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Radioactive Fallout through September 1955

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Soon after the fallout-monitoring network of the U.S. Atomic Energy Commission was established in the United States, it became apparent that daily fallout observations could be made by simple procedures at great distances from a nuclear detonation, and the network was expanded in October 1952 to include a number of locations beyond the continental United States. Additional stations were again added in February 1954, and since then the sampling program has been conducted at 88 stations, including 26 in the United States. These operate continuously. Additional stations in the United States are added when nuclear detonations are being conducted in Nevada. The overseas stations are listed in Table 1, the continental stations in Table 2.

The data collected within continental United States through August 1954 have been reported in previous publications (*1, 2*). In this report (*3*) are summarized the data obtained in the United States and abroad through September 1955.

The principal objective of the monitoring program is to enable one to estimate the levels of human exposure produced by radioactive fallout at great distances from nuclear detonations. Such exposure may result from external irradiation by radioactive dust deposited on the surface of the earth or from internal irradiation by specific radionuclides that enter the body by ingestion.

With respect to ingestion, strontium-90 is the nuclide selected for continuing attention because, from the point of view of the long-range behavior of the isotopes involved in biological systems, it is potentially the most hazardous. This is due in part to its relatively long half-life (about 28 years) but, more particularly, because of the chemical similarity of strontium to calcium and the possibility that strontium-90 can therefore be assimilated into biological processes involving calcium and ultimately be deposited in human bone.

The data from the monitoring network provide the following estimates for each place where a station is located. (i) The cumulative surface deposits of mixed fission products and strontium-90. These estimates are reported in millicuries per square mile. (ii) The cumulative gamma dose (in millirads) from external radiation.

It will be noted that the earlier publications did not include estimates of the gamma dose. However, in recognition of the increased interest in such estimates, these data have been computed for the full sampling period and will hereafter be available on a continuing basis.

Method of Sampling

Adhesive-coated films, as described previously, are used for collecting samples. An adhesive coated acetate film is supported horizontally on a frame about 3 feet above the ground. The coating retains its adhesive properties when it is wet, and the radioactive dust particles that are entrapped in raindrops are collected, possibly by impaction against the adhesive surface. Most stations maintain duplicate sampling units, thus providing some assurance against loss of samples. The films are changed each day at the same time and are mailed to the U.S. Atomic Energy Commission Health and Safety Laboratory in New York, where their radioactivity is assayed.

Studies have continued of the collection characteristics of gummed film in comparison with those of high-walled pots. Analyses of the data have been completed for an 84-week test period, indicating that the gummed film is 63 percent efficient if we make the assumption that the high-walled pots collect total fallout. We consider that the pots provide the best practical estimate of total fallout, and the data obtained from the use of gummed films have therefore been corrected by a factor of 1.6.

Methods of Estimation

Mixed fission product activity. When the sample is received in the laboratory, it is ashed at 550° to 600°C, and its beta activity is counted. The potassium-40 activity of a known weight of potassium carbonate is used as the basis for converting counts per minute to millicuries. This activity is then calculated as of (i) the sampling day and (ii) an arbitrary future date, usually the first day of the following calendar year. These extrapolations depend on knowledge of the age of the sample, its decay being assumed to be proportional to $t^{-1.2}$. At any given time, the sum of the extrapolated daily values provides an estimate of the cumulative radioactivity deposited at the sampling station as of the arbitrary date.

Strontium-90. The strontium-90 content of the samples can be estimated from Hunter and Ballou curves of relative isotopic abundance (*4*), or they can be measured directly by radiochemistry. When the latter procedure is used, it is desirable to collect the samples for a period of at least 1 month to facilitate the counting procedures, and a suitable vessel such as a high-walled pot can be exposed for this purpose.

The Hunter and Ballou curves were used during this reporting period.

Gamma dose. It is not possible to measure directly the cumulative gamma dose from fallout at a distance from a nuclear detonation. The normal gamma-radiation background of cosmic and terrestrial origin is much larger in magnitude than the gamma radiation from fallout, and usually masks the latter completely. It is

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thus necessary to estimate the cumulative gamma dose by indirect means.

In calculating the dose, it has been assumed that the daily fallout is deposited uniformly on an infinite smooth plane, where it remains to infinite time. The integrated infinite dose for each daily fallout is calculated from the measured beta activity, using the known isotopic composition of the sample and the known gamma characteristics. The sum of these integrated infinite doses from each of the daily samples collected during any given time represents the estimated gamma dose for infinite time delivered to populations who are exposed from the start of the sampling period. For practical estimates, the infinite dose is delivered in 2 to 4 years after fallout.

The integrated gamma dose is sensitive to the time of fallout, particularly in those parts of the world in which fallout may occur within 1 or 2 days of the detonation. In such cases, the bulk of the gamma dose is delivered within 1 month or so after fallout. It is thus desirable to know the time of fallout, at least to the nearest day, and for this reason daily samples are collected.

The United States collections were begun on a systematic basis in the fall of 1951. However, because of the way in which the data were organized prior to the fall of 1952, it has not proved practical to calculate the gamma dose estimates prior to this date. An exception has been made in Salt Lake City, Utah, which is the only location beyond the immediate vicinity of the Nevada test site known to have received a gamma dose during this period that is significant in relation to the total received since October 1952.

Table 1. Fallout at stations outside continental United States, October 1952 to September 1955. Stations that have not sampled continuously since October 1952 are indicated by an asterisk.

Station	Mixed fission products (mc/mi²)	Strontium-90 (mc/mi²)	Gamma dose (mrad)
1. Anchorage, Alaska	62	2.7	6.5
2. Edmonton, Alberta	78	2.8	7.6
3. Regina, Saskatchewan	82	3.0	9.3
4. Winnipeg, Manitoba	95	3.6	14
5. Churchill, Manitoba	50	1.9	4.1
6. Moosoonee, Ontario	67	2.8	12
7. North Bay, Ontario	100	3.1	12
8. Ottawa, Ontario	110	3.4	12
9. Montreal, Quebec	110	4.0	13
10. Seven Islands, Quebec	84	3.3	12
11. Moncton, New Brunswick	83	3.7	10
12. Goose Bay, Labrador*	160	4.0	13
13. Stephenville, Newfoundland	120	4.3	18
14. Thule, Greenland	50	2.0	3.6
15. Keflavik, Iceland	92	2.9	8.1
16. San Juan, Puerto Rico*	100	3.9	14
17. Bermuda	96	4.6	19
18. Mexico City, Mexico	110	5.1	22
19. San Jose, Costa Rica	63	3.2	11
20. Panama Canal Zone	89	4.1	15
21. Bogota, Colombia	50	2.6	10
22. Quito, Ecuador	51	2.6	10
23. Lima, Peru	50	1.8	5.0
24. La Paz, Bolivia	92	4.2	14
25. Belem, Brazil	74	3.4	12
26. São Paulo, Brazil	53	2.7	9.7
27. Buenos Aires, Argentina	60	2.8	9.4
28. Prestwick, Scotland	87	3.8	7.9
29. Oslo, Norway*	56	2.5	6.2
30. Rhein Main, Germany	100	3.5	9.0
31. Sidi Slimane, Morocco*	64	2.5	6.6
32. Tripoli, Libya	83	4.0	12
33. Dakar, French West Africa	74	3.6	8.0
34. Lagos, Nigeria*	33	1.9	4.6
35. Leopoldville, Belgian Congo*	70	3.4	8.0
36. Addis Ababa, Ethiopia*	110	4.2	10
37. Pretoria, Union of South Africa	39	2.0	4.2
38. Durban, Union of South Africa*	34	1.9	4.9
39. Colombo, Ceylon*	91	4.7	23
40. Singapore, Malaya*	95	4.6	18
41. Misawa, Japan*	73	2.8	9.6
42. Tokyo, Japan*	100	3.8	13
43. Hiroshima, Japan	66	3.2	11
44. Nagasaki, Japan*	92	4.9	16
45. Kadena, Okinawa*	76	4.0	14
46. Taipei, Taiwan*	100	4.6	16
47. Manila, Philippine Islands*	110	6.6	29
48. Iwo Jima*	290	24	150
49. Yap, Caroline Islands*	170	9.0	40
50. Guam, Caroline Islands	160	8.5	40
51. Truk, Caroline Islands*	190	9.2	47
52. Ponape, Caroline Islands*	240	14	63
53. Wake Island*	89	3.6	13
54. Noumea, New Caledonia*	60	3.2	12
55. Sydney, Australia*	65	3.5	12
56. Melbourne, Australia*	47	2.1	6.6
57. Wellington, New Zealand*	40	2.1	6.4
58. Honolulu, Hawaii	83	3.5	15
59. Johnston Island*	130	5.9	28
60. Canton Island*	86	4.2	19
61. Dhahran, Saudi Arabia*	57	3.1	7.9
62. Beirut, Lebanon*	73	3.3	8.4

Findings

The cumulative mixed fission product data as of 1 January 1956 are summarized on the world-wide map in Fig. 1, and strontium-90 and gamma dose data are given in Table 1. The United States data are listed individually in Table 2.

The estimates of mixed fission products and strontium-90 in the United States include the period from October 1951 to September 1955. Although some of the foreign stations began collections in October 1952, not all of them have sampled continuously since that time; these are indicated by an asterisk. Full data are available from the Pacific and Asian locations for the 1952 and 1954 Pacific tests, but they are not always available during 1953, when a Nevada test series was conducted. Some of the European, African, and South American stations did not collect samples during the Pacific test series in 1952. The missing data at these foreign stations have been estimated by regional interpolation. All stations have been sampling continuously since 1 January 1954, and it is since that time that the bulk of the fallout to date has occurred at most overseas stations.

The gamma doses, which are derived by calculation from the fallout of mixed fission products, are not available prior

to October 1952 for any United States stations except Salt Lake City. However, the values reported may be taken as a satisfactory approximation of the total gamma dose delivered to date. The fallout prior to October 1952 was of a low order compared with that during the subsequent period.

As expected, the high accumulations of mixed fission products (Fig. 1) exist in the vicinity of the proving grounds in Nevada and in the vicinity of the Pacific test area. The highest recorded deposition, Grand Junction, Colo., is 740 millicuries per square mile as of 1 January 1956. The values are lower everywhere else, the next highest deposition being 290 millicuries per square mile at Iwo Jima in the Western Pacific Ocean. The lowest value is Lagos, Nigeria, with 33 millicuries per square mile. In general, the mixed fission products data are characterized by a high degree of regional uniformity in which, with few exceptions, adjacent stations agree within ± 50 percent.

The estimates of strontium-90 are shown in Tables 1 and 2. Outside of the United States, the observed range is from 1.8 millicuries per square mile in Peru to 24 millicuries per square mile at Iwo Jima. In the United States, the accumulation varies from 2.1 at San Francisco to 23 at Salt Lake City. For most of the world covered by this network, the range of 2 to 5 millicuries per square mile may be taken as a representative estimate of the distribution of strontium-90.

With respect to the gamma dose, the average value for the United States is higher than it is for the rest of the world. The range of values in the United States is relatively narrow, 6 to 49 millirads, except for Salt Lake City (160), Grand Junction (120), and Albuquerque, N.M. (110). The representative dose for eastern United States is about 15 to 20 millirads, with slightly higher values in the Middle West and lower values on the West Coast.

The cumulative gamma dose at the foreign stations is in the range of 4 to 23 millirads, except for some of the Pacific islands, where the range is from 13 to 150 millirads.

These gamma values are somewhat lower than the average estimate of 100 milliroentgens for the United States as reported by Dunning (*5*) whose calculation was intended to provide a tentative upper limit of the estimated dose and was deliberately conservative.

Table 2. Fallout at stations within continental United States, October 1951 to September 1955.

Station	Mixed fission products (mc/mi²)	Sr^{90} (mc/mi²)	Gamma dose* (mrad)
Albuquerque, N.M.	400	20	110
Atlanta, Ga.	120	3.8	16
Billings, Mont.	160	5.7	24
Binghamton, N.Y.	61	2.2	7.8
Boise, Idaho	160	9.2	16
Chicago, Ill.	140	5.3	24
Dallas, Tex.	170	6.1	29
Des Moines, Iowa	170	6.2	28
Detroit, Mich.	140	4.2	21
Grand Junction, Colo.	740	18	120
Jacksonville, Fla.	88	3.3	13
Memphis, Tenn.	200	8.4	49
Minneapolis, Minn.	130	4.9	18
New Haven, Conn.	110	3.6	17
New Orleans, La.	170	5.7	27
New York, N.Y.	110	4.2	17
Philadelphia, Pa.	110	4.6	16
Pittsburgh, Pa.	100	4.1	13
Rapid City, S.D.	150	6.1	25
Rochester, N.Y.	99	3.7	13
St. Louis, Mo.	200	6.0	27
Salt Lake City, Utah	680	23	160
San Francisco, Calif.	47	2.1	5.8
Seattle, Wash.	89	3.5	11
Scottsbluff, Neb.	200	6.3	26
Washington, D.C.	86	3.0	11

* Gamma dose for the period October 1952 to September 1955, except for Salt Lake City, which covers the period October 1951 to September 1955.

Discussion

The collection efficiency of the gummed film is of fundamental importance in interpreting reported values. As noted previously, the over-all efficiency for collection of total fallout activity is taken to be 63 percent, and all of the data have been corrected accordingly.

A source of error which is more difficult to assess is that inherent in the assumption that the decay of the radioactivity is proportional to $t^{-1.2}$. Application of this decay law requires that the age of the debris be known with some certainty, and this has become increasingly difficult during the past 2 years. Prior to 1954, it was possible to predict the decay characteristics of a sample accurately because at any given time the debris was known to have originated from the most recent test series. After each series, the daily fallout would diminish rapidly and would ordinarily be undetectable before the next series of tests started.

The rapid diminution in fallout from tests conducted prior to the spring of 1954 can be explained by the fact that, except for Operation Ivy in November 1952, the yields from detonations were relatively low, and the bulk of the debris was distributed below the tropopause, where fallout is greatly hastened by precipitation and other factors. In contrast, the detonation of devices having yields equivalent to megatons of TNT produces clouds of radioactive debris which pierce the tropopause and become distributed in the stratosphere. From this relatively stable region of the earth's atmosphere, the particles descend slowly, and fallout to the earth's surface occurs over a period of time which is measured in years rather than weeks or months. The traces of relatively old debris from high-yield devices become mixed with the debris of subsequent detonations. This being the case, neither the decay characteristics of a sample nor the relative abundance of the long-lived isotopes can be predicted from theory.

In recent months, the procedure has been changed and, where necessary, either individual or pooled samples are followed for decay.

There is no practical way in which the accuracy of the estimates of accumulated mixed fission products can be tested experimentally. However, it is possible to test the validity of the strontium-90 estimates by direct radiochemical analyses of the soils from the vicinity of the sampling stations. In October 1955, immediately following the sampling period covered by this report, soil samples were collected from 17 widely scattered locations in the United States. The relationships between the predicted and measured values are given in Figure 2. It is seen that the data consist of two groupings, in which 14 points show a reasonably satisfactory regression of measured on predicted values. The correlation coefficient for these data is 0.70. The other three points for stations near the Nevada test site show low ratios

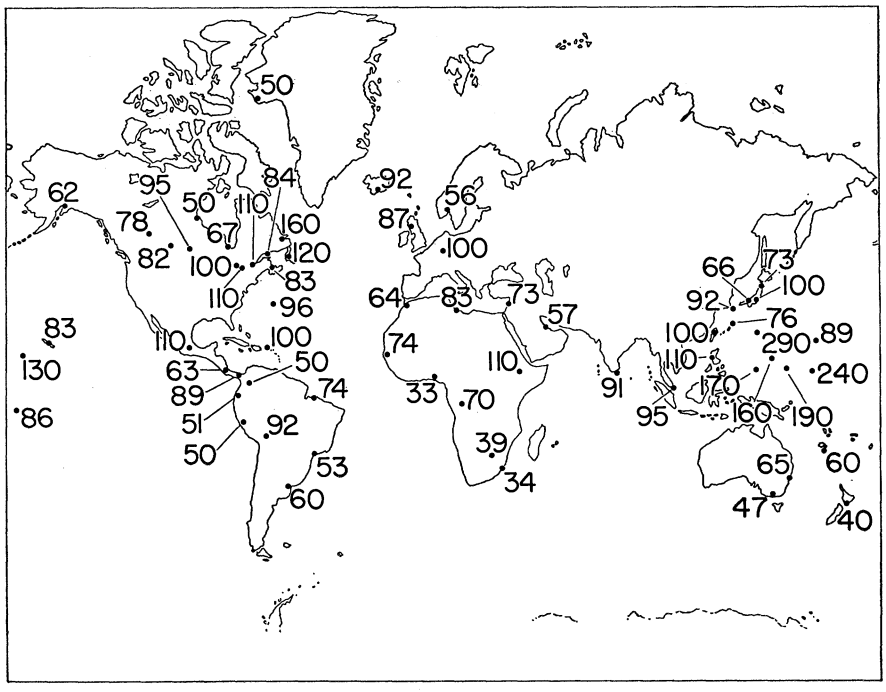

Fig. 1. Cumulative deposition of mixed fission products in millicuries per square mile at the stations shown. These data are for fallout from October 1952 to September 1955 and are extrapolated to 1 January 1956.

of measured to predicted strontium-90.

These findings are consistent with present knowledge of the way strontium-90 is formed and distributed. The estimates of strontium-90 were obtained by using the Hunter and Ballou curves of relative isotopic abundance in conjunction with the daily measurement of mixed fission products. Because strontium-90 is derived following fission from its precursor krypton-90, an inert gas which has a half-life of 33 seconds, some of the strontium-90 is formed relatively late in the life of the fireball. The relative abundance of strontium-90 in any given particle of dust is variable—there is a depletion of strontium-90 in debris which falls out relatively close to the site of detonation and a corresponding enrichment from debris which falls out at greater distances. This may explain the low ratios of measured-to-predicted values for Albuquerque, Salt Lake City, and Grand Junction, which are relatively close to the Nevada site. Similarly, fallout analyzed at great distances from detonations is known to be enriched in strontium-90 by as much as a factor of 2, which explains why the measured values at the other stations were higher than the predicted values. Based on this soil study, one might be justified in doubling the strontium-90 values given, but this factor was not used in preparing the tables.

The estimates of gamma dose, like the estimates of strontium-90 were derived by calculations from estimates of the fallout of mixed fission products. It should be noted that the assumptions underlying the calculation of gamma dose tend to produce values which are much higher than the doses to which populations are actually exposed. The reported values do not allow for weathering or shielding.

The effects of weathering and shielding cannot be treated quantitatively. For populations in cities, the true dose would be very much reduced by the fact that fallout to the surface is soon washed into gutters and storm sewers. For these reasons, it is likely that the actual dose to urban populations does not exceed 10 percent of the values reported here.

Rural populations are less shielded by buildings and do not have the advantage of large paved areas to encourage runoff during storms. Nevertheless, the true situation is never the infinite smooth plane on which these calculations are based, and some reduction is afforded by irregularities in the terrain, the plowing of fields, and other factors.

Significance of Findings

In interpreting the significance of these data, one needs to consider only the estimates of gamma dose and distribution of strontium-90. The total accumulations of mixed fission products are important only insofar as they serve as the basis for estimating the gamma dose delivered and the amount of strontium-90 present.

Geneticists are concerned with the average gamma dose to populations because this serves as the basis for estimating the number of radiation-induced chromosome mutations in the population as a whole. By referring to the tables, the average dose may be taken to be in the order of 10 millirads for the 4-year period covered by this report. This is small when compared with the gamma radiation received from natural sources by populations throughout the world. Libby (*6*) estimates that the average external dose from natural sources, both terrestrial and cosmic, is of the order of 75 millirads per year, or 300 millirads in four years. On this basis, the gamma dose delivered from fallout is about 3 percent of the average gamma dose from natural sources. Thus, even the maximum theoretical dose from fallout to date is a small fraction added to the gamma dose received from natural radioactivity, and this slight increment is considerably smaller in magnitude than the normal variations in natural dose which occur from place to place throughout the world.

The significance of the deposition of strontium-90 in the quantities shown in the tables can be understood in relationship to the occurrence of natural radium, with which strontium shares many chemical properties. In the upper 1 foot of the earth's crust, radium is present in amounts approximating 1000 millicuries per square mile. This radium, like other trace elements, is absorbed into all living things. Adult North Americans contain about

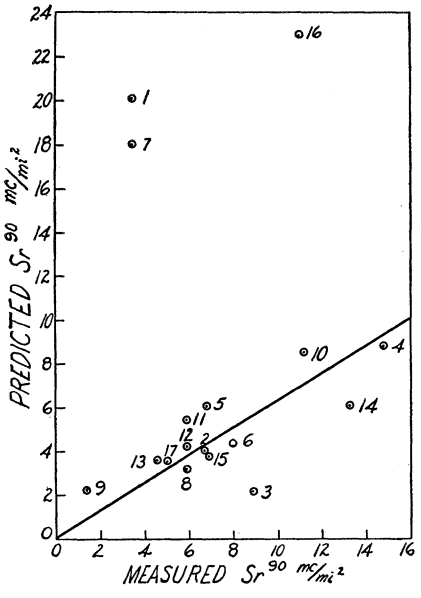

Fig. 2. Plot of measured versus predicted strontium-90 for soils from 17 locations in the United States: *1*, Albuquerque; *2*, Atlanta; *3*, Binghamton; *4*, Boise; *5*, Des Moines; *6*, Detroit; *7*, Grand Junction; *8*, Jacksonville; *9*, Los Angeles; *10*, Memphis; *11*, New Orleans; *12*, New York; *13*, Philadelphia; *14*, Rapid City; *15*, Rochester; *16*, Salt Lake City; *17*, Seattle.

10^{-4} microcuries of radium, which may be taken as the biological equilibrium of the radium burden of human beings in relation to the general environmental radium content of the upper 1 foot of soil. This amount (10^{-4} microcuries) is 1/1000 the maximum permissible radium burden of 0.1 microcurie and more nearly one 1/10000 the minimum amount that is known to have produced injury to human beings.

Natural radium and strontium-90 in fallout have differences in properties which may influence the ease with which they pass from soils into biological systems; therefore, one cannot conclude that, for any given soil content of strontium-90, the equilibrium human burden would be the same as the equilibrium body burden of radium at the same soil level. However, it is worth noting the minuteness of the present strontium-90 values in relation to the amount of radium present in all soils and, more particularly, in relation to the very much larger concentration of radium that could be safely tolerated.

A more direct method of evaluating the significance of the strontium-90 fallout is to measure the presence of this isotope, not only in soils, but in plants, animals, and human foodstuffs. Fortunately modern radiochemical techniques are sufficiently sensitive so that it is feasible to detect this isotope at concentrations comparable with that of radium and other naturally occurring isotopes. Measurements of this type have been undertaken and have succeeded in demonstrating the absorption of strontium-90 in foods. The concentration of this isotope, as expected, is dependent on the calcium content of the food, and for this reason the results can be expressed best as strontium-90 activity per gram of calcium. On this basis, milk in the United States during early 1956 contained about 3 micromicrocuries of strontium-90 per gram of calcium. One microcurie of strontium-90 (7) is the commonly accepted permissible content for the adult skeleton (*8*). The skeleton contains about 1000 grams calcium, and the permissible concentration would thus be 1000 micromicrocuries per gram of calcium, or about 350 times the presently observed concentration in milk.

According to the National Academy of Sciences (*9*), "Already some children have accumulated a measurable amount of radioactive strontium in their bodies. The amount, however, is quite small—a thousandth of what is considered a permissible dose."

References and Notes

1. M. Eisenbud and J. H. Harley, *Science* **117**, 141 (1953).
2. ———, *ibid.* **121**, 677 (1955).
3. We express our appreciation to our colleagues who participated in this program of fallout collection. In particular, A. E. Brandt is responsible for the IBM reporting of computations as well as the statistical analyses. Edward P. Hardy, Jr., and Robert S. Morse performed the soil analyses, and Naomi A. Hallden assisted in developing the procedure for computation of the gamma dose. C. L. Dunham made a number of helpful suggestions in the preparation of the manuscript. The continued cooperation of Lester Machta and his staff at the U.S. Weather Bureau has been invaluable.
4. H. F. Hunter and N. E. Ballou, *Nucleonics* **9**, No. 11, C-2 (1951).
5. G. M. Dunning, *Sci. Monthly* **81**, 265 (1955).
6. W. F. Libby, *Science* **122**, 57 (1955).
7. *Natl. Bur. Standards U.S. Handbook 52* (1953).
8. The permissible concentration of strontium-90 is probably lower by a factor of 10 than the concentration that would produce injury. On the other hand, the 1-microcurie level was established for occupational exposure, and the National Committee for Radiation Protection recommends that such levels be reduced to 10 percent for public exposure.
9. *Biological Effects of Atomic Radiation.* Report to the public (Natl. Acad. Sci.–Natl. Research Council, Washington, D.C., 1956); *Science* **124**, 60 (1956).

E. C. Crittenden, Physical Standards Expert

Dr. Eugene C. Crittenden died in Washington, D.C., on 28 March 1956 at the age of 75. He had been a member of the American Association for the Advancement of Science for 40 years. Born at Oswago, Pennsylvania, 19 December 1880, he graduated from Cornell University in 1905 with a B.A. degree—not, as his many associates assumed, in physics—but rather in classical languages. He did, however, have a divided interest in physics and remained as a graduate student and instructor at Cornell University until he accepted an appointment at the National Bureau of Standards as an assistant physicist in July 1909.

Dr. Crittenden was first assigned to the photometric laboratory, where he subsequently made many important contributions. He was named chief of the bureau's electrical division in 1921 and continued in this position until 1946. Under his leadership, the program of the division expanded substantially, keeping pace with the rapid developments in radio and electronics. Major organizational units of the bureau developed from nuclei assembled under Dr. Crittenden's leadership. These include the former ordnance development division, now the diamond ordnance fuze laboratories of the Department of the Army, and the bureau's central radio propagation laboratories at Boulder, Colorado.

In 1933, Dr. Crittenden was made assistant director of the bureau. Subsequently, this title was changed to associate director. Until his retirement in 1950, he was the bureau's senior associate director and, in this period of changing administration and directors, was often called upon to serve as acting director of the bureau. For his outstanding contribution to the Government and to science, he was honored with many awards and recognitions.

For his outstanding contributions to the work of the bureau, Dr. Crittenden was awarded the Department of Commerce gold medal for exceptional service in 1949, which was the year that these awards were established. In 1946 he was honored with the gold medal of the Illuminating Engineering Society for "meritorious achievement conspicuously furthering the profession, art, or knowledge of illuminating engineering." Also in 1946, the Case Institute of Technology awarded him an honorary D.Sc. degree as "a devoted servant of the public, exponent of precise measurement, and international authority on the standards of science and industry."

Dr. Crittenden took an active part in American and international scientific societies. He was president of the Illuminating Engineering Society in 1925, president of the U.S. National Committee of the International Electrochemical Commission from 1939 to 1946, and president of the Optical Society of America in 1932–33. He served as an associate editor of the *Review of Scientific*