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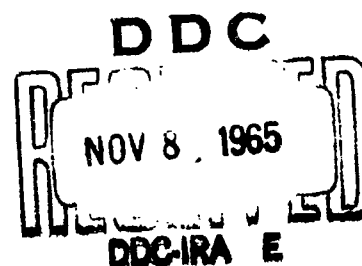
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THE EFFECT OF RADIONUCLIDE FRACTIONATION ON THE NORMALIZATION FACTOR FOR FALLOUT FIELDS

by

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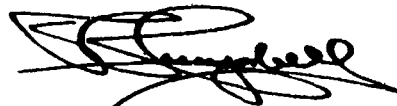
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ABSTRACT

This report compares the normalization factor, r/hr per kt/mi², calculated for unfractionated fission products with the normalization factors calculated from field data for a near-surface silicate soil burst and a silicate soil cratering burst. The large discrepancies between predicted and observed values appear to be caused by a combination of radionuclide fractionation, ground roughness and instrument self-shielding, and gradient effects. Fractionation effects can cause a difference of a factor of five in the normalization factors for surface and cratering bursts, allowing about 50 % reduction in radiation due to ground roughness and instrument self-shielding.

Ionization-chamber measurements on field-collected samples are correlated with their degree of fractionation in this report, and a reasonable correspondence between the ionization-chamber readings and the exposure rates measured in the field is established.

SUMMARY

Predictions of the hazards resulting from fallout are usually based on the exposure rates calculated for unfractionated fission products. In real situations, the unfractionated composition is encountered only rarely, if ever. Since the radionuclides which are the principal contributors to gamma radiation for many days after detonation are subject to strong fractionation, predictions based on the unfractionated composition may be greatly in error. In this report, field data on the radiological properties of debris from shots Smallboy, Johnny Boy, and Danny Boy are correlated with the radiochemical analyses on the debris. Large discrepancies occurred between predicted and observed values of the radiological properties. These can be at least partially resolved when the fractionation of the radionuclides is taken into account.

I. INTRODUCTION

1.1 One of the most basic parameters involved in interpreting and predicting fallout patterns is the concept of a normalization factor. This factor gives the exposure rate at a point 3 ft above an infinite smooth plane uniformly contaminated with fission products. The units generally used for exposure rate are r/hr and the contamination density is expressed as kt of fission products per square mile. The normalization factor, sometimes designated as K, is the ratio of exposure rate to contamination density "r/hr per kt/mi²." To be meaningful, the ratio must refer to fission products of a specified age, since it decreases as the fission products decay. In practice, this is usually taken as one hour.

1.2 Much confusion and uncertainty has arisen from attempts to assign a value to the normalization factor. A part of the difficulty arises from the fact that different procedures have been used in arriving at the value and communications do not always make clear exactly how a particular value was obtained. Some points which should be specified in proposing a value are: (1) whether the value was obtained by theoretical computation, by extrapolation and conversion of laboratory measurements of gamma-emission spectra or 4-pi ionization-chamber decay, or by the integration of 1-hour exposure rate contours from field data; (2) whether it refers to a laboratory-produced fission-product composition, such as that obtained from the thermal-neutron irradiation of U²³⁵, or to a composition more frequently encountered in the field, such as the fission-spectrum neutron fission of U²³⁵ or Pu²³⁹ or the thermonuclear fission of U²³⁸; (3) whether the value refers to fractionated or unfractionated debris; (4) whether the value takes into account the exposure rate contributions of capture products in the debris; and (5) whether or not it accounts for the loss of material into worldwide fallout.

1.3 Adding to the confusion is the fact that agreement is not always obtained where it is to be expected; e.g., theoretical computations by different authors do not agree among themselves or with values obtained by extrapolation and conversion of laboratory measurements. With regard to the latter, experimental determinations of the normalization factor have been limited by the impracticability of reproducing the

uniformly-contaminated infinite plane situation for laboratory measurements. Attention has centered mainly on the measurement of the gamma-emission spectra of fission-product mixtures and of the decay of such mixtures in 4- π ionization chambers. However, the gamma-spectral measurements so far available are not adequate in range and resolution for a precise determination of the normalization factor. Ionization-chamber measurements can be made very accurately, but are not in themselves sufficient to determine the factor. In order to transform from the 4- π geometry to the infinite plane geometry, specific assumptions about the spectral distribution must be made.

1.4 Table 1 lists some values which have been computed for the normalization factor for unfractionated fission products. Miller's¹ values were based on calculations of individual radionuclide activities in the fission-product mixture combined with available information on gamma-photon abundances, according to the method of Gates and Eisenhauer,² to yield the exposure rate. The values computed by Crocker, taken from a forthcoming report, were calculated in essentially the same way, but utilized updated and more complete input data. The values attributed to Björnerstedt³ have been calculated from Björnerstedt's spectral predictions.

1.5 Experimental gamma-spectral measurements on U²³⁵ fission products were reported by Zobel and Love⁴ and in revised form by Maienschein, et. al.⁵ Unfortunately, these did not extend to energies below 0.240 Mev. More recently, Fisher and Engle⁶ measured spectra with a low-energy cutoff at 0.120 Mev, but these were for times less than 45 seconds after fission. Zigman and Mackin and co-workers^{7,8} have studied the decay of fission products of thermal neutron fission of U²³⁵ and Pu²³⁹ in a 4- π ionization chamber. From these measurements they have calculated normalization factors. The conversion was made by combining the experimental data reported by Maienschein with the low-energy predictions of Miller. The results are reported to agree within $\pm 20\%$ with Miller's for thermal neutron fission of U²³⁵. Assuming the same spectral distribution for thermal neutron fission products of Pu²³⁹, the predicted gamma-energy release per fission is 5 to 30 % greater than that for U²³⁵.

1.6 There has not yet appeared in the literature a thorough evaluation of field data from the viewpoint of determining the effect of fractionation on the normalization factor. The effect of fractionation on the normalization factor has been discussed from the theoretical point of view by Freiling and Rainey.⁹ Freiling, Kay and Sanderson¹⁰ have estimated the effect that fission-product fractionation might have on the exposure rate for several conditions of interest. Their estimates were made for the thermal neutron fission of U²³⁵ and the pertinent aspects of their results are shown in Table 2.

TABLE 1
Some Reported Values for the Normalization Factor

Fission Process	Author	Normalization Factor (r/hr per kt/mi ²)	
		at 1 hr	at 10 hr
U ²³⁵ , thermal neutron	Miller	3950	
	Crocker	3260	167
	Björnerstedt	3096	
U ²³⁵ , fission spectrum neutrons	Miller	3940	
	Crocker	3110	169
	Björnerstedt	3050	
U ²³⁵ , 14 Mev neutrons	Crocker	2710	149
	Björnerstedt	2780	
U ²³⁸ , fission spectrum neutrons	Crocker	3120	163
	Björnerstedt	2890	
U ²³⁸ , 14 Mev neutrons	Crocker	2980	160
	Björnerstedt	2810	
Pu ²³⁹ , thermal neutrons	Miller	3480	
	Crocker	2870	152
	Björnerstedt	2690	
Pu ²³⁹ , fission spectrum neutrons	Miller	3400	
	Crocker	2720	149
Pu ²³⁹ , 14 Mev neutrons	Crocker	2370	132
U ²³⁸ , thermonuclear fission	Crocker	2910	158
U ²³⁸ , 8 Mev neutrons	Miller	3610	

TABLE 2

Percent of Unfractionated Exposure Rate
Remaining for the Most Extremely Fractionated Samples

Time After Detonation (hr)	Percent Remaining	
	Time of Debris 6 sec	Solidification 41 sec
1.12	19	30
23.8	32	48

1.7 This report undertakes to present an evaluation of field data from the extensively documented shot Smallboy. Pertinent evidence from the much smaller body of available data on the shots Johnny Boy and Danny Boy is also presented. The data are treated by separately analyzing the ionization chamber measurements on field samples and the exposure rates recorded in the field. Each of these kinds of data is correlated with radiochemical composition, and the results of the correlations are then compared. Also, the normalization factor from field exposure rates is compared with the ratio of integrated fallout patterns to total yield, and the significance of the agreement discussed. Sources of error are analyzed and the reliability of the apparent effect discussed.

1.8 A by-product of this report which should not be overlooked is the evaluation of the ionization chamber as a potential instrument for readily determining, by itself or in conjunction with other measurements, the number of Zr^{95} equivalent fissions in a sample, the degree of fractionation in a sample, and the exposure rate produced in the field by the sample.

II. SOURCES OF DATA

2.1 Most of the data discussed in this report are derived from measurements and analyses made by U. S. Naval Radiological Defense Laboratory (USNRDL) on local fallout from event Smallboy. Data from studies in connection with events Johnny Boy and Danny Boy are also referred to, either as supporting evidence or by way of contrast. USNRDL's participation in these events and the data derived therefrom are documented in the following reports:

Smallboy: Project Officer's Reports POR-2215,¹¹ POR-2216¹² and POR-2217¹³
Johnny Boy: Project Officer's Report POR-2289¹⁴
Danny Boy: USNRDL Technical Report in publication.*

Useful data on event Danny Boy were also obtained from Reference 15. With the exception of the entries in Table 3 referring to 1956 and 1957 events, all data used in this report were obtained from the references just quoted.

2.2 For purposes of orientation, the shot conditions for these events were as follows: Smallboy was a low-yield shot fired from a wooden tower 10 feet above alluvial soil in Area 5 at Nevada Test Site; Johnny Boy was a low-yield burst 23 inches below the surface of basaltic material in Area 18, and Danny Boy was a low-yield detonation buried at a depth of 33.5 meters in basalt on Buckboard Mesa (also in Area 18).

*F. K. Kawahara, L. R. Bunney, E. C. Freiling, and G. R. Crocker.
"Radiochemical Analysis and Some Physical Observations on Selected Danny Boy Fallout Samples." U. S. Naval Radiological Defense Laboratory Technical Report, San Francisco, California, in publication.

The local fallout fractionation effects observed in Smallboy and Johnny Boy are the kind expected in low-yield surface bursts, while those observed in Danny Boy appear to be typical of cratering events. That is to say, the gross samples from Smallboy and Johnny Boy were depleted in fractionating* radionuclides while the samples from Danny Boy were enriched.

*Such terms as "fractionating," "fractionation," and "fractionate," are strictly meaningful only insofar as they express or imply "fractionating from some reference substance," etc. In practice, radionuclides from mass chains with volatile members are often called fractionating and those from mass chains with all refractory members, serving as reference nuclides, are called non-fractionating. An alternative terminology refers to the two kinds of chains as "volatile" and "refractory" respectively. In fact, these latter terms are also commonly applied (through abbreviation of speech) to the nuclides themselves. Thus Sr^{89} is referred to as volatile, although the volatility is characteristic of the precursor bromine, krypton, and rubidium in the mass-89 chain and not of the strontium.

III. LABORATORY IONIZATION CHAMBER MEASUREMENTS

3.1 Extensive use was made of the USNRDL 4-pi ionization chamber¹⁶ in the study of the fallout collections from shots Smallboy and Johny Boy. Several features recommend this kind of instrument for radioactivity measurements. It is simple, stable, and capable of yielding reproducible measurements quite easily and rapidly over a wide range of activities. In addition, the results should correlate fairly well with exposure-rate measurements made in the field with such instruments as the USNRDL Gamma Intensity-Time Recorder (GITR) and the AN/PDR-39 (TLB) survey meter, since these also operate on the ionization-chamber principle.

Measurements with the 4-pi ionization-chamber were made in the field and were used to estimate the number of equivalent fissions* in the samples. The samples ranged in size from a few milligrams to several grams. The estimates of equivalent fissions were used as a basis for deciding the further disposition of the samples. Since this was an important field decision, it is of interest to determine how accurate the estimates are. The correlation of the measurements with the radiochemical results is also important to the study of the normalization factor for the fallout field. Although the relation between the 4-pi ionization-chamber measurements and the GITR measurements made in the field is not straightforward, because of the differing source geometries, it is clear that gross effects on the one kind of measurement due to varying radiochemical composition of the fallout samples should be reflected in the other kind of measurement.

3.2 Treatment of Data

Since the 4-pi ionization chamber was known to be slightly non-linear at high current readings, a linearity correction based on decay measurements of a standard $\text{Ba}^{140}\text{-La}^{140}$ sample was first applied in this region. Then readings were corrected for background and normalized

*Equivalent fissions is defined as the number of atoms of fissionable material which must have undergone fission to produce the amount of a given fission-product radionuclide observed in a sample. Unfractionated debris (assumed in making the estimates) will contain the same number of equivalent fissions of all the fission-product nuclides; in the case of fractionated debris, the number will vary.

on the basis of the daily instrument reading for a standard 100 μg equivalent Ra standard. The true reading of the Ra standard was taken to be 560×10^{-9} milliamps. The normalized ionization-chamber milliamp readings, which had been made at times varying from 50 to 220 hr, were decayed to a common time-point. The decay was calculated by use of the $t^{-1.2}$ rule, and 100 hr was chosen as a convenient intermediate time-point. The decayed readings were tabulated along with the corresponding numbers of Zr^{95} equivalent fissions found in the samples by radiochemical analysis. The ratio of milliamps of current at 100 hours to equivalent fissions of Zr^{95} (referred to hereafter as the ma/fission ratio) was then calculated for each sample. No appreciable variation in this ratio would be expected if the debris samples were unfractionated, or even if the degree of fractionation of each nuclide remained the same from sample to sample. In the latter case, however, the decay correction might introduce some error, as is shown in Section III.3.3.

3.3 Results

3.3.1 Range of ma/fission ratios. The ma/fission ratio calculated from both the Smallboy and the Johnny Boy data showed large variations. This was not unexpected, since a superficial comparison of the preliminary equivalent-fission estimates with the radiochemical data had disclosed disagreements ranging from minor to major proportions. For Smallboy, a few extreme values of the ratio differed by a factor larger than 100 and a large number of the values differed by a factor of at least 10. For the Johnny Boy samples, the ratio showed somewhat less variation than that for Smallboy (although this may be related to the fact that the number of data points was smaller) and lay within the same range.

Table 3 shows the range of values observed. The last three entries summarize, for purposes of comparison, the rather scanty data available from reports of previous operations^{17,18} which can be utilized for this calculation. The theoretically calculated value of the ratio for unfractionated fission products of the thermal-neutron fission of U^{235} at 100 hours is 2.7×10^{-20} . This value lies within all of the ranges shown in Table 3. It appears from the table that variations in the ma/fission ratio larger than an order of magnitude need to be accounted for.

3.3.2 Correlations of the ma/fission ratio. Since the range of values of the ratio was much larger than had been expected, attempts were made to correlate the ratio graphically with three other variables: (1) the degree of fractionation of the samples, as defined by Freiling, et. al.,¹⁰ (2) the weight and the specific activity of the sample, and (3) the time at which the ionization-chamber measurement was made.

TABLE 3

Values of the ma/Fission Ratio at 100 Hours After Detonation*

	Number of Data Points	Minimum Value of Ratio	Maximum Value of Ratio
Smallboy	108	2.35×10^{-21}	1.6×10^{-19}
Johnny Boy	27	1.2×10^{-20}	6.2×10^{-20}
1956 PFG Bursts	13	1.87×10^{-20}	1.08×10^{-19}
1957 NTS Bursts			
Gross Samples**	10	1.45×10^{-20}	6.44×10^{-20}
Individual Particles	9	1.8×10^{-20}	1.4×10^{-19}

* Some calibration differences may exist between the Smallboy and Johnny Boy data on the one hand and the 1956 and 1957 data on the other.

**D. Macdonald, P. Zigman, J. Mackin, P. Strom, "Measurements of Fall-out Samples From the Priscilla, Diablo and Shasta Detonations of Operation Plumbbob (U)," U. S. Naval Radiological Defense Laboratory, Technical Memorandum No. 94, 1958 (CONFIDENTIAL).

1. Degree of fractionation. The ionization-chamber data on Smallboy and Johnny Boy represent a fairly wide range of fractionation. This resulted from the treatment of sieve fractions as individual samples, since the degree of fractionation in local fallout varies rather strongly with particle size. The inclusion of cloud samples, which differed markedly in fractionation from the ground-collected samples, also extended the range.

The ratio of the equivalent fissions of Sr^{89} in a sample to the equivalent fissions of Zr^{95} , denoted by $r_{89,95}$, has been found to be a useful indicator of the degree of fractionation of the sample. Theoretical considerations predict a strong dependence of the ma/fission ratio on this parameter. A log-log plot gives a fairly satisfactory presentation of the correlation between these two variables. Figure 1 shows the Smallboy data treated in this way. The y-axis represents the ma/fission ratio and the x-axis is the $r_{89,95}$ ratio. In spite of a disconcerting scatter of points throughout the range, it seems clear that there is a definite trend of the ma/fission ratio with the degree of fractionation. The points inside the dotted rectangular box have been fitted to the line drawn through them. The coefficient of correlation for this fit is 0.93. The points outside the box exceed

Figure 1

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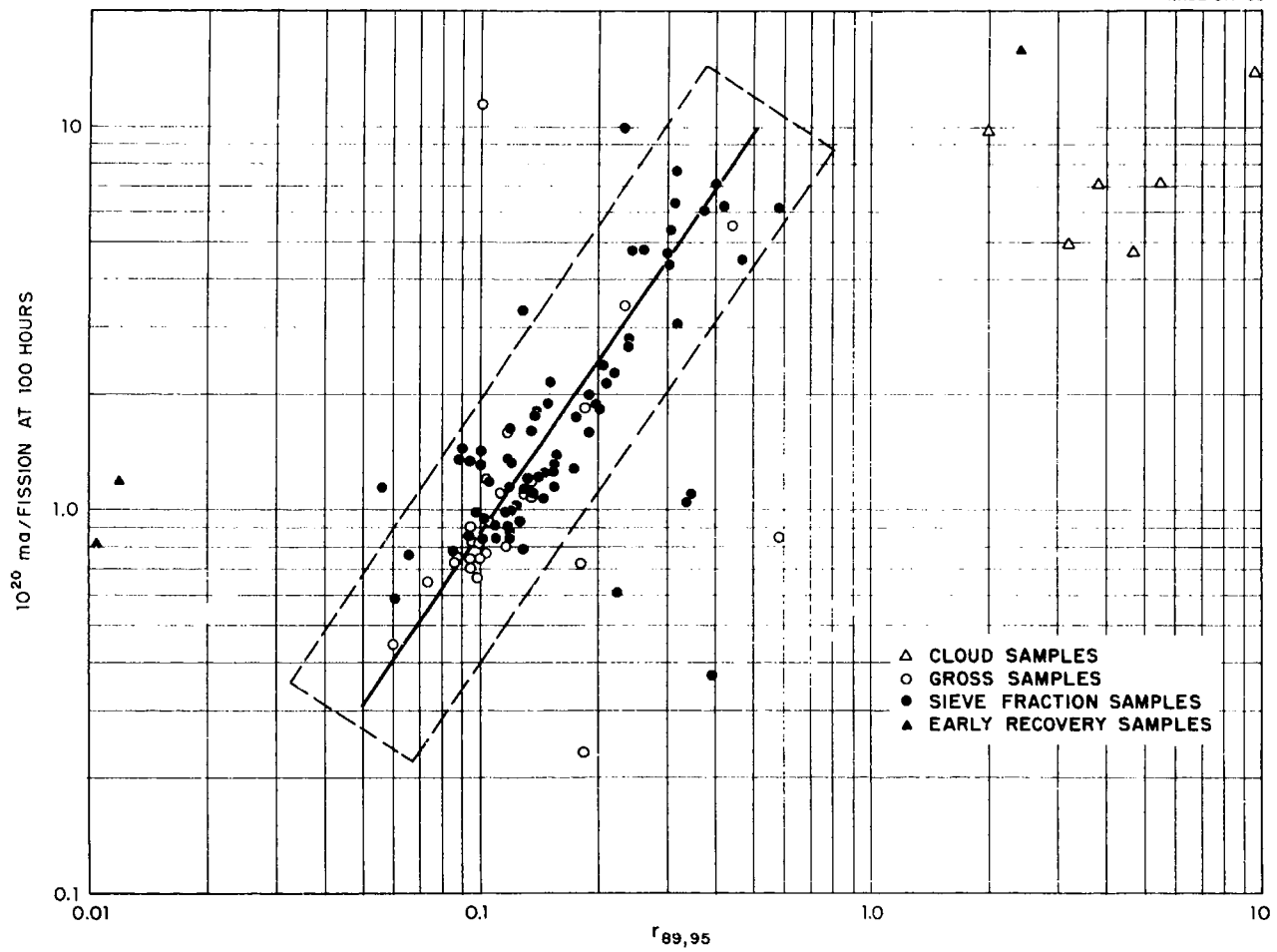


Fig. 1 Variation of the ma/Fission Ratio for Smallboy Samples With Degree of Fractionation, $r_{89,95}$. The points inside the dashed rectangle were fitted to the line by linear regression.

the standard error of estimate, with respect to the linear correlation. Note that the cloud samples do not fall on the line, nor do the early recovery (ER) samples. These latter resulted from separating a gross sample according to particle type. The Johnny Boy data are presented in the same way in Fig. 2 and show the same behavior as the Smallboy data, although the linear correlation indicated by the dashed line is less pronounced. However, the two sets of data are not compatible; i.e., although the ma/fission range for Johnny Boy coincides with the mid-portion of the range for Smallboy, the $r_{80,95}$ values for Johnny Boy are mostly much lower than those for Smallboy.

2. Sample weight and specific activity. The weights and specific activities of the Smallboy samples ranged over factors of 10^3 and 10^4 , respectively. It seemed possible that these wide variations might have introduced errors of geometry and self-absorption into the ionization-chamber measurement which were reflected in the ma/fission ratios. A plot of ma/fission ratio versus sample weights showed no discernible relationship; the very wide scatter of the points appeared to be random. However, the conclusion is less clear-cut in the case of specific activity. Figure 3 is a plot of the ma/fission ratio versus specific activity, expressed as Zr^{95} equivalent fissions per gram, on log-log paper. There appears to be some tendency toward high values of the ratio for low specific activities, but the relationship is far from definite. The dotted line in the figure was obtained by linear regression of the data. The slope is 0.14 and the coefficient of correlation is 0.42. The latter figure indicates that only 18 % of the variance can be accounted for by the correlation line. In view of the fact that most of the higher values of the ma/fission ratio are for sieve-fraction samples, this 18 % may simply reflect the tendency of the larger particle-size fraction of local fallout to be highly fractionated but contain relatively little of the activity.

3. Time of Ionization-chamber measurement. These measurements were made over a time span from one to ten days. Figure 4 shows the scatter of the ma/fission ratios (corrected to 100 hr) versus the various times of measurement. The data fall into two main groups; those in the neighborhood of 110 hours and those in the neighborhood of 230 hours. The spread of the values for the later times seems to be smaller than that at the earlier times. The probable explanation of this is discussed in Section III.3.4.

3.3.3 Effect of Fractionation on the Ionization-Chamber Measurements. It is certainly to be expected that depletion or enrichment of fallout in the volatile mass-chains, relative to Zr^{95} , will affect the amount of ionization current observed per Zr^{95} equivalent fission. Many of the radionuclides in the volatile mass-chains are fairly hard gamma-emitters, and thus must make important contributions

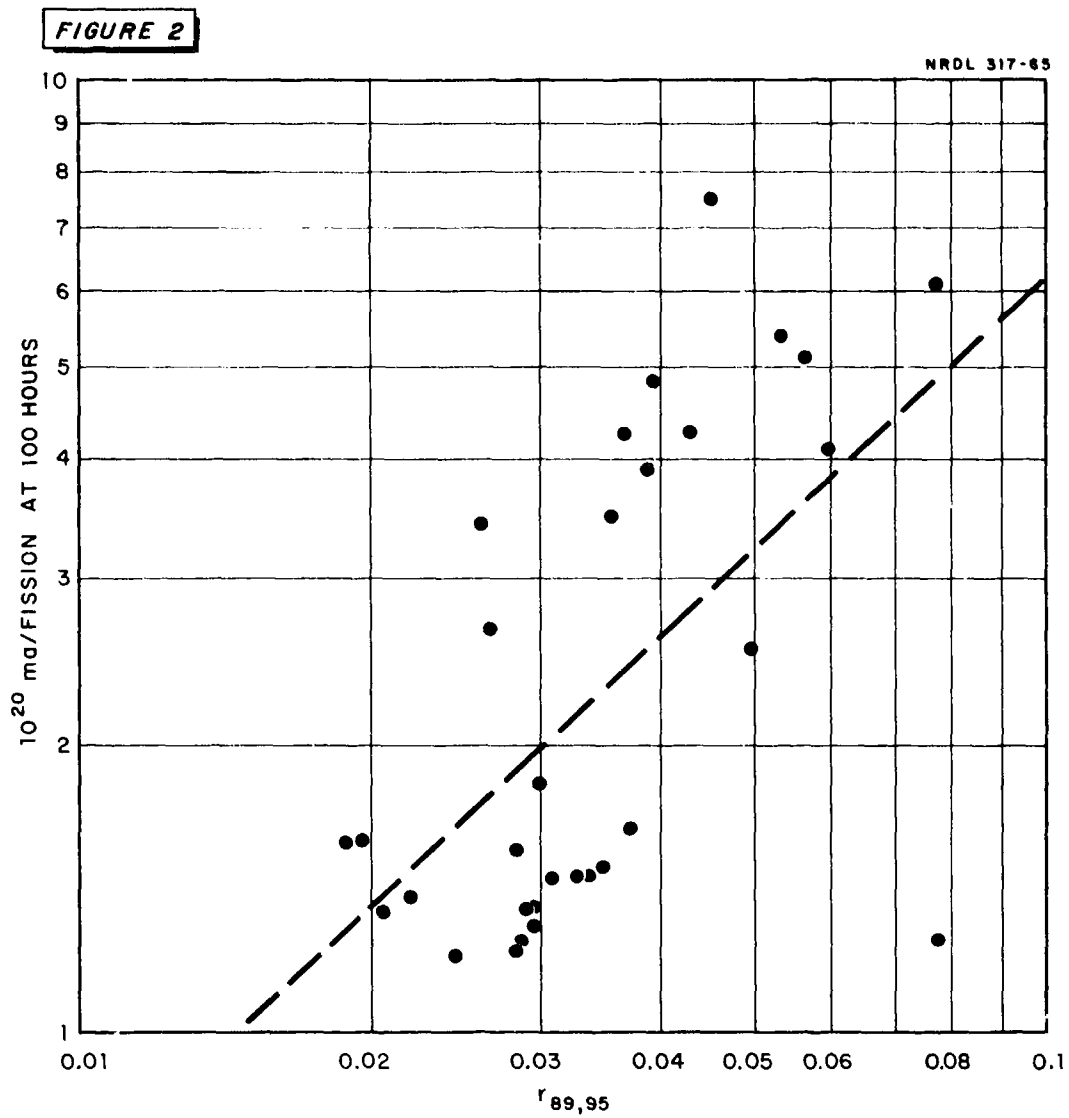


Fig. 2 Variation of the ma/Fission Ratio for Johnny Boy Samples with Degree of Fractionation, $r_{89,95}$. The points were fitted to the dashed line by linear regression. The coefficient of correlation was 0.63. All of these samples were sieve fractions.

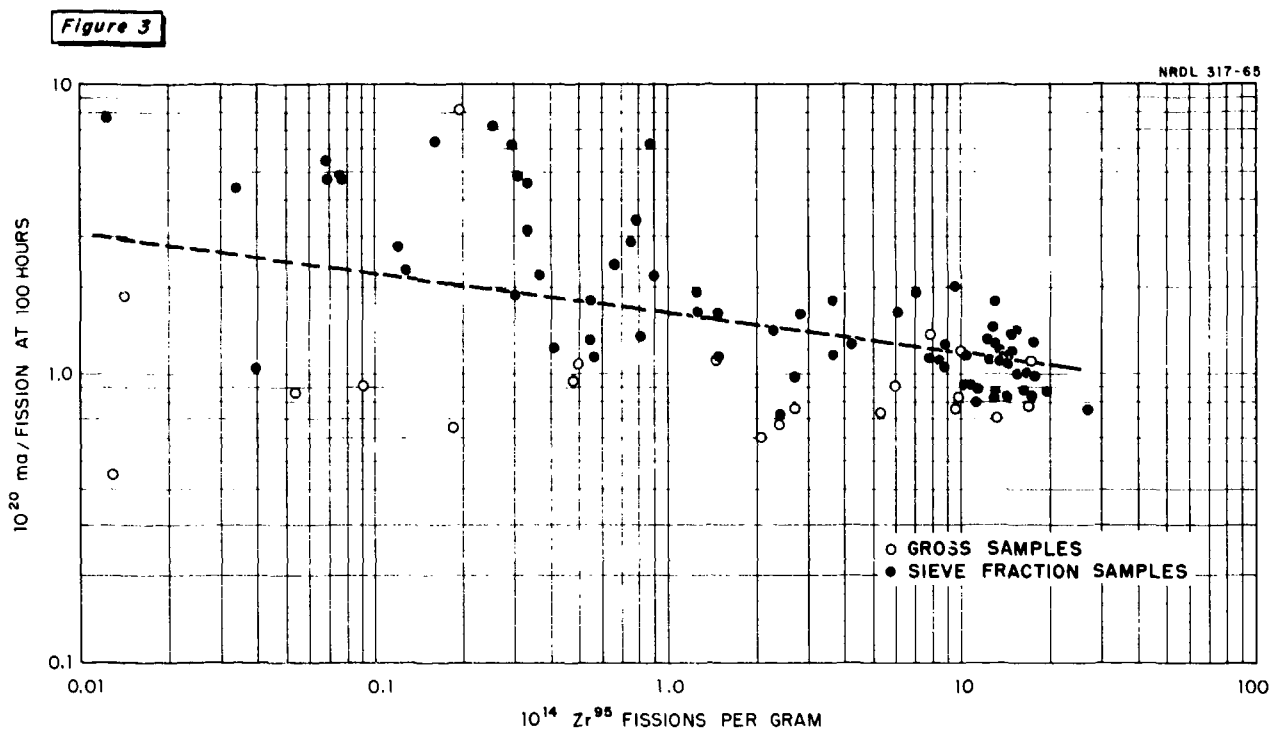


Fig. 3 The $ma/Fission$ Ratio for Smallboy Samples Versus the Specific Activity of the Samples. The points fit the dashed line with a coefficient of correlation of 0.42.

to the ionization current. It is worthwhile to examine the theoretically calculated values of these contributions in some detail, in order to decide whether the variation of the Smallboy ma/fission ratios can be explained as a fractionation effect. If attention is restricted to the points within the dashed rectangle of Fig. 1, a factor of at least 10 needs to be accounted for.

Table 4 is an analysis of calculated contributions to the ionization current by fission products from the thermal-neutron fission of Pu^{239} at 3 days after fission.* The first column lists all nuclides whose activities amount to 1 % or more of the total activity. These activities, given in the second column, are taken from recent unpublished computer calculations at NRDL. The multipliers in the third column convert the activities into the ionization current contributions shown in the fourth column. The multipliers have been calculated from the appropriate information on gamma-photon energies and abundances, along with the energy response curve of the ionization chamber. Note that a relatively short list of nuclides contributes most of the current; in fact, I^{132} alone accounts for nearly one-third of it. Most of the important contributors (the iodine and tellurium isotopes, and La^{140}) are subject to fractionation ranging from moderate to severe.

The importance of I^{132} in this list renders it impossible to make realistic estimates of the ma/fission ratio for the Smallboy samples on the basis of the radiochemical data, since analyses for I^{132} were not made. It is clear from the Te^{132} data available on some of the samples that the mass-132 chain fractionated severely. However, since evaporation of iodine from solid surfaces may continue for several days,^{19,20} the tellurium data are probably not representative of the iodine. This effect applies especially to the samples which were strongly enriched in Te^{132} .

A further complexity in estimating the ma/fission ratio which should have been observed is illustrated by Table 5 (also taken from unpublished calculations at NRDL). The table shows the principal contributions to the ionization current for unfractionated fission products from thermal neutron fission of U^{235} during the period from 2 to 10 days after detonation. This is the period during which the Smallboy ionization-chamber measurements were made. The necessary computer calculations for preparing such a table for Pu^{239} fission products are not available, but examination of activity lists indicates that only minor differences from U^{235} should be expected. The table shows clearly that significant changes occur in the list of principal contributors during the period in question. This means that an

*It should not be inferred from this either that the Smallboy device was a plutonium device or that the neutrons causing fission were thermal neutrons. All that is indicated is that the data for the thermal-neutron fission of Pu^{239} reasonably approximate the case at hand.

TABLE 4

Ionization-current Contributions, Pu²³⁹ Thermal-neutron Fission - 3 Days

Nuclide	Activity (10 ⁻³ dps per 10 ⁴ fissions)	Multiplier (10 ⁻¹⁴ ma per dps)	Ionization Current (10 ⁻¹⁷ ma per 10 ⁴ fissions)
Zr ⁹⁷	0.290	0.419	0.122
Nb ^{97m}	0.279	6.37	1.89
Nb ⁹⁷	0.313	6.15	1.93
Mo ⁹⁹	0.833	1.10	0.916
Tc ^{99m}	0.796	1.16	0.923
Ru ¹⁰³	0.107	4.53	0.483
Rh ^{103m}	0.107	-	-
Rh ¹⁰⁵	0.793	0.320	0.254
Te ^{131m}	0.102	16.3	1.663
I ¹³¹	0.283	4.39	1.242
Te ¹³²	0.639	2.40	1.53
I ¹³²	0.659	19.3	12.7
I ¹³³	0.589	5.99	3.53
Xe ¹³³	0.719	0.336	0.242
Xe ¹³⁵	0.181	2.92	0.526
Ba ¹⁴⁰	0.293	1.84	0.539
La ¹⁴⁰	0.236	21.3	5.03
Ce ¹⁴¹	0.122	0.934	0.114
Ce ¹⁴³	0.589	5.93	3.49
Pr ¹⁴³	0.187	-	-
Nd ¹⁴⁷	0.124	1.47	0.183
Pm ¹⁴⁹	0.193	0.056	0.011
Pm ¹⁵¹	0.094	1.70	0.160
	8.53		37.495

(Total Activity: 9.20×10^{-3} dps/10⁴ fissions)

TABLE 5

Principal Ionization-Current Contributors, Thermal-Neutron Fission,
of U^{235} , 2 to 10 Days

Time After Detonation (days)	Contribution (%)				
	I^{132}	I^{133}	Ce^{143}	$Nb^{97}-Nb^{97m}$	$Ba^{140}-La^{140}$
2	21.0	12.0	12.0	17.0	8.6
3	28.5	8.7	12.2	11.0	16.3
4	31.0	6.7	9.9	6.2	24.3
5	31.2	2.1	7.5	2.3	31.2
6	30.2	-	5.5	-	37.2
7	28.9	-	3.9	-	42.1
8	26.7	-	2.7	-	46.5
9	24.3	-	1.9	-	49.2
10	21.3	-	1.3	-	53.1

ionization-chamber measurement made on a fractionated sample at 50 hr and decayed forward by the $t^{-1.2}$ rule to 100 hr may differ considerably from a measurement made at 220 hr and decayed back to 100 hr.

It is interesting to note that the trend toward dominance of the ionization current by $Ba^{140}-La^{140}$, illustrated by Table 5, continues and reaches a maximum at about 20 days. At this time, nearly 70 % of the current predicted for unfractionated fission products is due to the mass-140 chain. At later times, the influence of $Zr^{95}-Nb^{95}$ becomes predominant; this pair accounts for about 90 % of the current from unfractionated fission products at 250 days.

Although sufficient radiochemical data for exact calculation are lacking on the Smallboy samples, it is easy to assign arbitrary values within the fractionation range suggested by the observed values, and thus make reasonable estimates of the fractionation effect on the $ma/fission$ ratio. For example, one might choose (1) a sample containing only 20 % of the unfractionated amounts of the 132, 133 and 140 chains but otherwise unfractionated, and (2) a sample enriched by a factor of 100 % in the 132 and 140 chains but having the unfractionated composition in all other nuclides. For such a pair it can be shown, by the aid of Table 5, that the $ma/fission$ ratios would differ by factors of from 2 to 4.5, depending on the time at which the ionization-chamber measurements were made. In order to account for the factor of 10

actually observed in the Smallboy ma/fission ratio, one must assume more extreme cases of fractionation. The calculation then becomes definitely speculative, in view of the absence of radiochemical data on I^{132} and I^{133} .

For event Johny Boy, the ma/fission ratio varied by a factor of about 4. Again, considerable variation can be accounted for by fractionation; but for these samples a factor of 4 seems excessive. Since all of the Johny Boy samples were depleted, the range of the ratio is somewhat restricted, compared to the Smallboy samples. Also, the ionization-chamber measurements were all made within a few hours of each other, (from 2 to 2.4 days) so that the time-of-measurement effect does not come into play. The lack of I^{132} and I^{133} data again precludes the possibility of making good theoretical estimates of the ionization current. Unless these nuclides behaved very erratically, it does not appear that the ma/fission ratio should have shown a range larger than a factor of 2. A possible effect in Johny Boy, which was missing in Smallboy, is the effect of induced Na^{24} activity. The unusual abundance of this nuclide in Johny Boy fallout is remarked on in Reference 14, although no quantitative data are available. Since the nuclide is a very hard gamma-emitter, sufficient quantities of it would have strongly affected the ionization-chamber readings. This might also explain the previously noted incompatibility of the Johny Boy and Smallboy ma/fission data.

3.3.4 Other Factors Influencing the ma/Fission Ratio. It is difficult to suggest factors, other than fractionation, which might produce variations in the ratio of an order of magnitude, especially in view of what Fig. 1 shows. It is not believed that induced activities were present in sufficient quantities in Smallboy samples to affect the values by more than a few percent. Similarly, the effects on the ionization-chamber measurements of geometry and self-absorption factors, due to variations in sample weight, should be small. The slight indication of a trend with specific activity, noted in Fig. 3, may result from a certain tendency for low specific activities to be associated with the highly fractionated samples. Figure 4 can be more confidently explained in terms of fractionation; the group of samples measured in the neighborhood of 110 hr included many sieve fractions which were enriched in fractionating nuclides, while those measured at around 230 hr were gross samples which were depleted in these nuclides. This was simply an accidental result of the sample-processing schedule.

A consideration which cannot be ignored is that the radiochemical data may not always have been representative of the sample. In many cases, only a portion of the sample was used for analysis. Due to the essential heterogeneity of fallout material, there is no assurance that the portion chosen was always representative of the radiochemical constitution of the sample taken as a whole.

FIGURE 4

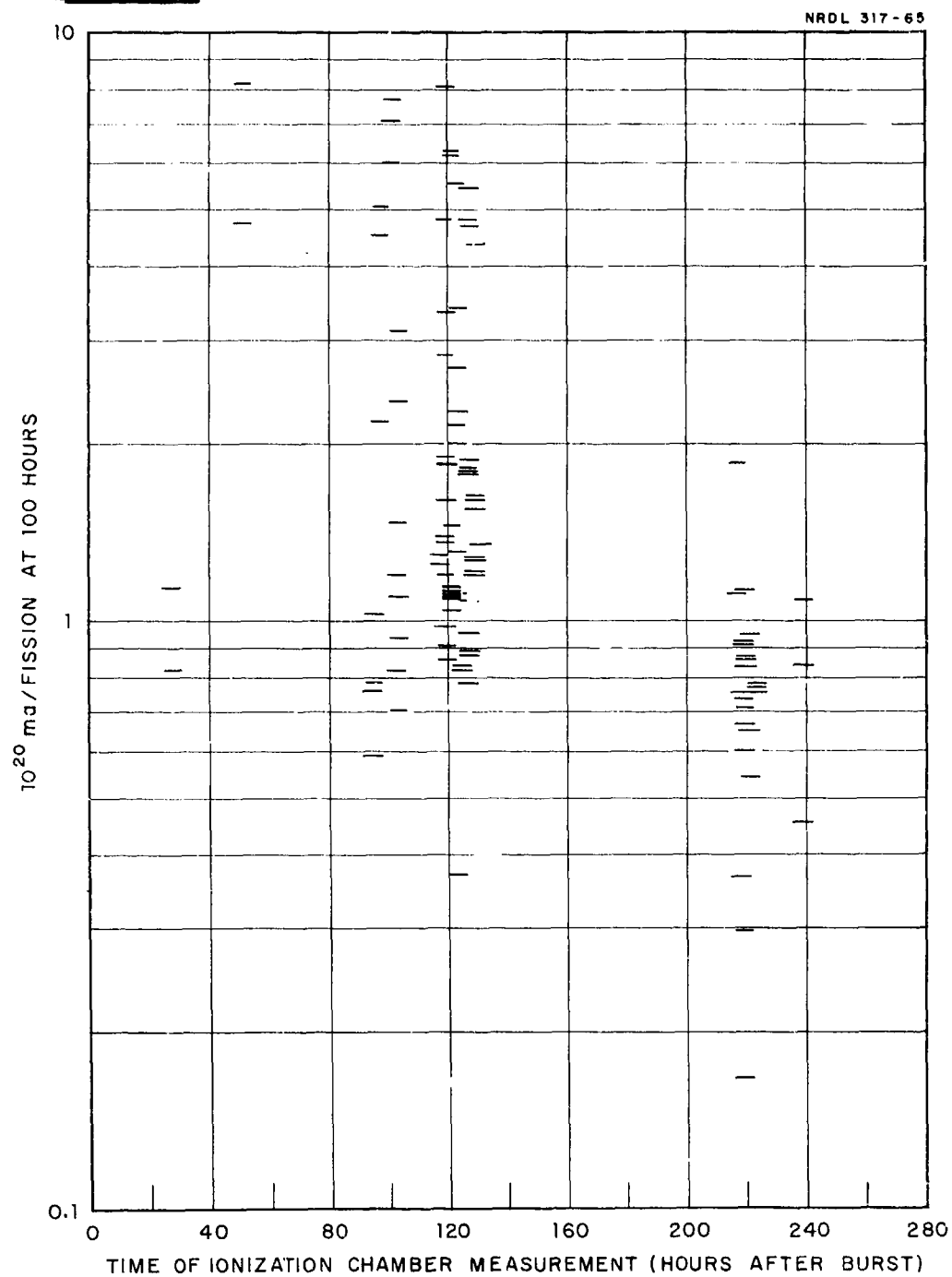


Fig. 4 The $\text{m}\alpha/\text{Fission}$ Ratio (corrected to 100 hr) for Smallboy Samples Versus the Time at Which the Ionization-chamber Measurement was made. The points in the neighborhood of 100 hours include many sieve-fraction samples, while those around 230 hours are for gross samples.

IV. FIELD EXPOSURE RATE MEASUREMENTS

4.1 At the Smallboy and Johnny Boy events the exposure rates in the field were monitored continuously for many hours after the events by USNRDL Gamma Intensity-Time Recorders (GITR's) installed near each collecting station. The GITR's are ionization-chamber devices and are described in Reference 21. Details of installation and operation as well as the results of the measurements at event Smallboy are reported in Reference 13. The measurements made at event Johnny Boy are reported in Reference 14.

4.2 Treatment of Data

It has become customary to reduce field exposure-rate measurements to their values at one hour after burst, for the purpose of standardization. Since there are a number of objections to this general practice,* it has not been adhered to in this study. The object here was to compare the radiochemical data on the deposited fallout with the recorded field exposure rates. Much of the GITR data at one hour for these events needs an appreciable correction for transit-exposure rate. As discussed in Reference 13, there is no really satisfactory method for making this correction with the data at hand. Accordingly, we have preferred to deal with the GITR exposure rates recorded at 10 hours, since these are essentially free of transit-exposure contributions.

The installation of the GITR's in the field is designed to produce an exposure-rate measurement which should be directly applicable to a determination of the normalization factor for the fallout field. A uniform contamination density is, of course, not attainable in the field. Nonetheless, when appropriate radiochemical data on the fallout deposited at the GITR stations is available, it should be possible to use the GITR

*These include the uncertainty involved in decaying measurements made at re-entry time back to 1 hour and the difficulty in correcting recorded GITR data for transit exposure rate. The exposure rates are invariably too high to permit movement of personnel into the field at 1 hour. The close-in fallout from high yield events and the longer range (greater than about 60 miles) fallout from lower yields may not all be deposited at 1 hour.

exposure rates to calculate a reasonably good value of the normalization factor if the deposit gradient is not too steep.

A slight complication arises from the fact that the Smallboy debris was fractionated. The quantity "kt per square mile" is, strictly speaking, undefined in such a case. In order to specify the contamination density of fractionated debris, reference must be made to some specific fission-product radionuclide. A convenient choice for a reference nuclide is Zr^{95} . We will retain the "kt per square mile" terminology by agreeing to call 1.45×10^{23} equivalent fissions of Zr^{95} a kt, and will calculate the denominator of the normalization factor, kt/mi^2 , from the number of Zr^{95} equivalent fissions deposited in a $4-ft^2$ collecting tray. With this convention, Zr^{95} and those nuclides which did not fractionate from Zr^{95} are present to the same extent as they would be had no fractionation occurred. Nuclides which fractionate from Zr^{95} , such as Sr^{89} and Cs^{137} , may, in general, be either depleted or enriched as discussed in Section IV.3.1. As long as the contamination has uniform composition and uniform distribution, the r/hr per 1.45×10^{23} equivalent fissions of Zr^{95} should be constant. It would not, however, be equal to the value calculated for unfractionated debris.

It appears, from examining the data in Reference 12, that the contamination in the Smallboy local fallout field was fairly uniform, insofar as one might expect to encounter this condition in the field. This can be verified by entering the proper $r_{89,95}$ values for the gross samples on a station layout of the field. The ratio varies from about 0.05 to about 0.25, but the majority of the stations lie within a considerably narrower range. No trend of the ratio with distance or direction appears to be present, except for a tendency toward low values at stations on the perimeter of the field. It is to be emphasized that the range of sample fractionation being dealt with in this section of this report is much narrower than that discussed in the preceding section, since only gross samples are being considered here. Measurements on sieved samples and cloud samples are not appropriate for comparison with exposure rates measured in the field.

4.3 Results

The results of the calculation of normalization factors from the Smallboy data are shown in column 4 of Table 6. (Discussion of columns 5 and 6 is deferred to Part V.) If one eliminates some of the extreme values for the reasons given in the footnotes to the table, the remaining values give an average value for the normalization factor of 26.4 r/hr at 10 hr per kt/mi^2 . The individual values range from 10.1 to 48.9. The most striking thing about the values is that not only the average but all of the individual values are far lower than the theoretical prediction for unfractionated debris. The predicted value for

this quantity at 10 hours is about 150 r/hr per kt/mi². (Predicted values for the various modes of fission in Table 1 show only small differences.) This is about 5.6 times the average value calculated for Table 6.

Similar calculations of the normalization factor using the Johny Boy data do not clarify the situation. Here only 5 stations have complete data. One station was very near the hot line while the other four were 1000 to 1500 feet distant. The value of the normalization factor for the hot line station is about one-half the theoretical value, while those for the remaining stations are many times too high. The Johny Boy fallout pattern was unique and did not at all approximate a uniformly contaminated field. Almost all the activity was concentrated in a narrow band along the hot line. There is evidence that the radiation from this band influenced GITTER's at a considerable distance. For these reasons the Johny Boy normalization factors will not be considered further in this report.

Further data of interest in connection with calculations of the normalization factor have recently appeared in a report by Miskel and Bonner on event Danny Boy.¹⁵ Table 1 of the report gives normalization factors, J_1 , based on a number of different nuclides and calculated as the ratio of disintegrations per minute per square foot to exposure rate in mr/hr. These are easily convertible to r/hr per kt/mi². Values are listed for each of nine different stations and are based on exposure rates measured at 29.75 hours. The factors based on Zr⁹⁵ are of principal interest here. These vary from 0.68 to 2.9×10^4 dpm/ft² per mr/hr, if one disregards the anomalous value for station Y-26. Note that this is comparable to the variation of the Smallboy factors in Table 6. The average value for the nine stations, when recalculated, is 50.5 r/hr per kt/mi². This compares with a prediction of 51.4 for unfractionated debris from the Danny Boy event. This agreement is anomalous, as will be discussed below, since the fallout from this event was highly fractionated.

4.3.1 Effect of Fractionation on the Normalization Factor.

In view of the pronounced effect of fractionation on the ma/fission ratios, as evidenced by the data of the preceding section, one would expect the field exposure rates (and therefore the normalization factors) to be similarly affected. In discussing the exposure rates, it should be borne in mind that they differ from the ionization-chamber measurements in two respects: (1) the exposure-rate measurements were made at an earlier time (10 hours), and (2) the range of fractionation of the samples is much smaller, for reasons given above.

Table 7 lists the principal contributions to the exposure rate at 10 hours from fission products of the thermal-neutron

TABLE 6

Calculation of Normalization Factor for Smallboy

Station Number	Zr ⁹⁵ (fissions per sq. ft.)	Exposure Rate (r/hr at 10 hr)	Normalization Factor (r/hr at 10 hr per kt/mi ²)	ma/fission at 100 hr $\times 10^{20}$	Ratio of Column 4 to Column 5
S1S-PC1 & 2	1.95×10^{14}	1.1	29.3	1.35	22
101-AO-6	2.14×10^{15}	11.0	26.7	1.17	23
103-AO-9	2.88×10^{12}	0.07	126 ^a	0.054	(2300)
200-AO-9	1.39×10^{13}	0.08	30.0	1.09	28
200-AO-10	1.03×10^{15}	5.0	25.2	1.28	20
202-AO-9	2.24×10^{11}	1.77×10^{-3}	41.1	0.46	(89)
209-AO-9	3.50×10^{11}	1.61×10^{-4}	2.4 ^b	0.17	14
S2-PC16 & 21	2.48×10^{14}	1.27	26.6	1.25	21
303-AO-9	9.23×10^{13}	4.18	236 ^b	11.4	21
305-AO-4 & 9	5.68×10^{14}	2.59	23.7	1.00	24
S3-PC16	3.20×10^{10}	5.97×10^{-4}	73.6 ^c	0.56	(130)
400-AO-9	9.23×10^{12}	3.54×10^{-2}	20.0	0.60	33
401-AO-9	8.90×10^{14}	2.66	15.5	0.71	22
403-AO-4	6.83×10^{14}	4.00	30.4	1.19	26
405-AO-9	4.43×10^{14}	1.07	12.6	0.75	17
407-AO-9	2.06×10^{12}	1.30×10^{-2}	32.8	0.91	36
501-AO-9	7.23×10^{12}	6.80×10^{-2}	48.9	0.66	(74)
503-AO-9	2.70×10^{14}	1.0	19.2	0.83	23
505-AO-6	3.15×10^{14}	1.92	31.7	1.30	24
S5-PC1 & 5	1.05×10^{14}	0.814	40.3	1.64	25
701-AO-9	3.23×10^{11}	4.14×10^{-3}	76.6 ^d	0.65	(118)
703-AO-9	2.80×10^{13}	6.20×10^{-2}	11.5	0.73	16
707-AO-3	6.50×10^{13}	0.126	10.1	1.26	(8)

a. Zr⁹⁵ value believed in error, also probable error in ma reading.

b. An error by a factor of 10 in Zr⁹⁵ value suspected.

c. GTR curve for this station is erratic, suggesting instrument malfunction.

d. Sample weight probably not reliable.

TABLE 7

Principal Exposure-Rate Contributors at 10 Hours, Thermal-Neutron
Fission of Pu²³⁹

Nuclide	Contribution (percent)	Nuclide	Contribution (percent)
I ¹³⁵	33.0	Sr ⁹²	3.8
Ru ¹⁰⁵	7.0	Xe ¹³⁵	3.5
I ¹³³	5.4	Sb ¹²⁹	3.4
Nb ⁹⁷ -Nb ^{97m}	10.1	Sr ⁹¹	2.8
I ¹³²	5.0	Ce ¹⁴³	2.5
Pr ¹⁴⁵	4.7	Kr ⁸⁸	2.5

fission of Pu²³⁹. Note that these contributors are very different from those listed in Table 5 for the time interval from 2 to 10 days. The present list is dominated by I¹³⁵. This nuclide almost certainly fractionates rather severely, but the extent of its fractionation is not known. In fact, the only nuclide on this list for which radiochemical data is available on the Smallboy samples is Sr⁹¹, which contributes only 2.5 % to the 10-hour exposure rate.

In spite of the absence of the desired radiochemical data, it is possible to use Table 7 to make a crude estimate of the effect of fractionation on the 10-hour exposure rate for Smallboy. Gross samples from all stations in the field were depleted in Sr⁸⁹ and Cs¹³⁷. The value of r_{89,95}, as mentioned earlier, was roughly the same throughout the field. For purposes of estimation, we will assume that this ratio was constant at all points and equal to 0.2. Three of the mass chains in Table 7 - mass 97, 143, and 145 - can be assumed with some confidence not to have fractionated from Zr⁹⁵. These chains account for 17.3 % of the unfractionated exposure rate. Now assume that all the remaining mass chains fractionated and that, on the average, they fractionated to the same extent as the mass-89 chain. This means that only 0.2 of their contribution to the unfractionated exposure rate (82.7 %) will remain. When this figure is added to the contribution from the three unfractionated mass chains, the result suggests that the fractionated exposure rate should have been 33.8 % of the unfractionated exposure rate.

The average value of the normalization factor for Smallboy, as calculated in Table 6, is only 17.7 % of the theoretical unfractionated normalization factor. To account for a reduction of this

magnitude, it would be necessary to assume that the fractionated mass chains were completely absent from the Smallboy fallout. This was certainly not the case. It appears therefore that some other factor was operating to reduce the exposure rates measured in the field.

The fallout from the Danny Boy event was strongly fractionated and in the reverse sense from the Smallboy event; i.e., the fallout was much enriched in volatile mass chains.* The evidence for this fractionation is presented in Miskel and Bonner's Table 3.¹⁵ Accordingly, the observed normalization factor should have been much larger than that predicted for unfractionated debris. It is difficult to estimate how large this effect should have been. The enrichment factors in Table 3 of Reference 15 run as high as 12, with values very commonly in the range from 2 to 7. The enhancement of the exposure rate due to this enrichment is not apparent from the Danny Boy average normalization factor, which agrees well with the value calculated for unfractionated fallout. One can only conclude that some other factor or factors, operating to reduce the field exposure rates, almost exactly counterbalanced the fractionation effect.

4.3.2 Effect of Ground Roughness. A possible explanation of the reduced field exposure rates in both Smallboy and Danny Boy lies in the ground roughness effect and the shielding due to the detector itself. The radiation-attenuating effect of irregularities in the ground surface is known to be appreciable, but there is no agreement as to its magnitude. A reduction of 20 % to 50 % is often suggested, but much larger reduction factors have sometimes been suggested.²² If one applies a 50 % reduction to account for both ground roughness and shielding by the detector to the Smallboy normalization factor as estimated from Table 7, the result is well in line with the observed value as calculated from the field data. Supposing the same factor to apply to Danny Boy, one finds an "average" enrichment factor for all fractionating nuclides of about 1.7. (This assumes that the exposure-rate contributions at 2 days from the Danny Boy device are about like the 2-day ionization-chamber contributions given in Table 5.) Such a factor is certainly not out of reason, insofar as one can judge from the comparatively scanty radiochemical data on the Danny Boy samples.

4.3.3 Other Possible Effects on the Normalization Factor. The treatment of the data in this report has assumed that the GTR's give a good measurement of the exposure rate at a point 3 feet above an infinite,

*It might be more appropriate to regard this debris as being depleted in refractory mass chains. This kind of fractionation seems to be typical of local fallout from cratering detonations. Apparently the refractory mass chains condense so quickly that a large proportion of their activity is returned to the crater with the fall-back. However, for consistency in the discussion, it is better to continue to regard Zr^{95} as the reference substance and to speak of enrichment or depletion of the volatile mass chains.

contaminated plane. This may not be true. A careful review of GTR installation, operation, and calibration is in order, possibly in conjunction with some experimental measurements under controlled conditions. Another possibility is that the theoretical apparatus for predicting infinite plane exposure rates may be inadequate for the purpose at hand. Until recently, all such predictions relied ultimately on the air "build-up" factors of Gates and Eisenhauer.² These factors, which are based on a solution of the radiation transport equation by the method of moments, were computed by a very complicated machine method which necessarily involved certain approximations. However, it is regarded as rather unlikely that inherent uncertainties in these factors could account for any large discrepancies between theoretical and observed exposure rates. This has now been confirmed by the Monte Carlo calculations of R. L. French²³ which give good agreement with the Gates and Eisenhauer factors.

4.3.4 The Area Integral of the Exposure-Rate Contours and the Normalization Factor. If the iso-exposure rate contours of a fallout pattern are integrated over the area of the fallout field, by some such procedure as those described in References 24, 25 or 26, and the integral divided by the device yield, the resulting ratio has the same units as the normalization factor. The value of this ratio is often compared to a theoretical value of the normalization factor for unfractionated debris (after making some adjustment for instrument and terrain shielding), in an effort to determine the partition of the fission products between local and world-wide fallout. The "escape fraction" thus determined can only be applied, at best, to the whole collection of fission products, since the partition fractions of individual nuclides will often differ by an order of magnitude. Even when restricted to its very limited proper meaning, the "escape fraction" determined from the 1-hr exposure rate contours can be misleading. The 1-hr exposure rates are almost entirely contributed by a handful of nuclides whose fractionation behavior is not documented at all. There is no good reason to suppose that the effect of fractionation on the 1-hr exposure rates is representative of any sort of "average" fractionation effect at times appreciably later. Thus, the 1-hr "escape fraction" is not of much use in predicting the radiological properties of the debris. In particular, it cannot yield any useful information about the state of fractionation of such biologically important radionuclides as I^{131} , Sr^{90} , and Cs^{137} , since none of these (or their mass chains) make any appreciable contribution to the exposure rate until very much later times.

If some radiochemical data on the fallout is available, and if the contamination in the area over which the exposure rates are integrated is known to have been reasonably uniform, the integral of

the iso-exposure contours can be interpreted with considerably more confidence.* A normalization factor can be calculated which is based on the actual field contamination level (in equivalent fissions) of some particular nuclide, as given by the radiochemical data. Since the factor from iso-exposure contour integration is based on the total number of fissions produced by the device, a comparison of the two factors will give a good idea of what fraction of the nuclide in question is present in the area over which the integration was carried out.

The normalization factor for Smallboy at 10 hours, according to the preceding section is 26.4 r/hr at 1 hr per kt/mi², based on the contamination level of Zr⁹⁵ in the field. The value derived from integration of 1-hr exposure-rate contours is reported by Bouton²⁵ to be 491 r/hr at 1 hr per kt/mi². LaRiviere and co-workers²⁶ report it to lie in the range of 560 to 1200 r/hr at 1 hr per kt/mi². It appears that both values refer to the contour pattern as constructed by Bouton. LaRiviere et. al., remark on the disagreement and suggest possible explanations; they also offer some important observations on the problems encountered in integrating exposure-rate patterns.

In order to make a valid comparison, some means must be devised to put the 10-hour value and the 1-hour values on the same time basis. The simplest procedure is to assume that the Smallboy fallout decayed like unfractionated fallout; the ratio of 1-hour and 10-hour values will then be given by the appropriate entries in Table 1. Bouton's 1-hr exposure-rate pattern is, in fact, based on survey measurements made at times ranging from 1 hour to 6 days after the event. These were all extrapolated to 1 hour by rather complicated methods. Since there is no satisfactory way to adjust his original data to 10-hour values, we choose to adjust our 10-hr field normalization factor to 1 hour by the use of Table 1. This yields a value of about 480 r/hr per kt/mi². The close comparability of this value with Bouton's value and with the lower limit assigned by LaRiviere et al. strongly suggests that most of the Zr⁹⁵ produced by the device came down in the local area. One must allow a few percent in the long range fallout cloud, of course, since Zr⁹⁵ was still detectable in Smallboy fallout at great distances.²⁷ However, it seems quite possible that as much as 90 % of the Zr⁹⁵ and the refractory nuclides which did not fractionate from it, like Mo⁹⁹ and Ce¹⁴⁴, were brought down in the local area. Since $r_{89,95}$ and $r_{137,95}$ lie someplace between 0.1 and 0.2 for the local field, it follows that only 10 to 20 percent of the Sr⁸⁹ and Cs¹³⁷ produced by the burst were deposited in the local fallout.

This method of calculating the partition of fission products between local and world-wide fallout is subject to several

*The objections offered in Section IV.2 of this report to 1-hr exposure rates, as opposed to exposure rates referring to later times, remain valid.

reservations, based largely on deficiencies in the data, but in the absence of better methods it yields important indications as to magnitude and direction. Russell* has arrived at reasonably similar values of the partition of these nuclides in Smallboy by a mass balance method applied to the radiochemical data on the cloud samples and the ground-collected samples. Fairly large uncertainties must also be admitted in his estimates, on account of the insufficiency of the cloud sampling.

*Col. I. J. Russell, Kirtland AFB, Albuquerque, New Mexico, private communication.

V. COMPARISON OF MA/FISSION RATIOS WITH NORMALIZATION FACTORS

For purposes of comparison, Table 6 includes the ma/fission ratio from one of the trays at each of the stations for which field normalization factors were calculated. The last column of the table shows the ratio of the normalization factor to the ma/fission ratio. This ratio should be constant, since it is simply the ratio of two ionization-chamber measurements on samples of the same material. This should be approximately true even if the spectral characteristics of the radiation from samples from different stations are not exactly the same, inasmuch as the energy responses of the two detectors should be similar. Some allowance should be made for the differences in scattering effects of the two different geometries employed, but these would probably not be large except in the case of gross differences in spectral characteristics.

If the parenthesized values in the last column of Table 6 are omitted, the remainder are reasonably well clustered around an average value of $(23 \pm 5) \times 10^{20}$. Three of the six parenthesized values are associated with normalization factors which were suspected of error on other grounds, as noted in the footnotes. Errors in the Zr^{95} value cancel out of the ratio of the normalization factor to the ma/fission value. Note that the values of this ratio for stations 209-A0-9 and 303-A0-9 tend to support footnote (b). For station 103-A0-9 an error of a factor of 5 in the Zr^{95} value coupled with an error of 100 in the ionization-chamber reading must be assumed to account for the data, although it might also be accounted for by a steep deposit gradient. (Since reading the ionization-chamber meter requires noting the position of a decade switch, the former kind of error can easily occur.)

The predicted exposure rate at 10 hours for unfractionated fission products of thermal neutron fission of U^{235} is 167 r/hr per kt/mi².* The predicted ionization current at 100 h is 2.7×10^{-20} ma/fission. Allowing 50 % for ground roughness and instrument self-shielding, as before, the calculated ratio of the normalization factor to the ma/fission ratio is 31×10^{20} . This is slightly outside the range of the

*This case is chosen for purpose of illustration and has no implication in regard to the conditions of the Smallboy event.

average value found for Smallboy. The difference may be due to differences among the fractionation of I^{135} , the principal radiation contributor at 10 hours, and of I^{132} , I^{133} and $Ba^{140}-La^{140}$, which account for most of the radiation at 100 hours. One should also bear in mind that the sample collected in a 4 ft² collecting tray was relatively small compared with the contaminated area being surveyed by the GTR. It seems likely that, in some instances, the radiochemical composition of the sample was not completely representative of the fallout field which was influencing the detector.

VI. DISCUSSION

The radiochemical data and field exposure rates from Smallboy, Johnny Boy, and Danny Boy provide evidence of strong effects of radionuclide fractionation on the gamma-radiation characteristics of fallout. Both 4-pi ionization-chamber measurements and field exposure-rate measurements appear to be highly sensitive to these effects. This is due to the fact that many of the radionuclides which are usually observed to fractionate make strong contributions to the total gamma radiation. In view of the magnitude of the effects observed, the following points are suggested for careful consideration in interpreting radiation measurements and predicting radiation hazards from fallout:

6.1 Predictions of field exposure-rate levels must allow for fractionation effects. Predictions based on the ordinarily used normalization factor for unfractionated debris may overestimate the levels for fallout depleted in volatile mass-chains and underestimate those for enriched fallout by large factors.

6.2 The interpretation of 4-pi ionization chamber measurements in terms of "device fissions" or "fraction of device in a sample" gives misleading impressions if fractionation is unaccounted for. Neither of these concepts is useful in defining the contamination level of fractionated debris. The 4-pi ionization chamber remains a very useful instrument for comparing the relative gamma-radiation strengths of samples, so long as the measurements are made at essentially the same time. However, there is no way presently known to deduce from these measurements alone the concentration of any particular fission product, at least within an order of magnitude; nor can they be used, without supplementary information, to estimate enrichment or depletion of any nuclide or nuclides, relative to others. Ionization-chamber measurements can be used to good advantage in checking exposure rate survey measurements and radiochemical results, and to fill some of the inevitable gaps in this kind of field data.

6.3 The practice of integrating exposure rate contours for fallout fields and dividing by a normalization factor to determine "fraction out" or "% vented," which has been applied to cratering events,²⁴ is

misleading. As Miskel and Bonner¹⁵ have pointed out, these concepts are meaningless, as applied to fractionated debris, since the percents of the radionuclides escaping into world-wide fallout may vary by orders of magnitude depending on their fractionation behavior.

6.4 Since the unfractionated fission-product composition cannot be used to predict radiological properties of fractionated fallout with reasonable accuracy, means must be sought to predict the fractionated composition. A scheme for doing this, based upon a knowledge of $^{89,95}\text{Sr}$ for the fallout (or some means of predicting it), has been advanced by Freiling and co-workers.¹⁰ The scheme depends upon previously established correlations between the fractionation behavior of ^{89}Sr , relative to ^{95}Zr , and that of other fractionating nuclides. Unfortunately, radiochemical data on which to base such correlations for ^{132}I , ^{133}I , and ^{135}I do not exist. Moreover, there is no reason to believe that, if it did exist, it would follow a similar pattern. As pointed out in the earlier parts of this report, these radionuclides strongly affect the radiological properties of fallout at times extending to several days after burst. It is, accordingly, of great importance to formulate some method of describing their fractionation behavior.

6.5 The basic concept of a normalization factor for unfractionated fission products remains useful; since when the appropriate fractionation parameters are known, it can be properly adjusted and used to predict the radiological properties of uniformly contaminated fields.

VII. RECOMMENDATIONS

If it should become possible in the future to collect fallout from the testing of surface or cratering explosions of nuclear devices, no opportunities for clarifying aspects of the fractionation phenomena should be overlooked. Some suggestions, based on the study of Smallboy, Johnny Boy, and Danny Boy, follow:

7.1 Sufficient fallout collection and analysis should be performed to define as accurately as practicable the radiochemical composition of the fallout in the local area. The fallout field itself should be monitored continuously with gamma-intensity-time recorders for 1 or 2 days. Any effects of induced activities during this period should be anticipated and methods devised to determine their magnitude.

7.2 As much information as possible concerning the fractionation behavior of the iodine isotopes should be obtained. The half-life of I^{131} is long enough to permit determination on all samples, if desired. Special efforts to expedite sample handling should allow a reasonable number of determinations of I^{133} along with I^{132} and its precursor, Te^{132} . Perhaps methods of analysis in the field can be devised to supplement information on these latter isotopes and to give some indication of the behavior of I^{135} . Special attention should be directed to resolving the question of evaporation of iodine from fallout after its deposition. These studies would be designed to establish whether this process actually occurs to an appreciable extent and, if so, the rate at which it occurs. Iodine solubility studies and subsequent oxidation-state determinations might throw additional light upon the secondary fractionation of iodine.

7.3 Careful and adequate sampling of the fallout cloud is needed to estimate the partition of the fission products between local and world-wide fallout. The radiochemical composition of the cloud varies greatly from top to bottom and changes rapidly during early times after burst. Sampling penetrations at several altitudes on a fairly extensive time schedule are indicated. The Air Force studies of the Johnny Boy cloud can serve as a model for this operation.

7.4 In the event of a nuclear cratering experiment, it is important to gain an accurate idea of the three-way partition of the radionuclides among the fall-back into the crater, the local fallout and the world-wide fallout. The local area and cloud studies outlined above should be supplemented by an examination of the radiochemical contents of the crater itself. Radiation measurements in the crater will yield little information on this point, since most of the gamma radiation from the buried radionuclides will be absorbed by the soil. Core sampling should be conducted to provide adequate definition of a concentration profile of the key radionuclides throughout the crater. It will undoubtedly be necessary to defer such sampling for weeks or even months, to minimize radiation hazards to personnel. Nonetheless, it should still be possible to obtain good analyses for a fairly wide range of fractionating and non-fractionating radionuclides. Depending upon local climatic conditions, weathering effects may become a consideration in this kind of study. The Nevada Test Site conditions, however, are favorable in this regard. The usual conditions in this area are such that weathering effects should not be appreciable except in the top inch or two of the material in the crater.

7.5 It is highly desirable that the disagreements about the value of the normalization factor for unfractionated fallout fields (see Table 1) be resolved. This objective can probably best be accomplished by careful laboratory studies on unfractionated fission products, extending from the earliest practical time after fission to times of a few days. Measurements in a 4-pi ionization chamber on samples of known Zr^{95} content could be used to predict early-time normalization factors on the basis of the proportionality between $ma/fission$ and r/hr per kt/mi^2 derived in this report from the Smallboy data. As pointed out earlier, this ratio should not be very sensitive to changes in spectral characteristics. Nonetheless, it is very desirable that the gamma-spectral characteristics of the sample radiation also be determined at these early times. These data would serve as an independent and more sophisticated method of arriving at the normalization factor. These experimental approaches to the problem of the normalization factor seem preferable to further calculational efforts, since the latter suffer from certain defects of input information which are not likely to be remedied in the near future.

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