role in space exploration. On the consulting and decision-making level the prestige and resources of the Academy will, as in the past, be utilized, though now there will be an intermediate link —the Committee on Coordination— which will in turn exert pressure upon the Academy.

Perhaps these measures are a recognition of a turning point in Soviet technological development: the point of diminishing returns from adaptation of Western technology has been reached, and new and vigorous domestic technological development becomes a necessity. The Soviet political leadership appears to be convinced that the invigoration of technological research activities can be more profitably achieved by separating functions, and by freeing the Academy of Sciences of the U.S.S.R. to concentrate its attention on basic research and the long-run problems of science. Reorganization of the Soviet research setup could provide an effective mechanism for channeling scientific manpower and material resources into strategic areas of the physical sciences and engineering toward the achievement of the most ambitious long-run goal of Soviet power —world leadership in science and technology.

Note added in proof. Right after this article had gone to press, Mr. Khrunichev died of a heart attack on 2 June. His successor, Konstantine N. Rudnev, was named on 10 June.

References and Notes

1. *Pravda* (12 Apr. 1961).
2. Some of the statistical information in this article is taken from N. DeWitt, *Education and Professional Employment in the U.S.S.R.* (National Science Foundation, Washington, D.C., in press).
3. A 14th union-republic academy of sciences is scheduled to begin functioning in the Moldavian S.S.R. in 1962; it is presently a branch of the Academy of Sciences of the U.S.S.R.
4. *Pravda* (14 Feb. 1956).
5. *Uchitel'skaia gazeta* (20 May 1958).
6. *Pravda* (2 July 1959).
7. *Izvestiia* (9 Aug. 1959). Semenov was a Nobel prize winner in chemistry.
8. *Ibid.* (16 Dec. 1959).
9. *Pravda* (31 Dec. 1960).
10. *Ibid.* (12 Apr. 1961).
11. This information was given me by A. V. Topchiev, vice-president of the Academy of Sciences of the U.S.S.R., in Washington, D.C., on 26 and 27 April. Dr. Topchiev attended the plenary session just before his trip to the United States. He indicated that no public announcement of this decision has been made as yet, since the Presidium is still considering the possibility of additional transfers of research institutes.
12. There are seven branches: Bashkir, Dagestan, Karelian, Kazan', Kola, Komi, and Ural. An eighth branch—Moldav—is currently being reorganized into a union-republic academy. The seven branches are being transferred to the jurisdiction of the Council of Ministers of the Russian S.F.S.R.
13. Dr. Topchiev gave as examples the Institutes of Complex Transportation Problems, Metallurgy, and Hydraulic Engineering and Water Economy. In addition, the Institute of Mining will be transferred from the Academy [*Vestnik Akad. Nauk.* **31**, No. 4, 3 (1961)].
14. Dr. Topchiev gave the following as examples: in the Geography-Geology Division, Institutes of Geological Prospecting and of Coal Geology; in the Chemical Division, Institutes of Silicate Chemistry and of Forestry and Wood Chemistry; in the Biological Division, the Institute of Soil Sciences.
15. *Ekonomicheskaia gazeta* (20 Apr. 1961).
16. *Pravda* (20 May 1961); *Ekonomicheskaia gazeta* (20 May 1961).
17. Some of the Academy's institutes which deal with long-range planning, such as the Institute of Complex Transportation Problems, have been transferred to the operating auspices of the State Economic Council.
18. This was especially emphasized by Dr. Topchiev.
19. *Pravda* (21 Apr. 1961).

Radionuclide Fractionation in Bomb Debris

The fractionation systematics for high-yield bursts at sea-water and coral surfaces are delineated.

E. C. Freiling

In radiochemical studies of nuclear detonation debris, the term *fractionation* is used to indicate any alteration of radionuclide composition occurring between the time of detonation and the time of radiochemical analysis which causes the debris sample to be nonrepresentative of the detonation products taken as a whole. The phenomenon has recently been discussed by Adams *et al.* (*1*) and Edvarson *et al.* (*2*) and treated theoretically by Magee (*3*). The alteration observed may have taken place in various stages, and it is helpful to classify these according to the type of processes involved, to list them in approximately chronological order, and to group them under two headings—natural and artifactitious.

Natural fractionation begins with the condensation of radioactive and inert material from the fireball, some radionuclides being preferentially taken up by the condensed phase. The intimate mixture of condensed and solid phase may begin to separate while condensation is still in progress, with further separation of the condensate, according to size, density, and shape, occurring under the influence of wind, gravity, and the turbulence of the cloud. The fractionation taking place through these processes is called primary fractionation in this article.

Further fractionation may then occur through contact of debris with radioactively inert surroundings. For example, soluble radionuclides may be preferentially leached from fallout by sea water, or small particles may preferentially adhere to available surfaces. Fractionation occurring by processes such as these is called secondary fractionation.

Artifactitious fractionation can be induced by sample-collection processes which result in biased samples, by incomplete removal of debris from sampling apparatus, and by faulty analytical procedures.

At this point it appears advisable to introduce two further terms to describe primary fractionation. It is conceivable that in one burst only a small portion of the debris will be sensibly fractionated with respect to two given radionuclides, but that highly unrepresentative ratios of these radionuclides will be produced. In a second burst this pair of radionuclides may be fraction-

---

The author is acting head of the Nuclear and Physical Chemistry Branch of the U.S. Naval Radiological Defense Laboratory, San Francisco, Calif.

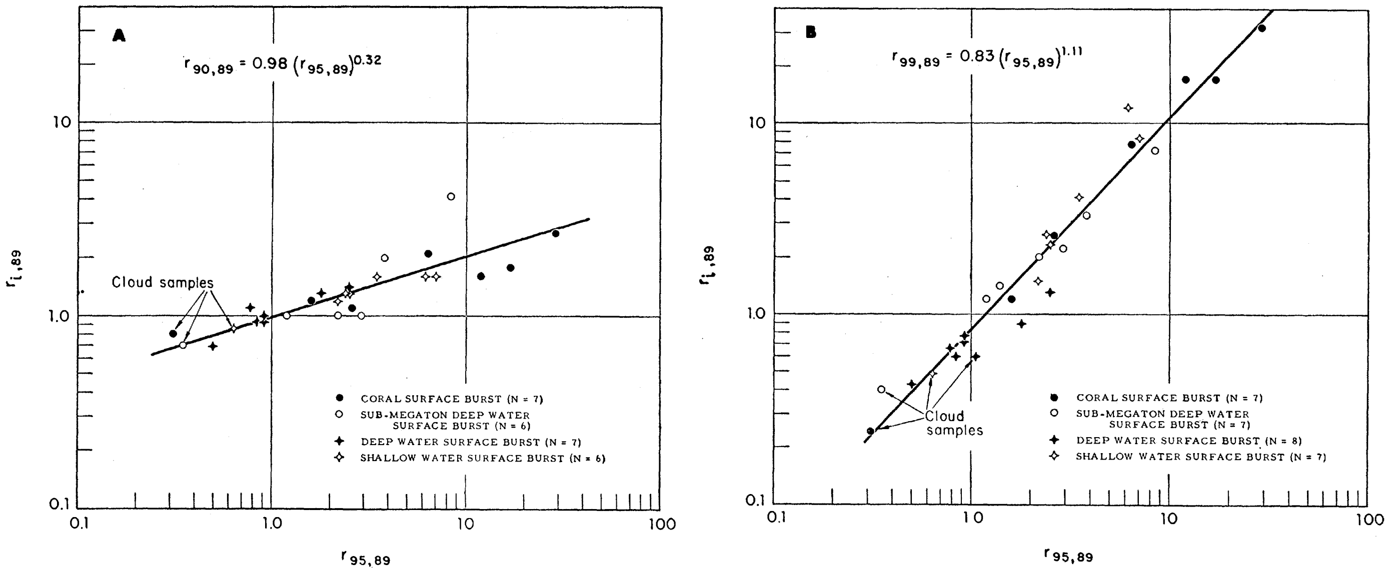


Fig. 1. Logarithmic fractionation correlations for (*A*) strontium-90, (*B*) molybdenum-99, (*C*) tellurium-132, (*D*) cesium-137, (*E*) lanthanum-140, (*F*) cerium-144, (*G*) uranium-237, and (*H*) neptunium-239.

ated throughout the entire debris, but with only small departures from the representative ratio. To distinguish between such types of behavior the term *degree of fractionation* is used to refer to the range of variability of the radionuclide ratio and the term *extent of fractionation* is used to refer to the fraction of the total quantity of these radionuclides which are not present together in the representative ratio. Thus, the first burst referred to above would be said to have fractionated this pair to a high degree but to a small extent, while the second would be said to have fractionated the pair to a low degree but to a large extent.

Importance of Fractionation

An understanding of fractionation phenomena is essential to an understanding of the problems of fallout-contour prediction, device distribution, contamination, ingestion hazard, world-wide fallout, and the nuclear chemistry of the detonation process.

Fallout theories are based upon the gravitational and micrometeorological forces exerted upon the particles resulting from nuclear bomb detonations. The nature of the particles present depends upon the environment, and the ensuing variations in particle size and type are accompanied by variations in radiochemical composition according to the fractionation pattern. Without a knowledge of the latter, correlation and interpretation of field data and prediction of future results is difficult, if not impossible.

In the study of device distribution the term *fraction of the device* is frequently used. This term is undefined for fractionated samples, and the occurrence of completely unfractionated samples is seldom, if ever, demonstrated. Although the term can be redefined in various ways without a knowledge of the degree and extent of fractionation (for example, as "fraction of the device's total gamma-ray activity at time *t*"), this knowledge is necessary to convert results based upon one definition to results based upon another. Furthermore, through considerations of mass balance, a knowledge of fractionation data can contribute to knowledge of device distribution.

The radiation fields produced by contamination and remaining after decontamination vary with the initial radionuclide composition if fractionation is induced by either of these processes.

The hazard of ingestion of bomb-debris particles is proportional to the presence of isotopes with long biological half-life. To evaluate ingestion hazard from field data, the effect of fractionation must be taken into account.

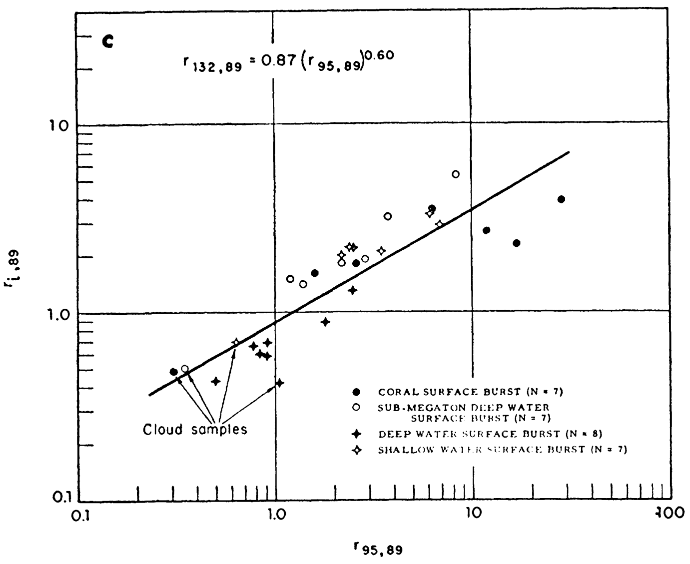
The fraction of strontium-90 contributed to world-wide fallout from a fractionated nuclear detonation cannot be considered equal to the fraction of the gamma-ray activity contributed to world-wide fallout. In the case of high-yield surface bursts, as discussed below, it should be significantly greater.

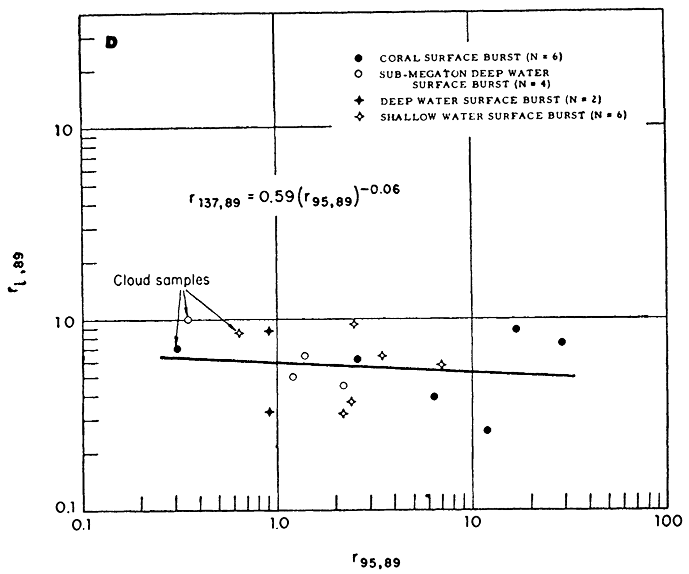
The nuclear chemistry of the detonation process can be studied only in the light of fractionation. An understanding of the fractionation phenomena could lead to a knowledge of the unfractionated composition, which is essential to the study of the primary process. In addition, the dependence of fractionation upon the half-lives and independent yields of volatile precursors relative to condensation time may eventually make possible the study of these parameters through the analysis of fractionated samples.

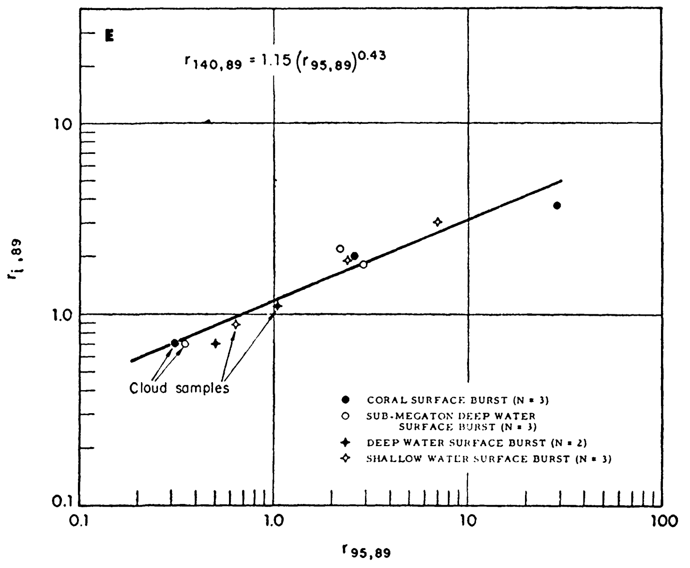
The work discussed here was undertaken to develop empirical relationships for organizing available observations on high-yield surface bursts, illustrate the influence of the nature of the environment, and possibly provide a basis for predicting future fractionation behavior (*4*).

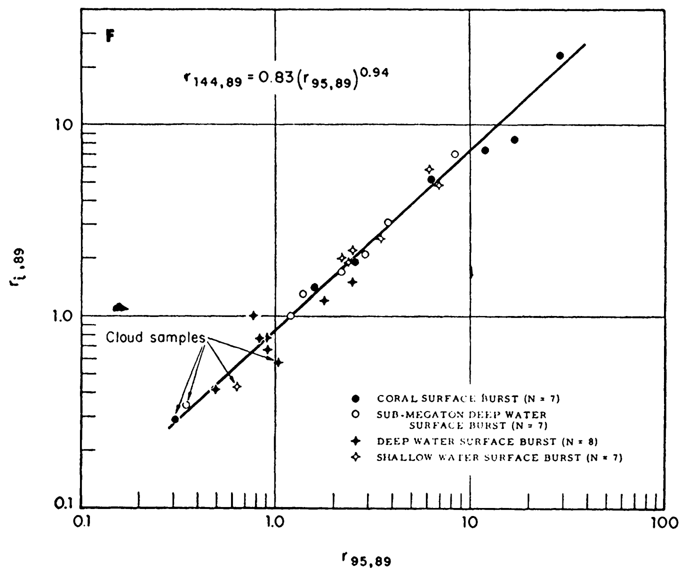
Background

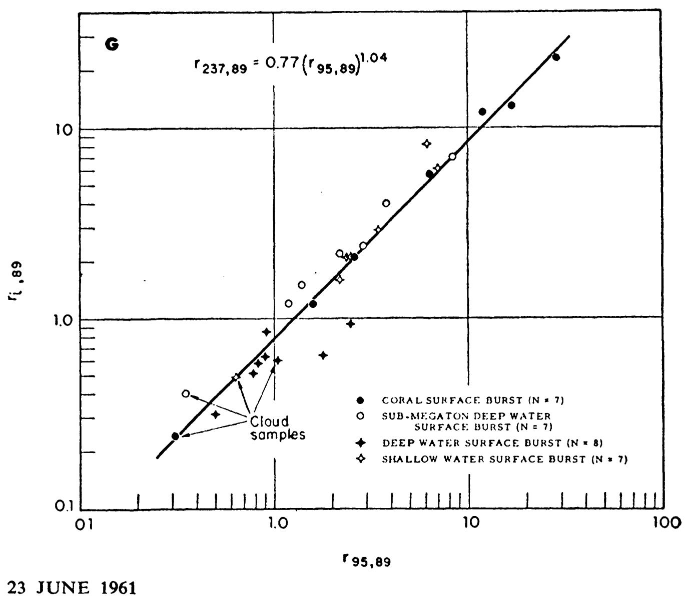
In the atomic cloud the processes of cooling, condensation, coagulation, mixing, and separation occur simultaneously but to different extents in different regions. Furthermore, the initial radioactive products change in elemental form through processes of radioactive decay. The final distribution of each mass chain will therefore be related to its elemental distribution during the various phases of the primary fractionation process. This elemental distribution is simplest in the case of induced activities (Na^{24}, S^{35}, Ca^{45}, Br^{82}, U^{237}, and U^{240}) and of shielded fission


C
$r_{132,89} = 0.87 (r_{95,89})^{0.60}$
Cloud samples
CORAL SURFACE BURST (N = 7)
SUB-MEGATON DEEP WATER SURFACE BURST (N = 7)
DEEP WATER SURFACE BURST (N = 8)
SHALLOW WATER SURFACE BURST (N = 7)
$r_{i,89}$
$r_{95,89}$
0.1
1.0
10
100


D
CORAL SURFACE BURST (N = 6)
SUB-MEGATON DEEP WATER SURFACE BURST (N = 4)
DEEP WATER SURFACE BURST (N = 2)
SHALLOW WATER SURFACE BURST (N = 6)
$r_{137,89} = 0.59 (r_{95,89})^{-0.06}$
Cloud samples
$r_{i,89}$
$r_{95,89}$
0.1
1.0
10
100


E
$r_{140,89} = 1.15 (r_{95,89})^{0.43}$
Cloud samples
CORAL SURFACE BURST (N = 3)
SUB-MEGATON DEEP WATER SURFACE BURST (N = 3)
DEEP WATER SURFACE BURST (N = 2)
SHALLOW WATER SURFACE BURST (N = 3)
$r_{i,89}$
$r_{95,89}$
0.1
1.0
10
100


F
$r_{144,89} = 0.83 (r_{95,89})^{0.94}$
Cloud samples
CORAL SURFACE BURST (N = 7)
SUB-MEGATON DEEP WATER SURFACE BURST (N = 7)
DEEP WATER SURFACE BURST (N = 8)
SHALLOW WATER SURFACE BURST (N = 7)
$r_{i,89}$
$r_{95,89}$
0.1
1.0
10
100


G
$r_{237,89} = 0.77 (r_{95,89})^{1.04}$
Cloud samples
CORAL SURFACE BURST (N = 7)
SUB-MEGATON DEEP WATER SURFACE BURST (N = 7)
DEEP WATER SURFACE BURST (N = 8)
SHALLOW WATER SURFACE BURST (N = 7)
$r_{i,89}$
$r_{95,89}$
0.1
1.0
10
100

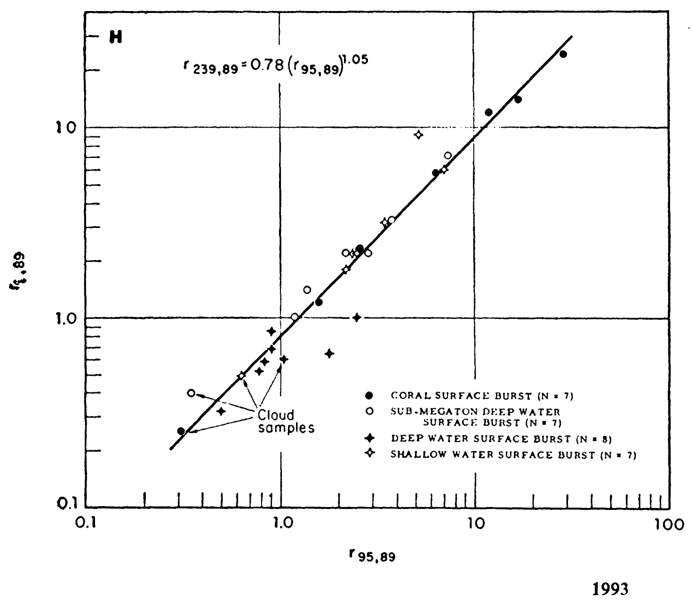

H
$r_{239,89} = 0.78 (r_{95,89})^{1.05}$
Cloud samples
CORAL SURFACE BURST (N = 7)
SUB-MEGATON DEEP WATER SURFACE BURST (N = 7)
DEEP WATER SURFACE BURST (N = 8)
SHALLOW WATER SURFACE BURST (N = 7)
$r_{i,89}$
$r_{95,89}$
0.1
1.0
10
100

Table 1. Ratio of linear variance from linear fit to linear variance from logarithmic fit.

Burst	Sr^{90}	Mo^{99}	Te^{132}	Cs^{137}	La^{140}
Coral-surface burst	1.13		1.26	0.86	2.85
Sub-megaton deep-water surface burst	0.20	0.37	1.03	2.41	1.36
Deep-water surface burst	1.85	1.52	1.15		
Shallow-water surface burst	1.87		1.18	0.92	4.38

products (Br^{82}, Rb^{86}, Nb^{96}, and Cs^{36}), which do not change form between the time of formation and the time of measurement. Neptunium-239 remains predominantly in the form of uranium-239 during the condensation process. The behavior of fission products such as zirconium-95, molybdenum-99, ruthenium-103, ruthenium-106, and the heavier rare earths is simplified by the absence of volatile precursors, but even here the precursors existing during condensation may have varying affinity for the surfaces available for deposit. Volatile precursors are prominent in mass chains 88 through 97 and 137 through 140.

Factors which favor rapid condensation after detonation cause radionuclides with volatile precursors to fractionate more severely than those with refractory precursors, due to the fact that the volatile precursors are abundant at early times after detonation. Such factors would be low device yield and entrainment by the fireball of cool material with high heat capacity.

Factors which promote occlusion of volatile species tend to offset fractionation. Such factors are a rapid rate of cooling and a high concentration of vaporized material.

Factors, such as the formation of heavy fallout particles, which cause rapid initial settling of condensate from the cloud favor fractionation.

From these considerations it would be expected that fractionation in general would decrease with device yield and increase with proximity to the earth's surface. Thus, airbursts would be expected to fractionate less than surface bursts, surface bursts would be expected to fractionate less than subsurface bursts; and solid environments would be expected to produce more severe fractionation than aqueous environments. However, the phenomenon is so complex, and the data are so scattered, that even such generalizations must be advanced with caution. Air bursts, for example, have been known to fractionate severely.

The number of observations that can be made through radiochemical analysis of fractionated debris is very small compared with the number of different sample types produced and the number of variables requiring investigation. Particulate samples consist not only of particles of many sizes but of particles of many types. Available samples have usually been taken at different times during the period of fallout. In view of this situation, an empirical approach to the problem of fractionation appears to offer the most immediate rewards.

Correlation of Data from High-Yield Surface Bursts

Data suitable for fractionation correlations have been obtained from four high-yield surface bursts: a burst of megaton range at the surface of a coral atoll; a burst of megaton range at the surface of deep water; a burst of sub-megaton range at the surface of deep water; and a burst of megaton range at the surface of shallow water. Cloud and fallout samples were obtained from these bursts and were analyzed radiochemically for strontium-89, strontium-90, zirconium-95, molybdenum-99, tellurium-132, cesium-137, lanthanum-140, cerium-144, uranium-237, and neptunium-239. Standard procedures were used for all radionuclides except lanthanum-140, which was determined by gamma-ray spectrometry. The data obtained are assumed to represent the results of natural fractionation. The unknown but presumably minor effect of artifactitious fractionation has been neglected.

Preliminary treatment of data. Consider a fission event in which a total of F fissions occur and in which the yield of radionuclide i (either a fission product or a product of induced activity) is Y_i atoms per fission. If the total number of atoms A_i of any such radionuclide be determined by radiochemical analysis, a value of F can be calculated from the results as

$$F_i = A_i/Y_i$$

and all values of F_i so obtained should be equal. If, however, only a sample of the products is available for analysis, and the number of atoms a_i are determined by radiochemical analysis, values f_i may be similarly calculated as

$$f_i = a_i/Y_i$$

but agreement will be obtained only among those radionuclides which have not fractionated from one another, and variance of f_i values will be indicative of fractionation. Initial conversion of the data into this form by use of the appropriate Y_i values was necessary to eliminate nuclear-physical effects, such as the dependence of the yield of a given mass chain on neutron energy and fissile material, which have no bearing on the present problem.

With zirconium-95 and strontium-89 chosen as reference radionuclides, values of $r_{95,\,89}$, defined as the ratio f_{95}/f_{89}, were then calculated for each sample

Table 2. Least-squares slopes and 95-percent confidence limits of logarithmic fractionation correlations.

Burst	Sr^{90}	Mo^{99}	Te^{132}	Cs^{137}	La^{140}	Ce^{144}	U^{237}	Np^{239}
Coral-surface burst	0.24 ± 0.12	1.10 ± 0.11	0.40 ± 0.21	−0.03 ± 0.39	0.37 ± 0.86	0.92 ± 0.12	1.02 ± 0.22	1.02 ± 0.06
Sub-megaton deep-water surface burst	0.52 ± 0.40	0.89 ± 0.08	0.72 ± 0.13	−0.42 ± 0.51	0.52 ± 1.82	0.94 ± 0.09	0.91 ± 0.09	0.90 ± 0.12
Deep-water surface burst	0.41 ± 0.21	0.60 ± 0.23	0.59 ± 0.50		0.62	0.70 ± 0.44	0.48 ± 1.26	0.56 ± 1.20
Shallow-water surface burst	0.38 ± 0.18	1.32 ± 0.34	0.60 ± 0.23	−0.04 ± 0.70	0.55 ± 0.62	1.06 ± 0.19	1.15 ± 0.22	1.16 ± 0.30
Cumulative slope for high-yield surface bursts	0.32 ± 0.08	1.11 ± 0.09	0.60 ± 0.14	−0.06 ± 0.31	0.43 ± 0.09	0.94 ± 0.06	1.04 ± 0.10	1.05 ± 0.09

Table 3. Least-squares intercepts and 95-percent confidence limits of logarithmic fractionation correlations.

Burst	Sr^{90}	Mo^{99}	Te^{132}	Cs^{137}	La^{140}	Ce^{144}
Coral-surface burst	−0.01 ± 0.11	0.06 ± 0.10	0.03 ± 0.19	−0.24 ± 0.39	0.07 ± 0.58	−0.07 ± 0.11
Sub-megaton deep-water surface burst	0.04 ± 0.23	0.00 ± 0.04	0.04 ± 0.06	−0.20 ± 0.08	0.09 ± 0.61	−0.06 ± 0.05
Deep-water surface burst	0.00 ± 0.46	−0.16 ± 0.05	−0.24 ± 0.10		0.03	−0.10 ± 0.09
Shallow-water surface burst	−0.06 ± 0.09	−0.12 ± 0.19	−0.03 ± 0.13	−0.25 ± 0.35	0.05 ± 0.34	−0.12 ± 0.10
Cumulative intercept for high-yield surface bursts	−0.08 ± 0.05	−0.08 ± 0.05	−0.06 ± 0.08	−0.23 ± 0.20	0.06 ± 0.04	−0.08 ± 0.04

as a measure of the degree of fractionation present. These nuclides were chosen because their tendency to fractionate from one another makes $r_{95,89}$ a sensitive function of the degree of fractionation, because their yields and half-lives are such that they can be determined in samples of relatively low activity, and because they are sufficiently long-lived to be measurable at late times after detonation. The $r_{95,89}$ values obtained varied by a factor of 5 for the deep-water surface burst of megaton range, of 20 for the deep-water surface burst of submegaton range, of 12 for the shallow-water surface burst, and of 100 for the coral-surface burst.

The ratios $r_{i,89}$, defined as f_i/f_{89}, were then calculated for each of the remaining radionuclides in each sample studied. These were plotted logarithmically against $r_{95,89}$. It was found that the data thus plotted could be fitted with straight lines and that all lines passed near the intersection of the unit axes. Slopes of approximately 1 were observed for cerium-144, uranium-237, and neptunium-239, and, in the cases of the land-surface and shallow-water surface bursts, for molybdenum-99. The cumulative plots of the data are shown in Fig. 1.

Comparison of linear and logarithmic correlation. Stevenson (*5*) has made linear correlations of fractionation data from less highly fractionated samples than we have observed. When a wide range of fractionation is observed, however, linear graphs are incapable of portraying both extremes of fractionation on an equal basis, with the result that the detail near the origin is suppressed. Since the data appear to fall near a straight line in either case, the first question to be examined is whether linear or logarithmic correlations fit the observed data more closely. This was investigated by making least-squares fits of the data in both linear and logarithmic form and comparing the variances. Strictly speaking, the usual least-squares equations do not apply to the case at hand. Inherent in their derivation is the assumption that the abscissa values are known with much greater precision than the ordinate values, while here the two are known with approximately equal precision. However, because it was expected that results would be used as though abscissa values were accurately known, the usual equations were used. Both linear and logarithmic variances were expressed in the linear form

$$s^2 = \frac{\Sigma(r_{i,89} - \tilde{r}_{i,89})^2}{N - 2}$$

for comparability. Here N is the number of observations, and the tilde signifies the values of the ordinate as computed from the empirical equation. The ratios of logarithmic to linear variance thus expressed are shown in Table 1 for those cases where the slopes differed appreciably from unity. From the data it would appear that logarithmic correlation is definitely preferred for tellurium-132 and barium-140, while for strontium-90, molybdenum-99, and cesium-137 the type of correlation preferred may depend upon the yield. On the whole, the data favor logarithmic correlations, but the preference is far from decisive. Since the results of a single analysis can have a large effect on the ratio presented, further evidence is needed before any firm conclusions can be drawn.

Comparison of slopes for logarithmic correlations. The values of the slopes obtained from least-squares fits to the logarithmic data are shown in Table 2, together with the 95-percent confidence limits calculated from the *t*-table. In addition there are shown the slopes and confidence limits for the cumulative data for each radionuclide. These were obtained by fitting the lines to all the data for a given radionuclide, regardless of burst. The cumulative least-squares lines are shown in Fig. 1. (*A–H*). It may be seen that, except for molybdenum-99 in the two deep-water surface bursts, the cumulative slopes are within the confidence limits for the individual slopes. In these cases the slope is at least an insensitive function of the environments studied and may be used to predict fractionation behavior in future high-yield surface bursts. In the water-surface bursts, molybdenum-99 appears to behave more like tellurium-132 than like zirconium-95.

Comparison of intercepts for logarithmic correlations. The least-squares values of the intercepts and the associated 95-percent confidence limits are shown in Table 3. If the intercepts represent the unfractionated composition, all values should be zero within the confidence limits of the results. Consistent departures from zero, as shown in the case of cerium-144, may merely indicate systematic errors in the calibration and conversion factors used. The cumulative values of the intercepts fall within the limits of the individual values for all radionuclides except molybdenum-99 and tellurium-132. For each of these radionuclides the discrepancy appears to be due to the deep-water surface burst of megaton range, since all other values are zero within the limits of the data. The cumulative intercepts for the remaining radionuclides are not sensitive to the nature of the surfaces studied and may be used to predict fractionation behavior.

Goodness of fit. In addition to the variance, a more readily interpretable measure of the goodness of fit is given by the ratio of predicted-to-observed value for the point that shows the poorest fit. This ratio is given in Table 4 for each radionuclide studied. Where the ratio is less than unity, the recip-

Table 4. Predictability factors for points with the poorest fit.

Radionuclide	Predictability factor
Mo^{99}	1.9
Sr^{90}	2.2
Te^{132}	2.1
Cs^{137}	2.0
La^{140}	1.4
Ce^{144}	1.5
U^{237}	2.2
Np^{239}	2.1

rocal is given instead, in order to facilitate comparison. It may be seen that, even in the cases of poorest fit, the cumulative lines predict the data to within a factor of about 2.

Correlation of slopes with precursor volatility. It has long been recognized that fractionation is primarily caused by the presence of volatile precursors in the decay chains of the fractionating radionuclides. The decay chains of the fission-product radionuclides correlated were taken from the Bolles-Ballou compilation (*6*) and modified according to more recent results; they are presented in Fig. 2. Included are chains for uranium-237 and neptunium-239. Below each element in a given chain is presented the fractional chain yield, as calculated (i) by the theory of Present (*7*) and (ii) by the theory of Glendenin, Coryell, and Edwards (*8*).

To illustrate the dependence of the slopes found upon the volatility of the precursors, and at the same time to obtain an empirical correlation between slopes and volatility, the following procedure was followed. First, the fraction of the decay chain existing in a refractory form at a given time was calculated from the Bolles-Ballou compilation, according to Present's theory, and designated F_R. It was assumed that halogens, rare gases, alkali metals, and tellurium were volatile. The cumulative slopes were then plotted against $F_R^{1/2}$ for values of F_R calculated at 35 seconds. The results are shown in Fig. 3. It may be seen from the figure that the cumulative slopes equal $F_R^{1/2}$ within their confidence limits. It should not be inferred from this relationship that the condensation time is actually 35 seconds or that there is any theoretical significance to the function $F_R^{1/2}$. Condensation probably occurs over an extended period of time. The fractional chain yields are not well known, nor are some of the half-lives involved. Furthermore, the fractional chain yields vary with fissile material and neutron spectrum. To repeat, for emphasis: the correlation is intended only to illustrate the relationship between precursor volatility and slope and to provide a rough rule of thumb for estimating slopes of unobserved radionuclides. This rule may be written

$$\log r_{\mathrm{i},\,89} = F_R^{1/2} \log r_{95,\,89}$$

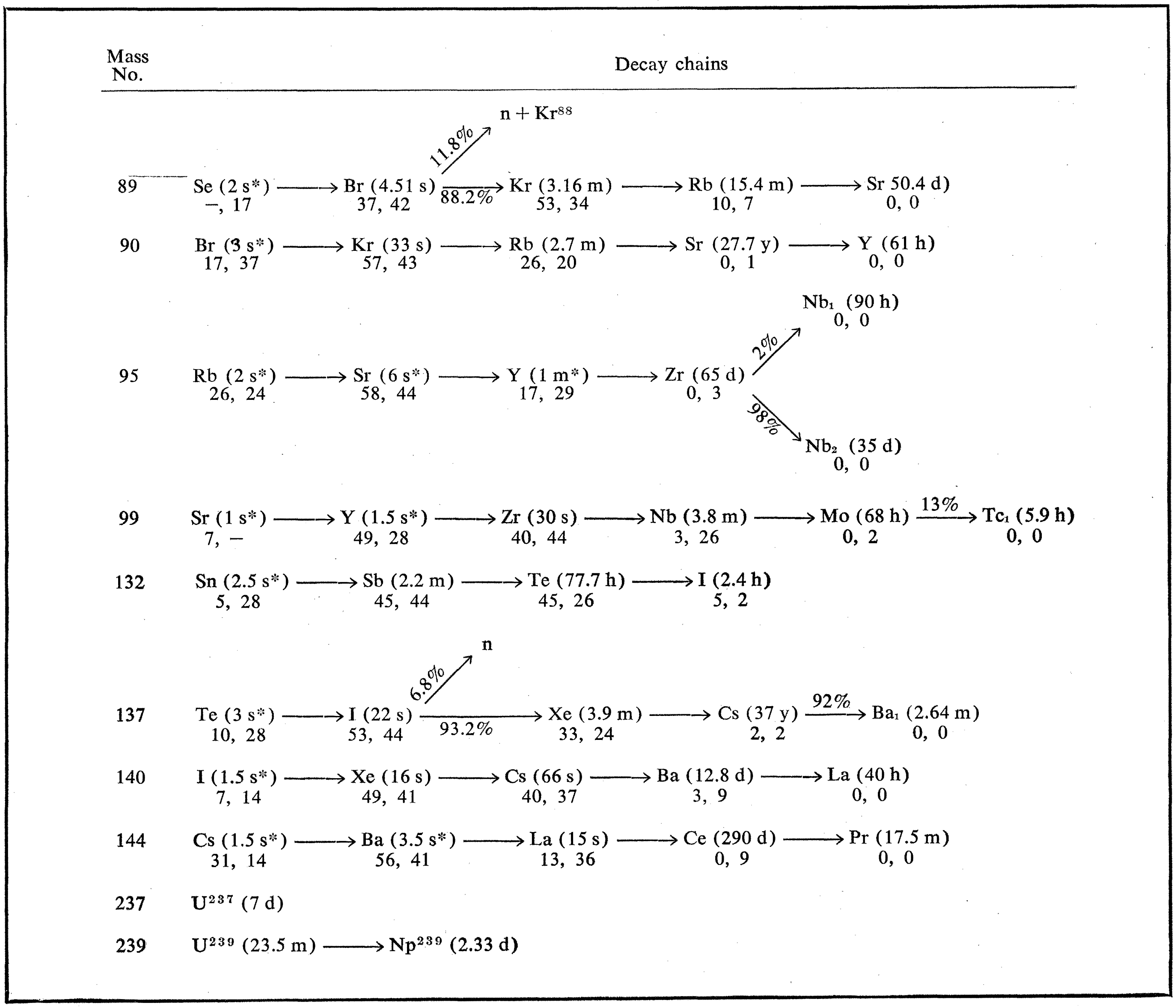


Fig. 2. Decay chains of radionuclides studied. Half-lives in parentheses (asterisks indicate estimated values).

Possible Applications

Correlation with fallout pattern. In all cases except that of the deep-water surface burst, the cloud sample was found to be rich in strontium-89. Beyond this it has not been possible to make any meaningful correlation between sample location and degree of fractionation for the samples studied. This is at least partially due to the fact that the number of samples studied is inadequate as compared to the complexity of the fallout patterns. The correlations developed, however, give rise to the expectation that in any future detonation, sufficient documentation could be achieved to obtain the desired correlation by analyzing a large number of samples for one refractorily behaving nuclide (Zr^{95}, Ce^{144}, U^{237}, or Np^{239}) and for one volatilely behaving nuclide (Cs^{137} or Sr^{89}), inferring the remaining composition from more complete analyses performed on selected samples of varying degrees of fractionation.

Fraction of the device. From the results presented, it may be seen that, in shots of the type studied here, there is a fairly large group of refractorily behaving radionuclides which do not grossly fractionate from zirconium-95 in even highly fractionated samples. It is expected that niobium-95, zirconium-97, niobium-97, rubidium-103, and all mass chains above 144 would behave similarly. If one of these radionuclides —say zirconium-95—were chosen for documentation, it would appear most reasonable and convenient to relate device distribution to the time-independent quantity "fraction of the device's zirconium-95." This quantity should then be very nearly equal to the fraction of the device's refractorily behaving radionuclides. Because of the large number of high-yield radionuclides involved, the corrections necessary to convert from this quantity to other quantities of interest, such as the "fraction of the device's gamma-ray activity at time t," would be minimized in both number and magnitude. The same principle would apply to the correction for variation of survey-instrument response with radiochemical composition.

Device distribution. From the correlation data the number of fissions at any location calculated from the mass i radionuclide is related to similar quantities for zirconium-95 and strontium-89 through the slope b by the equation

$$f_{\mathrm{i}} = f_{95}^{\,\mathrm{b}_{\mathrm{i}}} f_{89}^{\,1-\mathrm{b}_{\mathrm{i}}} = r_{95,\,89}^{\,\mathrm{b}_{\mathrm{i}}-1} f_{95}$$

From mass balance considerations, integration over the contaminated environment gives the total number of fissions

$$\int \mathrm{d}f_{\mathrm{i}} = F = \int \mathrm{d}f_{95}$$

Therefore, as a check of consistency between device distribution data and fractionation data, the equations

$$\int r_{95,\,89}^{\,\mathrm{b}_{\mathrm{i}}-1}\ \mathrm{d}f_{95} = \int \mathrm{d}f_{95}$$

should be satisfied for every radionuclide studied.

Contamination-decontamination. The first and second beta-decay products of rare-gas radionuclides are alkali metals and alkaline earth metals, respectively. These daughters tend to be concentrated in the surface regions of fallout particles and also to exhibit high solubilities. As a result, these volatilely behaving fission products are more easily leached from the fallout by aqueous media than the refractorily behaving radionuclides. One would therefore expect that the results of contamination or decontamination processes involving an aqueous phase, when measured in terms of gross activity, might differ considerably for volatile-rich and volatile-poor debris which were otherwise similar. In order to insure meaningful results from such studies, then, the radiochemical composition of the debris would have to be known, and contributing variations in composition would have to be accounted for. For high-yield surface bursts, the composition could be estimated from a knowledge of $r_{95,\,89}$, on the basis of the results presented here.

Nuclear chemistry. Suppose that a device has been detonated for which the values Y_{i} are unknown, and that no unfractionated samples are available. Suppose, however, that the fractionation correlation curves can be assumed to intersect at a point, from previous experience with devices of similar yield detonated under similar conditions. Correlation curves, either linear or logarithmic, can then be constructed. If it

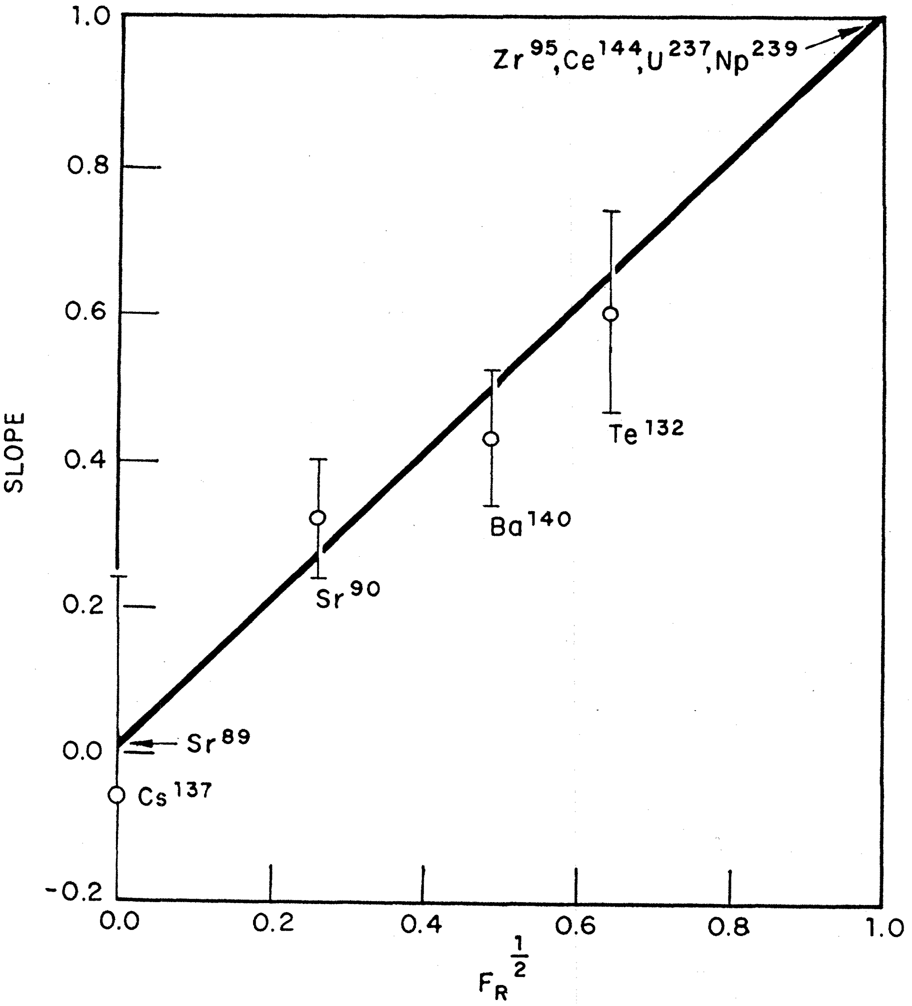


Fig. 3. Dependence of correlation slope on volatility of the precursor.

is now assumed that $Y_{95} = Y_{140}$, the values for unfractionated composition can be determined from the intersections of the curves with the vertical line through the point at which $f_{95} = f_{140}$. An estimate of the validity of the assumption that $Y_{95} = Y_{140}$ can be obtained from values in Table 5, as calculated from the Katcoff compilation (*9*).

Table 5. Variation of mass-140/mass-95 yield for various conditions of neutron fission.

Fissioning nucleus	Neutron energy	Y_{140}/Y_{95}
U^{235}	Thermal	1.03
U^{233}	Thermal	1.02
Pu^{239}	Thermal	0.96
U^{238}	Fast	1.00
Th^{232}	Fast	0.94

Prediction of future behavior. The correlations presented here may be useful in estimating fractionation behavior in future high-yield sea-water surface and coral-surface bursts. It is to be expected that fractionation behavior from lower-yield devices, and from high-yield devices detonated over other surfaces (for example, silicate), may differ significantly from that presented here. In the absence of specific information, however, the behavior presented here may prove to be a useful basis of approximation for planning purposes.

Summary and Discussion

The empirical approach taken here to the problem of fractionation has led to a number of valuable correlations.

1) The composition of fractionated samples from high-yield surface bursts can be correlated logarithmically.

2) The slopes obtained for the various mass chains (except for molybdenum-99) are relatively insensitive to the environment and are empirically related to precursor volatility.

3) Zirconium-95, cerium-144, uranium-237, and neptunium-239 do not fractionate grossly from one another, nor does molybdenum-99 fractionate from these radionuclides when coral is in the environment; cesium-137 does not fractionate grossly from strontium-89. The fact that the slopes of the correlation curve for two radionuclides are identical within their respective margins of error cannot be interpreted as meaning that no fractionation was observed. Small degrees of fractionation between similarly behaving radionuclides are best tested by plotting the ratio of their f values against the fractionation index.

4) Even the data with the poorest fit fall wide of the correlation lines only by a factor of about 2.

5) In all cases except that of the deep-water surface burst, the cloud sample was found to be rich in strontium-89.

It is evident at this point that several pieces of information are lacking for the data presented here. First in importance is the effect of artifactitious fractionation. The extent to which this has contributed to the scatter of points about the correlation lines is undeterminable. Because of the relatively small number of points, any error in the data from the most highly fractionated sample for a given burst would severely affect the type of correlation preferred and the resulting slope.

The behavior of the deep-water surface burst is anomalous in a number of respects, and supporting information on such detonations is needed.

The conditions studied represent a small proportion of the many possible conditions. The findings will have to be viewed in the light of similar studies on many other types of bursts before any firm conclusions and reliable generalizations can be reached.

References and Notes

→ C. E. Adams, N. H. Farlow, W. R. Schell, *Geochim. et Cosmochim, Acta* **18**, 42 (1960).
→ K. Edvarson, K. Low, J. Sisefsky, *Nature* **184**, 1771 (1959).
3. J. L. Magee, *Mechanisms of Fractionation, Rept. No. M-7140* (1953).
4. I wish to acknowledge the excellent work of L. R. Bunney, who supervised the preparation and distribution of samples, coordinated the radiochemical analyses, and accumulated the fission-product results; of L. Wish, who performed the majority of U^{237} and Np^{239} analyses; of Miss E. M. Scadden, who made the Mo^{99} determinations; of J. Pascual, who made the Te^{132} determinations; of J. L. Mackin, P. O. Strom, and D. MacDonald, who performed the Sr^{89}, Sr^{90}, Cs^{137}, and Ce^{144} analyses; and of W. E. Thompson and P. D. LaRiviere, who obtained the La^{140} results. The comments of C. E. Adams have been most helpful in preparing this article.
5. P. C. Stevenson, Univ. of California, Lawrence Radiation Laboratory, private communication.
6. R. C. Bolles and N. E. Ballou, "Calculated activities and abundances of U^{235} fission products," *U.S. Naval Radiological Defense Laboratory Rept. No. USNRDL-456* (1956).
→ R. D. Present, *Phys. Rev.* **72**, 7 (1947).
8. L. E. Glendenin, C. D. Coryell, R. R. Edwards, *National Nuclear Energy Ser. Div. IV* **9** (1951); L. E. Glendenin, *Mass. Inst. Technol. Tech. Rept. No. 35* (1949).
9. S. Katcoff, *Nucleonics* **16**, No. 4, 78 (1958).

Jerome T. Syverton, Microbiologist

Jerome T. Syverton began an outstanding, active career in microbiology as an instructor in bacteriology at the University of North Dakota in 1928. Born in Courtenay, North Dakota, on 29 March 1907, he entered the University of North Dakota in 1923, obtaining an A.B. degree in 1927 and a B.S. degree in 1928. He graduated from Harvard University School of Medicine in 1931, and after an internship and assistant residency in medicine at Duke University Hospital in 1931–32, he became an assistant in pathology and bacteriology at the Rockefeller Institute for Medical Research. There he obtained basic knowledge in virology under the guidance of P. K. Olitsky, and in 1932 he became a member of the faculty of the University of Rochester School of Medicine and Dentistry. He remained at Rochester until 1947, except for a sabbatical leave in 1942 at Vanderbilt University School of Medicine, as visiting associate professor of pathology and bacteriology.

Syverton's development of outstanding skill as a teacher and student adviser and his fundamental and pioneering work, involving, for example, tumor viruses and multiple viral infections of single animal cells, occurred during the period 1934–47, while he was in the department of bacteriology at the University of Rochester. For his outstanding research in virology he received the Lilly award in bacteriology