137}		Total	Background	Bone-Sr^{90}
Time Interval	(A)*	(B)	(C)	(D)			
30 years	42	32	10	71	42-113	3750	40-160
			Most probable		52 (1.4%)**		
70 years	52	40	10	118	48-170	8750	93-372
			Most probable		62 (0.7%)		

* Assumption (A) Shielding factor of 5 for all fission products

" (B) Shielding factor of 5 and weather factor of 2 for Cs^{137}

" (C) Cs^{137} body burden proportional to fallout rate

" (D) Cs^{137} body burden proportional to accumulated fallout.

** Percentage of background dose.

TABLE V

Probable Values of Whole Body Dose in mrads

Conditions	30-year Dose	70-year Dose
Tests continue for 5 years beyond 1959	79 (2.1%)*	95 (1.1%)
Tests continue indefinitely	234 (6.2%)	546 (6.2%)
Natural Background	3750	8750

* Percentage of Background Dose.

FIGURE LEGENDS

Figure 1. Latitude distribution of stratospheric fallout.

Figure 2. Gamma-ray spectra of soil.

Figure 3. Gamma-ray dose from fission products tests stopped in 1958.

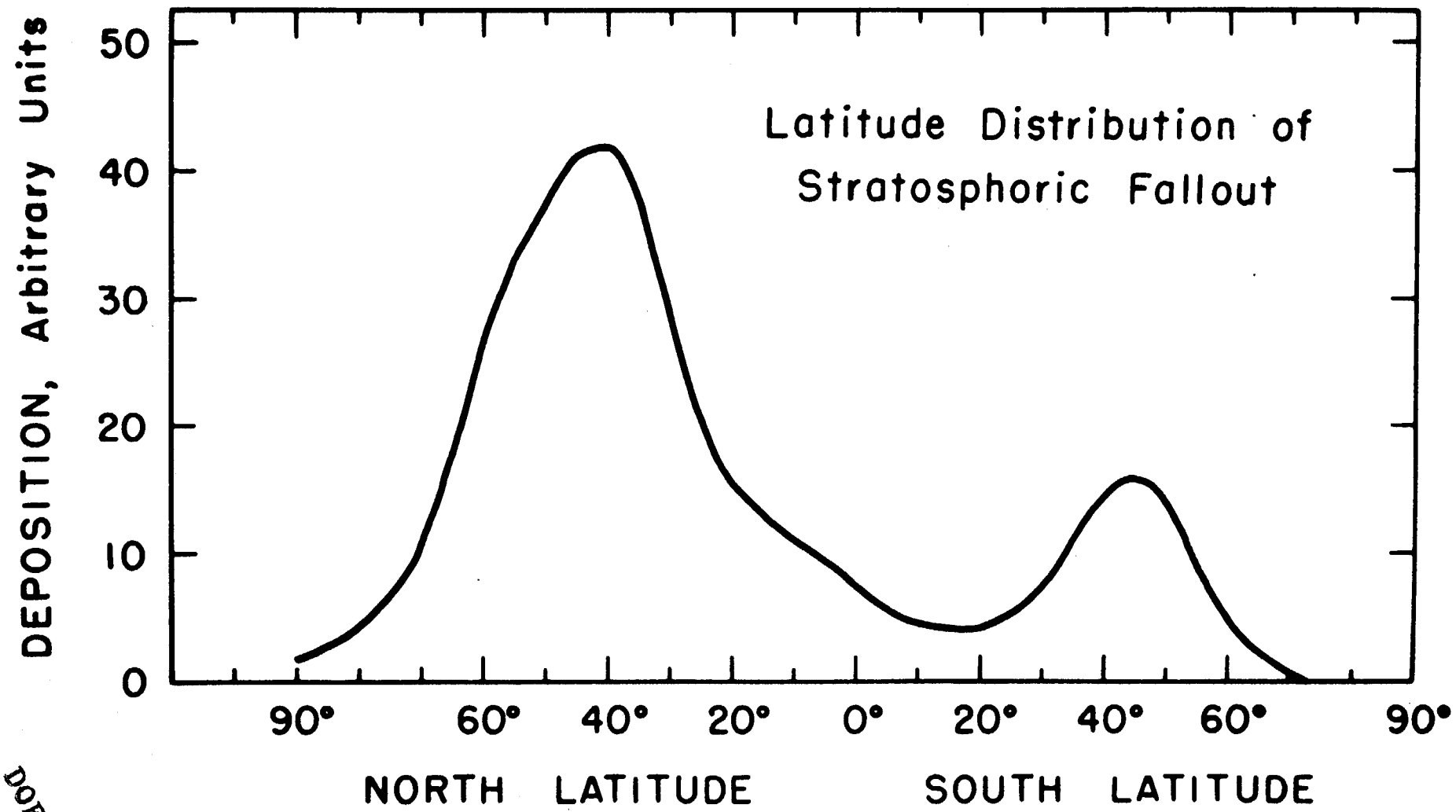

Fig. 1

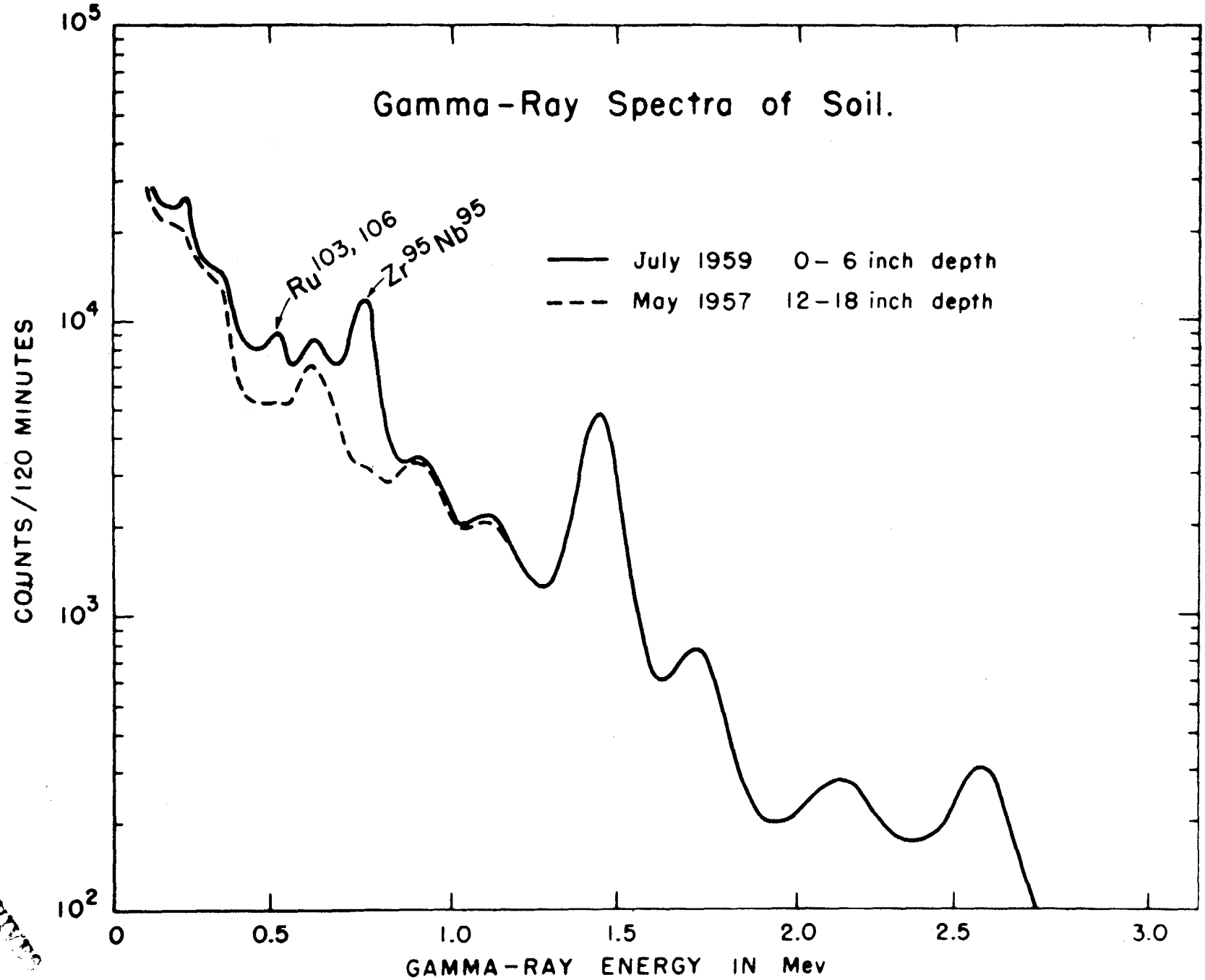

Fig. 2

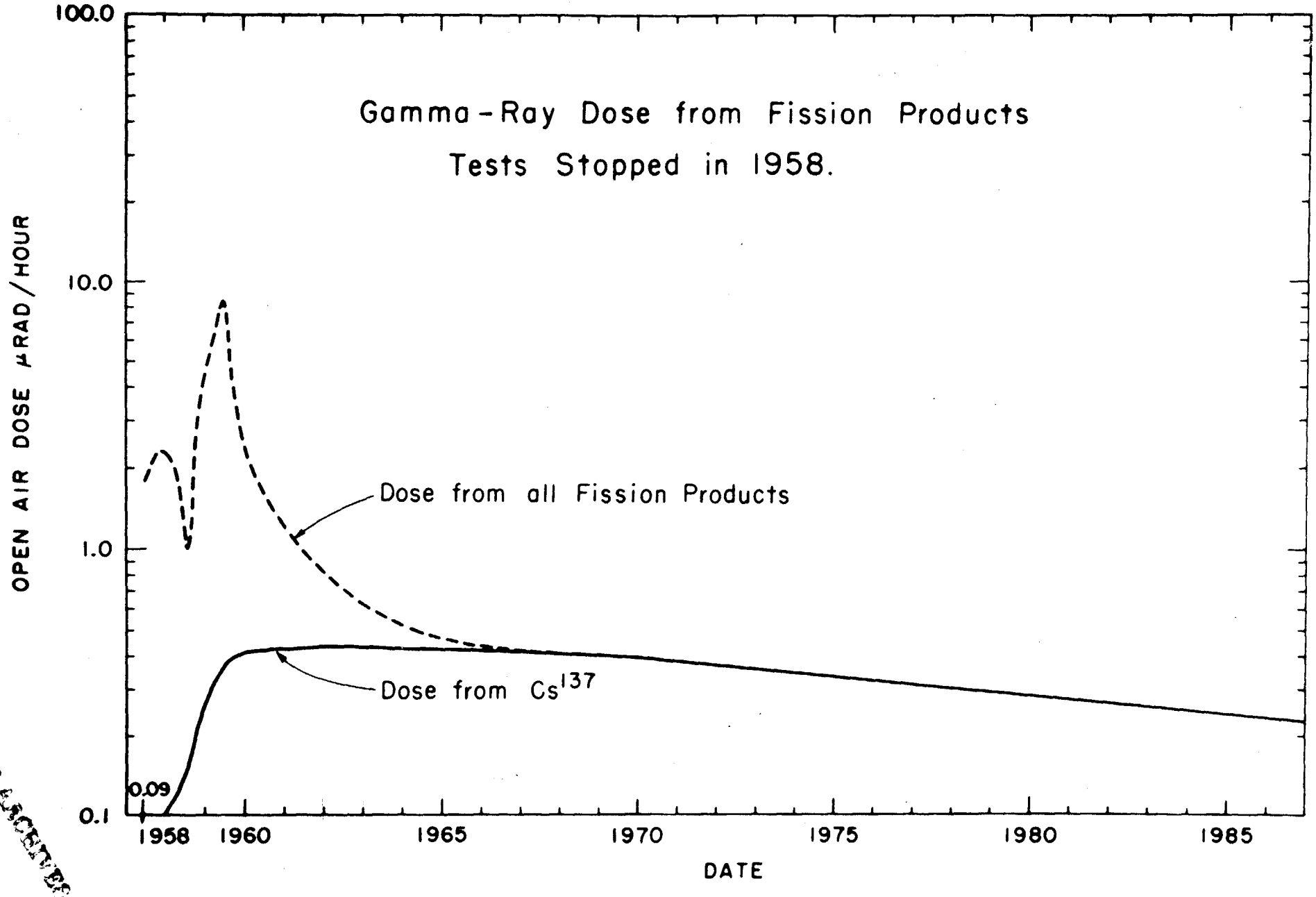
Gamma-Ray Dose from Fission Products
Tests Stopped in 1958.
OPEN AIR DOSE μRAD/HOUR
100.0
10.0
1.0
0.1
0.09
Dose from all Fission Products
Dose from Cs137
1958
1960
1965
1970
1975
1980
1985
DATE

Fig. 3