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FRACTIONATION IV. ILLUSTRATIVE CALCULATIONS OF THE EFFECT OF RADIONUCLIDE FRACTIONATION ON EXPOSURE-DOSE RATE FROM LOCAL FALLOUT

by
E.C. Freiling
M.A. Kay
J.V. Sanderson

431227

U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY

SAN FRANCISCO · CALIFORNIA · 94135

PHYSICAL CHEMISTRY BRANCH
E.C. Freiling, Head

CHEMICAL TECHNOLOGY DIVISION
L.H. Gevantman, Head

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Eugene P. Cooper
Scientific Director

D.C. Campbell, CAPT USN
Commanding Officer and Director

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ABSTRACT

This report illustrates a proposed definition of surface density of contamination for the deposit of fractionated fallout. The illustrations employed give estimates of the dependence of exposure-dose rate on the degree of radionuclide fractionation and of the sensitivity of this dependence to independent fission yield input data, total yield of the device, and time of exposure.

SUMMARY

Problem

The effects of radionuclide fractionation severely complicate the prediction of many properties of nuclear bomb debris, including the definition of surface density of contamination and the dependence of exposure-dose rate on the degree of fractionation.

Findings

A semi-empirical model can be used to illustrate a new definition of surface density of contamination for fractionated fallout. The model is recommended for rule-of-thumb estimates of the effect of fractionation on exposure-dose rate, and as a stop-gap until either better models or more extensive information becomes available.

PREFACE

This series of reports presents and discusses the effects of radionuclide fractionation in nuclear bomb debris. Part I (Reference 1) defined fractionation as "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." It showed how the radionuclide compositions of fractionated samples could be correlated empirically by logarithmic relations. Part II (Reference 2) used these relations as the basis of a technical discussion of contamination density as applied to fractionated nuclear debris. Part III (Reference 3) presented a theoretical foundation for the observed logarithmic correlations of Part I. It used this as a simplified means of estimating fractionation as a function of particle size and the partition of product radionuclides among local, intermediate, and worldwide fallout.

The present report extends the calculations of Part III to show how fractionation-correlation parameters can be used to estimate the exposure-dose rate from nuclear debris with various degrees of fractionation. It serves to illustrate the proposals made in Part II.

INTRODUCTION

Local fallout from a land-surface nuclear detonation consists of the larger radioactive particles formed in the explosion. Because of radionuclide fractionation, it is depleted in volatilely behaving mass chains (such as the mass-89 chain) relative to refractorily behaving mass chains (such as the mass-95 chain). Worldwide fallout, on the other hand, consists of the smaller particles of debris and is relatively enriched in volatilely behaving mass chains. The effect of fractionation on exposure-dose rate from fallout deposits is to produce a wide variation in the ratio of dose-rate to fission-product abundance, for any given mass chain, or of one fission-product abundance to another, for certain pairs of mass chains.

The radiochemical composition of fallout will depart from the representative composition of debris by a quantity which we have called the degree of fractionation and have defined quantitatively as the base-10 logarithm of the fractionation ratio:

$$\Phi = \log_{10} r_{89,95}$$

The fractionation ratio ($r_{89,95}$) is the ratio of the number of fissions (from the device in question) required to produce the quantity of mass-89 chain found at a given location, to the number required to produce the quantity of mass-95 chain at that location. The relation of our notation to that of other authors is given in Appendix A.

Two theoretical methods exist for estimating Φ in local fallout,[3,4,5,6] and empirical relationships are available for estimating the relative amounts of other mass chains as a function of Φ.[1] By combining either of these theoretical methods with the empirical relations, one obtains a semiempirical means of estimating the radionuclide composition, and hence the resulting exposure-dose rate, as a function of particle size. A fallout-transport model can then be used to calculate these same quantities at any point in the fallout pattern.

The purpose of this report is threefold: (1) to illustrate the empirical estimation of radionuclide composition as a function of ϕ; (2) to estimate the resulting exposure-dose rate as a function of ϕ, (3) to obtain a preliminary estimate of the sensitivity of this relationship to several kinds of input data; and (4) to illustrate the application of a new definition of surface density of contamination.[2] In presenting this material, familiarity with the previous reports in this series is assumed.

The procedure for estimating the radiochemical composition of a fission-product mixture as a function of ϕ is as follows. We consider only those nuclides which, at some time, contribute significantly to the dose rate. These nuclides and their isobars form the significant mass chains. Next, the fission-product elements are divided into refractory and volatile groups, according to their vapor pressures at a chosen temperature. The temperature chosen in this report corresponds roughly to the solidification temperature of molten silicate soil. For any nuclear detonation, the total yield of the device determines the solidification time.* In any chain, the fraction of atoms that exists in the form of refractory elements at the time of solidification (F_R) is a measure of the refractory nature of that mass chain taken as a whole. According to the magnitude of the fraction, the chains are grouped into three categories: volatile, refractory, or mixed. The specification of ϕ (i.e., the degree of fractionation) will be shown to fix the value of the fractionation coefficient ($r_{k,95}$) for each chain k and, therefore, the radiochemical composition.

Part II of this series[2] showed that one could estimate the effect of fractionation on exposure-dose rate by summing the products of the fractionation coefficients and the unfractionated contributions for the individual chains. In so doing, it was convenient to divide the chains into groups, according to the volatility shown by their behavior. This procedure will be carried out in detail in the present report. Thus, these calculations will illustrate the definition of surface density of contamination proposed in Part II.

*The energy release from a nuclear device is given sometimes as the yield from fission reactions and sometimes as the total (fireball) yield. The latter includes the former plus the energy released by fusion reactions. The yield from fission reactions plays the dominant role in producing radioactivity. The total or fireball yield, together with the nature and location of environmental material, determines the condensation time and solidification time for the debris.

PRELIMINARY CONSIDERATIONS

Selection of Significant Chains

Although some 90 mass chains are produced in the thermal neutron fission of U^{235}, only about half of these will, at some time or other, make a significant contribution to the exposure dose rate. Therefore, calculations can be considerably shortened by considering only significant mass chains. In selecting those chains, and in all subsequent dose-rate calculations, the contributions of individual fission products, such as those compiled by Miller and Loeb for the products of thermal-neutron fission of U^{235}, will be used.[7] For any given time, the nuclides contributing less than 0.3 % to the total dose rate at that time will be considered insignificant. A significant nuclide and its isobars form a significant mass chain. These 47 significant chains will be listed in Table 1, together with the time interval in which they are significant. The earliest time considered will be 45.8 m.

Criterion of Volatility

The empirical relationships derived in Reference 1 depend upon a quantity, F_R, equal to the fraction of a particular chain that is refractory at the time of condensation in the fireball. Miller's thermodynamic model[4,6] for land-surface-burst fallout adopts an idealized soil that melts at 1400°C. Above this temperature, Miller's first stage of condensation pertains, and the product nuclides distribute themselves uniformly throughout the volume of molten particles. Below this temperature the particles are frozen and produce nuclides can only deposit on the surfaces. Miller calls this the second stage of condensation. In order to maintain consistency between the two approaches, we will also adopt this approximation and call all elements volatile whose predominant species have a normal boiling point less than 1400°C. These elements are As, Sc, Br, Kr, Rb, Mo, Tc, Te, I, Xe, and Cs. All other elements will be called refractory.

Calculation of F_R

To calculate F_R, we now need an equation to estimate the time at which 1400°C is reached by the cooling fireball. From Table 3.8 of Reference 6, the equation

$$t\ (\text{sec}) = 1.88\ W^{0.363}$$

can be inferred, where W is the total yield in kt.

TABLE 1

F_R Values for Significant Chains

Chain No.	6-sec Solidification		41-sec Solidification		Time of Significance[a]	
	Present	Glendenin	Present	Glendenin	From	To
83	0.01	0.04	0.00	0.00	45.8 m	1.12 h
84	0.00	0.01	0.00	0.00	45.8 m	3.52 h
85	0.00	0.00	0.00	0.00	2.40 h	23.8 h
87	0.00	0.00	0.00	0.00	45.8 m	11.1 h
88	0.00	0.00	0.00	0.00	45.8 m	23.8 h
89	0.00	0.00	0.00	0.00	45.8 m	2.40 h
90	0.01	0.01	0.08	0.08	2.60 y	55.3 y
91	0.06	0.07	0.16	0.17	45.8 m	3.12 d
92	0.28	0.30	0.82	0.83	45.8 m	1.45 d
93	0.60	0.62	0.99	0.99	45.8 m	3.12 d
94	0.88	0.87	0.99	0.99	45.8 m	2.40 d
95	0.97	0.97	1.00	1.00	16.2 h	2.60 y
97	1.00	1.00	1.00	1.00	45.8 m	6.70 d
98	1.00	1.00	1.00	1.00	45.8 m	3.52 h
99	0.99	0.97	0.86	0.83	7.56 h	21.1 d
101	0.80	0.60	0.09	0.07	45.8 m	2.40 h
102	0.61	0.39	0.02	0.01	45.8 m	1.64 h
103	0.34	0.20	0.17	0.17	26.2 h	1.20 y
105	0.12	0.21	0.75	0.80	45.8 m	4.57 d
106	0.20	0.36	0.93	0.95	66.4 d	8.18 y
107	0.36	0.54	0.99	0.99	45.8 m	45.8 m
118	1.00	1.00	1.00	1.00	1.20 y	55.3 y
125	1.00	1.00	1.00	1.00	1.20 y	12.0 y
126	1.00	1.00	1.00	1.00	2.40 h	2.13 d
127	1.00	1.00	1.00	1.00	2.13 d	14.4 d
128	1.00	1.00	1.00	1.00	1.64 h	4.57 d
129	0.97	0.98	0.97	0.98	45.8 m	23.8 h
131	0.74	0.86	0.73	0.84	45.8 m	66.4 d
132	0.48	0.70	0.40	0.58	1.12 h	30.9 d
133	0.25	0.53	0.23	0.50	45.8 m	30.9 d
134	0.09	0.24	0.01	0.02	45.8 m	11.1 h
135	0.00	0.10	0.00	0.00	45.8 m	3.12 d
137	0.00	0.00	0.00	0.00	66.4 d	250 y
138	0.00	0.00	0.00	0.00	45.8 m	5.26 h
139	0.00	0.02	0.04	0.06	45.8 m	1.11 h
140	0.06	0.11	0.28	0.32	4.56 h	143 d
141	0.38	0.47	0.96	0.96	45.8 m	301 d
142	0.71	0.78	1.00	1.00	45.8 m	1.11 h
143	0.94	0.96	1.00	1.00	45.8 m	14.4 d
144	0.98	0.99	1.00	1.00	14.4 d	8.18 y
145	1.00	1.00	1.00	1.00	45.8 m	2.13 d
146	1.00	1.00	1.00	1.00	45.8 m	3.52 h
147	1.00	1.00	1.00	1.00	23.8 h	97.3 d
149	1.00	1.00	1.00	1.00	45.8 m	14.4 d
151	1.00	1.00	1.00	1.00	7.56 h	4.57 d
152	1.00	1.00	1.00	1.00	45.8 m	3.52 h
155	1.00	1.00	1.00	1.00	1.78 y	3.80 y

a. No time earlier than 45.8 m is considered.

Bolles and Ballou[8] tabulate, for U^{235}-thermal neutron fission, the number of atoms of each fission-product mass chain present in various elemental forms at these times. These elemental distributions are calculated both according to Present's minimum kinetic energy (MKE) theory of charge distribution,[9] and according to the equal charge displacement (ECD) theory of Glendenin, Coryell and Edwards.[10] Although the latter treatment is more widely preferred, we will carry out calculations on both bases in order to illustrate the sensitivity of the calculations to the fractional chain yields.*

A sample calculation of F_R for the mass-92 chain, according to Present's theory, for a solidification time of 41 sec, is given below. Values of F_R for the significant mass chains are given in Table 1 for both theories of independent yield (nuclear charge distribution) for solidification times of 6 and 41 sec. According to the equation above, these correspond to total yields of about 25 kt and 5 Mt, respectively.

Mass-92 Decay Chain	Half-Life	Atoms per 10^4 Fissions at 41 sec. (Ref. 8)	Refractory Atoms per 10^4 Fissions
Br	1.5 s[a]	0	0
↓			
Kr	3 s	0	0
↓			
Rb	17 s[a]	102	0[b]
↓			
Sr	2.7 h	477	477
↓			
Y	3.5 h	1	1
↓			
Zr	stable	0	0
	Total	580	478

$$F_R = 478/580 = 0.82$$

a. Estimated half-life.
b. For silicate soil, Rb would probably behave refractorily because it forms refractory silicate compounds.

*The fractional chain yield of a fission-product radionuclide is the ratio of the independent (primary) yield of that nuclide to the total chain yield for that nuclide's mass chain. The concept applies only at the instant of fission.

Volatile, Mixed, and Refractory Chains

The significant mass chains can now be divided into three groups (volatile, mixed, or refractory) according to the F_R value for that chain. Chains with $0.98 \leqslant F_R \leqslant 1.00$ are called refractory, chains with $0.02 < F_R < 0.98$ are called mixed, and chains with $0.00 \leqslant F_R \leqslant 0.02$ are called volatile.

For unfractionated debris, the total exposure-dose rate and the contributions of the individual groups are shown as functions of time in Figs. 1 through 4 for the two yield theories and the two condensation times. The total curve is, of course, the same in all four figures.

The percentage contributions of each group to the total at 1.12 and 23.8 hr are shown in Table 2. As can be seen, 98 % or more of the dose rate is accounted for by the significant chains.

TABLE 2

Contribution of Groups to Unfractionated Dose Rate (%)

Group	1.12-hr dose rate		23.8-hr dose rate	
	Present	Glendenin	Present	Glendenin
	6-sec Solidification Time			
Volatile	33.4	30.9	16.8	0.4
Mixed	47.0	49.2	51.0	67.8
Refractory	18.5	18.8	30.4	29.9
Neglected	1.1	1.1	1.8	1.9
	41-sec Solidification Time			
Volatile	48.4	48.4	16.8	16.8
Mixed	21.2	20.9	38.6	38.0
Refractory	29.2	29.5	42.7	43.3
Neglected	1.2	1.2	1.9	1.9

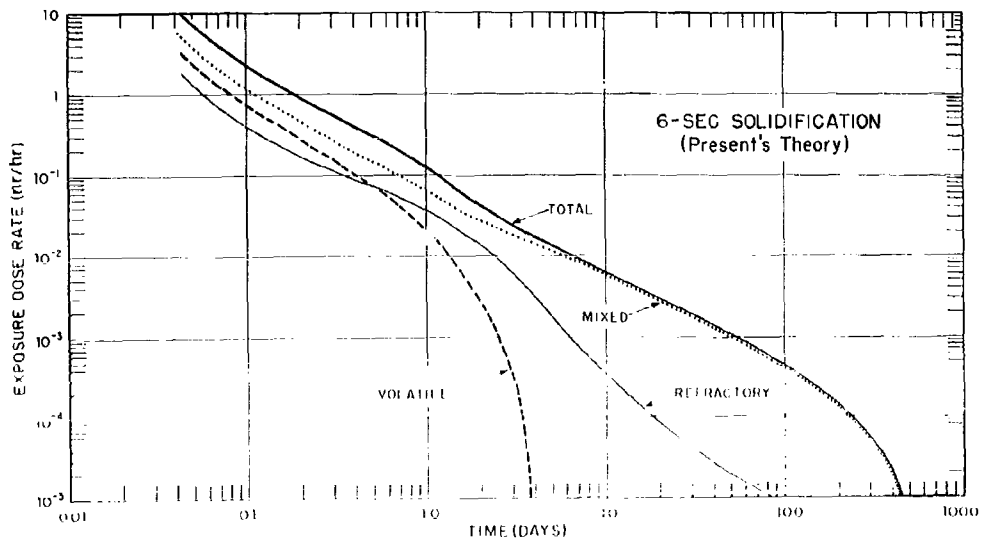

Fig. 1 Exposure-dose Rates From Volatile, Refractory, Mixed, and Total Fission-Product Mass Chains as Functions of Time. Calculated for 3 ft above an infinite, smooth, impenetrable plane, uniformly contaminated with the products of 10^4 thermal-neutron-induced fissions of U^{235} per sq. ft. (6-sec solidification, Present's Theory.)

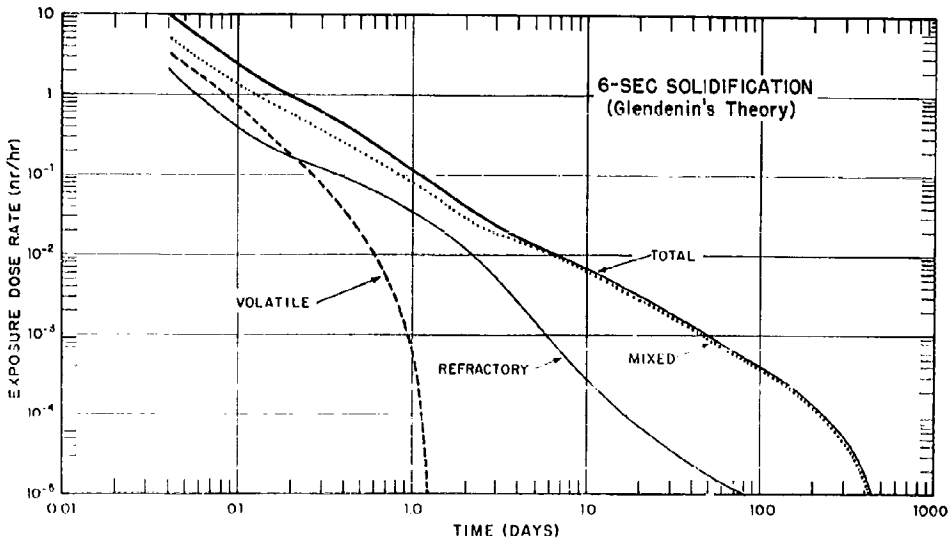

Fig. 2 Exposure-dose Rates From Volatile, Refractory, Mixed, and Total Fission-Product Mass Chains as Functions of Time. Calculated for 3 ft above an infinite, smooth, impenetrable plane, uniformly contaminated with the products of 10^4 thermal-neutron-induced fissions of U^{235} per sq. ft. (6-sec solidification, Glendenin's Theory.)

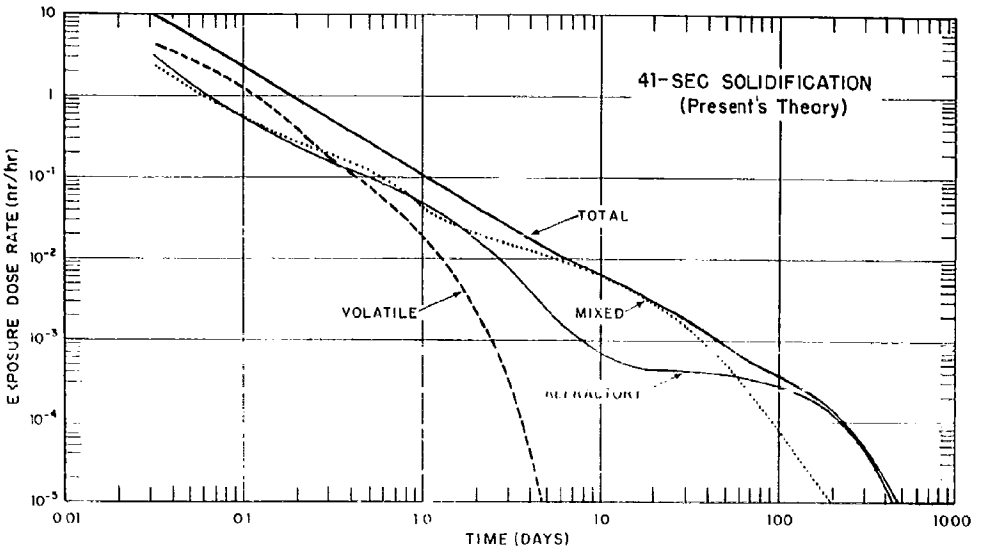

Fig. 3 Exposure-dose Rates From Volatile, Refractory, Mixed, and Total Fission-Product Mass Chains as Functions of Time. Calculated for 3 ft above an infinite, smooth, impenetrable plane, uniformly contaminated with the products of 10^4 thermal-neutron-induced fissions of U^{235} per sq. ft. (41-sec solidification, Present's Theory.)

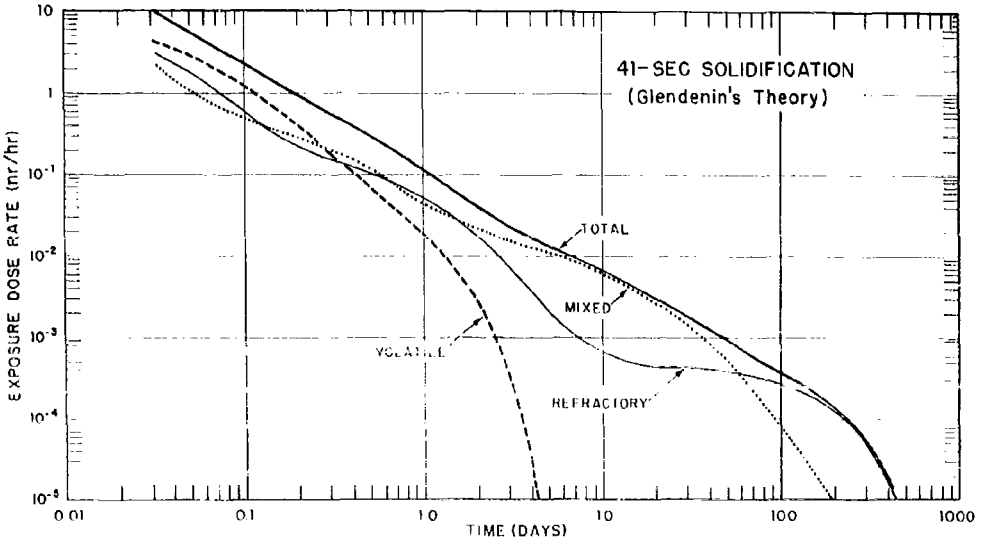

Fig. 4 Exposure-dose Rates From Volatile, Refractory, Mixed, and Total Fission-Product Mass Chains as Functions of Time. Calculated for 3 ft above an infinite, smooth, impenetrable plane, uniformly contaminated with the products of 10^4 thermal-neutron-induced fissions of U^{235} per sq. ft. (41-sec solidification, Glendenin's Theory.)

RELATION TO THE NEW DEFINITION OF CONTAMINATION LEVEL

Part II of this series[2] proposed a new definition of surface density of contamination which was applicable to fractionated debris. The new definition was summarized by Eq. 18 of that report as

$$D(t) = \sigma_{95} \left[\underbrace{\Sigma\, Y_k \bar{D}_k(t)}_{\text{quasi-refractory chains}} + \underbrace{\Sigma\, Y_k \left(r_{89,95}\right)^{1-b_k} \bar{D}_k(t)}_{\text{quasi-volatile chains}} + \underbrace{\Sigma\, Y_k \bar{D}_k(t)}_{\text{induced chains}} \right]$$

where σ_{95} is the contamination surface density in mass-95 chain equivalent fissions per unit area, Y_k is the average number of fission-product or induced atoms of mass k produced initially per fission, and $\bar{D}_k(t)$ is the dose rate contribution of the mass-k chain per atom of mass k per unit area at the time t. The quantity b_k is a correlation parameter which is dicussed in Ref. (3), and which has been observed to be approximately equal to $F_R^{1/2}$ in debris from high-yield surface bursts.[1,*]

For illustration and interim predictions, the "air ionization rates" (actually the exposure-dose rates) of individual fission-product radionuclides tabulated by Miller and Loeb provide a convenient source of data. These authors tabulate in units of mr/hr, the "air ionization rate" three feet above an infinite plane, each square foot of which is uniformly contaminated with the products of 10^4 thermal-neutron-induced fissions of U^{235}. In their Table A-1, they list the individual contribution of each fission-product radionuclide at various convenient times. In the notation of Ref. 2, (cf. Eq. 6) each entry would be written

$$D_j = \sigma_k Y_k d_j G_{jk}(t)\, \lambda_j$$

with the contamination surface density (σ_k) in units of 10^4 fissions per sq ft, the total chain yield for mass k (Y_k) being the value for thermal-neutron fission of U^{235}, and the product of the dose-rate conversion factor (d_j) the chain fraction (G_{jk}) and the decay constant (λ_j) being in units of mr per hr per fission. Since the tabulated data

*This relation neglects the departure from zero of the term a_k of Refs. 2 and 3. When dealing with samples of fallout, rather than particles of a single size, this is justifiable.

is for unfractionated fission products, each radionuclide has the same value of σ_k. Thus, D_j gives the dose rate from each radionuclide under the conditions stated, and if the values are summed for all nuclides of a given mass k, one obtains D_k, the dose rate from the mass-k chain at the time t. In Ref. 2, it was found convenient to abbreviate the sum $\Sigma_j d_j g_{jk}(t)\lambda_j$ by $\bar{D}_k(t)$ and write (Ref. 2, Eq. 7)

$$D_k = \sigma_k Y_k \bar{D}_k(t)$$

To illustrate the application of Eq. 18 with Miller and Loeb's data, we will consider first the quasi-refractory term. The value of this term is estimated by (1) choosing from Table 1 the mass chains with $1.00 \geq F_R \geq 0.98$ for the appropriate conditions, (2) summing all the data tabulated by Miller and Loeb for these chains at the time of interest t.

In calculating the value of the quasi-volatile term, it has been found convenient and instructive to consider the mass chains to fall into two classes: a purely volatile class with $0.02 \geq F_R \geq 0.00$ and a mixed class with $0.98 > F_R > 0.02$. For the purely volatile class, we (1) choose the appropriate mass chains as above, (2) sum their contributions for the time of interest, and (3) multiply the sum by the fractionation ratio we wish to apply. The contribution for the mixed chains is calculated similarly, except that in each case the fractionation ratio must be raised to the appropriate power $1-b_k$, here taken as $1 - F_R^{1/2}$, for the chain in question.

The induced chains are not considered in this report. The value of σ_{95} is taken as 10^4 fissions per square foot.

CALCULATIONS AND RESULTS

Exposure-Dose Rates

Each term in the summation of D(t) can be considered as the product of the contribution from unfractionated fission products ($\sigma_{95}\, Y_K D_K$) and a fractionation correction factor ($r_{89,95}$)$^{1-b_K}$. Figure 5 illustrates the variation of this correction factor with fractionation ratio ($r_{89,95}$) as computed for several values of F_R.

As a specific example, consider the mass-132 chain contribution at 1.12 h from a burst with a 41-sec solidification time. From Table 1, the value of F_R according to Present's theory is 0.40. From Fig. 5, the correction factors for $r_{89,95}$ values of 1/3, 1/10, 1/30 and 1/100 would be 0.667, 0.427, 0.284 and 0.182 respectively. The unfractionated contribution of this chain to the total dose rate is calculated from Ref. 7 as 0.01010 nr/hr/10^4 fissions/ft^2. Contributions which the chain would in fact provide, for the fractionation ratios listed, are therefore 0.00674, 0.00431, 0.00287 and 0.00184 nr/hr/10^4 fissions/ft^2, respectively. Similar calculations of contributions for all the other mixed chains and summation of these contributions give values of 0.846, 0.554, 0.434 and 0.347 nr/hr/10^4 fissions/ft^2 respectively, at these fractionation ratios. Tables 3 through 6 list the contributions of each group for each set of conditions considered, together with their totals. They are given both in units of nr/hr for 10^4 mass-95 chain equivalent fissions*/sq ft and in kr/hr for 1.45 x 10^{23} mass-95 chain equivalent fissions/sq mi. To illustrate the functional behavior of the contributions, Figs. 6 through 9 show the variations of component contributions and total dose rates with the fractionation ratio, so that the effects of solidification time, independent-yield theory and exposure time can be independently observed. The figures are drawn for a constant number of mass-95 chain fissions. On this basis the contribution of the refractory group (b_K=1) is constant and the contribution of the volatile group (b_K=0) is directly proportional

*The number of mass-95 chain equivalent fissions (f_{95}) is equal to the number of device fissions which produced the quantity of mass-95 chain observed.

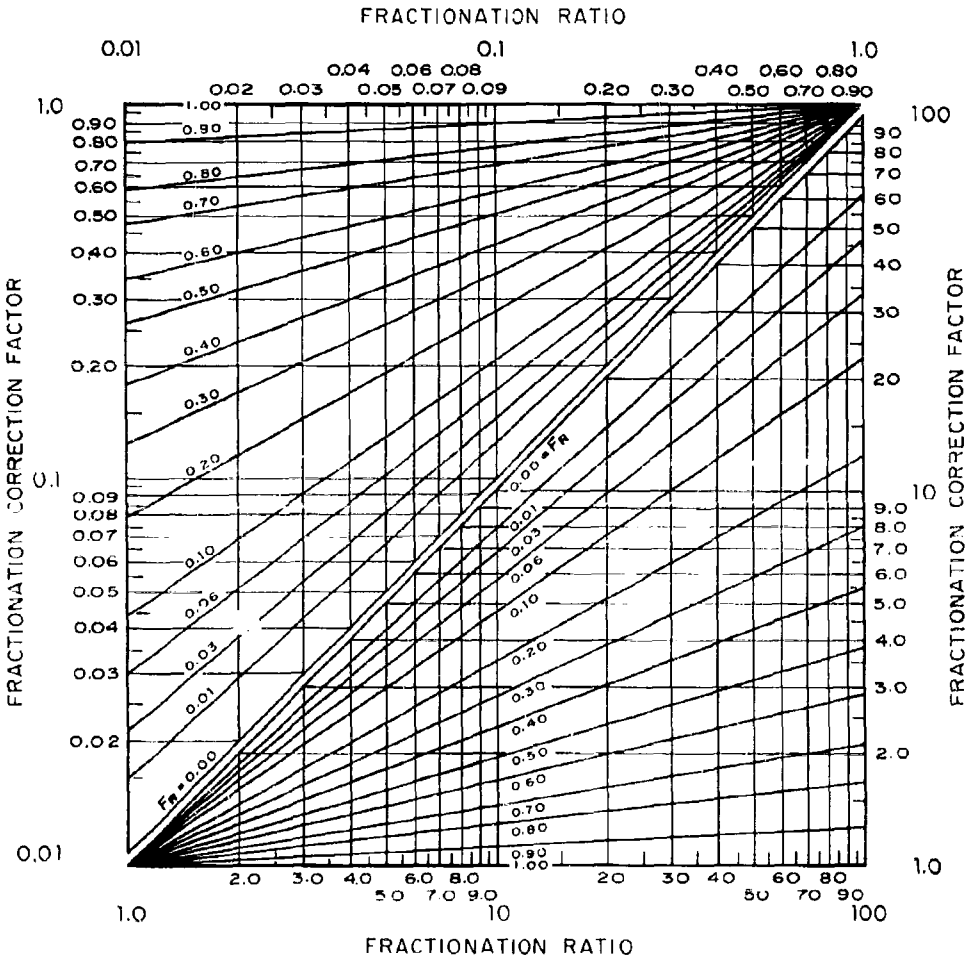

Fig. 5 Fractionation Correction Factors as Functions of F_R and $r_{89,95}$

TABLE 3

1.12-HR EXPOSURE-DOSE RATE FOR FRACTIONATED DEBRIS FROM A BURST WITH 6-SEC SOLIDIFICATION TIME

$r_{89,95}$	Present				Glendenin			
	Volatile	Mixed	Refractory	Total	Volatile	Mixed	Refractory	Total
	Dose rate in mr/hr for $\sigma=10^4$ mass-95 chain equivalent fissions per ft^2							
1	2.086	2.941	1.155	6.182	1.932	3.074	1.176	6.182
1/3	0.695	1.918	1.155	3.768	0.544	2.162	1.176	3.982
1/10	0.209	1.238	1.155	2.602	0.193	1.533	1.176	2.902
1/30	0.070	1.011	1.155	2.236	0.064	1.196	1.176	2.436
1/100	0.021	0.804	1.155	1.980	0.019	0.943	1.176	2.138
0	-	-	1.155	1.155	-	-	1.176	1.176
	Dose rate in kr/hr for $\sigma=1.45 \times 10^{23}$ mass-95 chain equivalent fissions per mi^2							
1	1.034	1.529	0.601	3.214	1.004	1.598	0.612	3.214
1/3	0.362	0.998	0.601	1.961	0.335	1.124	0.612	2.071
1/10	0.108	0.644	0.601	1.353	0.101	0.797	0.612	1.510
1/30	0.036	0.526	0.601	1.163	0.034	0.622	0.612	1.268
1/100	0.011	0.418	0.601	1.030	0.010	0.491	0.612	1.113
0	-	-	0.601	0.601	-	-	0.612	0.612

Note: The total exposure dose rate for the case $r_{89,95}=1$ is always the exposure dose rate resulting from unfractionated fission products, and in this case is obtained by summing all the contributions from the significant chains listed in Table A-1 of reference 7.

TABLE 4

23.8-HR EXPOSURE-DOSE RATE FOR FRACTIONATED DEBRIS FROM A BURST WITH 6-SEC SOLIDIFICATION TIME

$r_{89,95}$	Present				Glendenin			
	Volatile	Mixed	Refractory	Total	Volatile	Mixed	Refractory	Total
	Dose rate in nr/hr for $c=10^4$ mass-95 chain equivalent fissions per ft^2							
1	0.020	0.060	0.036	0.116	0.001	0.080	0.035	0.116
1/3	0.007	0.039	0.036	0.082	0.000	0.052	0.035	0.087
1/10	0.002	0.026	0.036	0.064	0.000	0.035	0.035	0.070
1/30	0.001	0.019	0.036	0.056	0.000	0.026	0.035	0.061
1/100	0.000	0.014	0.036	0.050	0.000	0.020	0.035	0.055
0	-	-	0.036	0.036	-	-	0.035	0.035
	Dose rate in kr/hr for $c=1.45 \times 10^{23}$ mass-95 chain equivalent fissions per mi^2							
1	0.010	0.031	0.019	0.060[a]	0.001	0.042	0.018	0.061[a]
1/3	0.003	0.020	0.019	0.042	0.000	0.027	0.018	0.045
1/10	0.001	0.013	0.019	0.033	0.000	0.018	0.018	0.036
1/30	0.000	0.010	0.019	0.029	0.000	0.014	0.018	0.032
1/100	0.000	0.007	0.019	0.026	0.000	0.011	0.018	0.029
0	-	-	0.019	0.019	-	-	0.018	0.018

a. Difference due to rounding-off errors.

TABLE 5

1.12-HR EXPOSURE-DOSE RATE FOR FRACTIONATED DEBRIS FROM A BURST WITH 41-SEC SOLIDIFICATION TIME

$r_{89,95}$	Present				Glendenin			
	Volatile	Mixed	Refractory	Total	Volatile	Mixed	Refractory	Total
	Dose rate in nr/hr for $\sigma=10^4$ mass-95 chain equivalent fissions per ft^2							
1	3.026	1.328	1.827	6.181[a]	3.026	1.309	1.848	6.183[a]
1/3	1.009	0.846	1.827	3.682	1.009	0.911	1.848	3.768
1/10	0.303	0.554	1.827	2.684	0.303	0.667	1.848	2.818
1/30	0.101	0.434	1.827	2.362	0.101	0.532	1.848	2.481
1/100	0.030	0.347	1.827	2.204	0.030	0.436	1.848	2.314
0	-	-	1.827	1.827	-	-	1.848	1.848
	Dose rate in kr/hr for $\sigma=1.45 \times 10^{23}$ mass-95 chain equivalent fissions per mi^2							
1	1.573	0.690	0.950	3.213[a]	1.573	0.681	0.961	3.215[a]
1/3	0.525	0.440	0.950	1.915	0.525	0.474	0.961	1.960
1/10	0.157	0.288	0.950	1.395	0.157	0.347	0.961	1.465
1/30	0.052	0.226	0.950	1.228	0.052	0.277	0.961	1.290
1/100	0.016	0.180	0.950	1.146	0.016	0.227	0.961	1.204
0	-	-	0.950	0.950	-	-	0.961	0.961

a. Differences due to rounding-off errors.

TABLE 6

23.8-HR EXPOSURE-DOSE RATE FOR FRACTIONATED DEBRIS FROM A BURST WITH 41-SEC SOLIDIFICATION TIME

$r_{89,95}$	Present				Glendenin			
	Volatile	Mixed	Refractory	Total	Volatile	Mixed	Refractory	Total
	Dose rate in nr/hr for $\sigma=10^4$ mass-95 chain equivalent fissions per ft^2							
1	0.020	0.045	0.050	0.115[a]	0.020	0.045	0.051	0.116[a]
1/3	0.007	0.029	0.050	0.086	0.007	0.031	0.051	0.089
1/10	0.002	0.019	0.050	0.071	0.002	0.022	0.051	0.075
1/30	0.001	0.014	0.050	0.065	0.001	0.017	0.051	0.069
1/100	0.000	0.010	0.050	0.060	0.000	0.013	0.051	0.064
0	-	-	0.050	0.050	-	-	0.051	0.051
	Dose rate in kr/hr for $\sigma=1.45 \times 10^{23}$ mass-95 chain equivalent fissions per mi^2							
1	0.010	0.023	0.026	0.059[a]	0.010	0.023	0.027	0.060[a]
1/3	0.003	0.015	0.026	0.044	0.003	0.016	0.027	0.046
1/10	0.001	0.010	0.026	0.037	0.001	0.012	0.027	0.040
1/30	0.000	0.007	0.026	0.033	0.000	0.009	0.027	0.036
1/100	0.000	0.005	0.026	0.031	0.000	0.007	0.027	0.034
0	-	-	0.026	0.026	-	-	0.027	0.027

a. Differences due to rounding off errors.

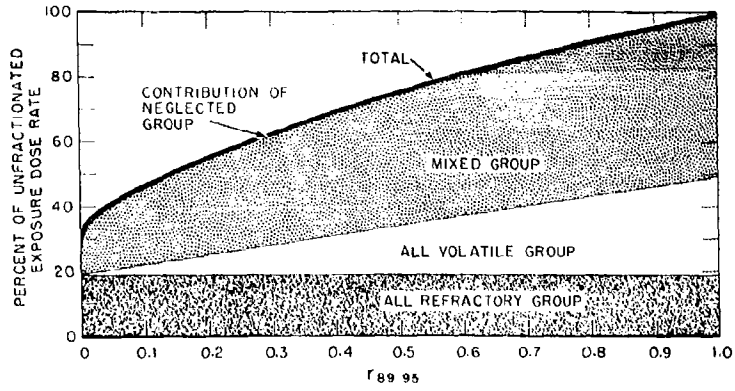

Fig. 6 Relative Exposure-Dose Rate From Volatile-Depleted Fission Products as a Function of Fractionation Ratio. Same geometry as Fig. 1. (1.12 h, 6-sec, solidification, Glendenin's Theory)

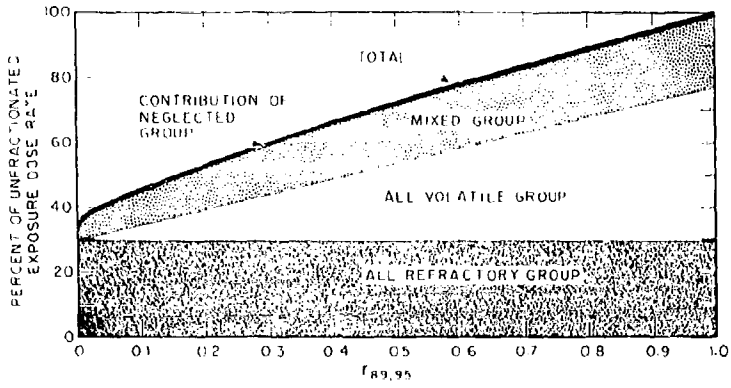

Fig. 7 Relative Exposure-Dose Rate From Volatile-Depleted Fission Products as a Function of Fractionation Ratio. Same geometry as Fig. 1. (1.12 h, 41-sec, solidification, Glendenin's Theory)

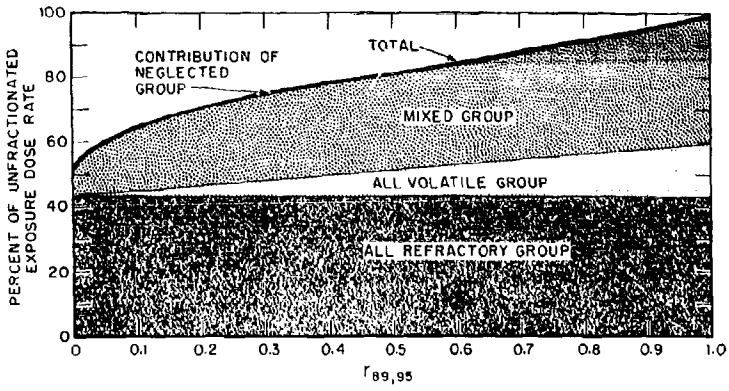

Fig. 8 Relative Exposure-Dose Rate From Volatile-Depleted Fission Products as a Function of Fractionation Ratio. Same geometry as Fig. 1. (23.8 h, 41-sec. solidification, Glendenin's Theory)

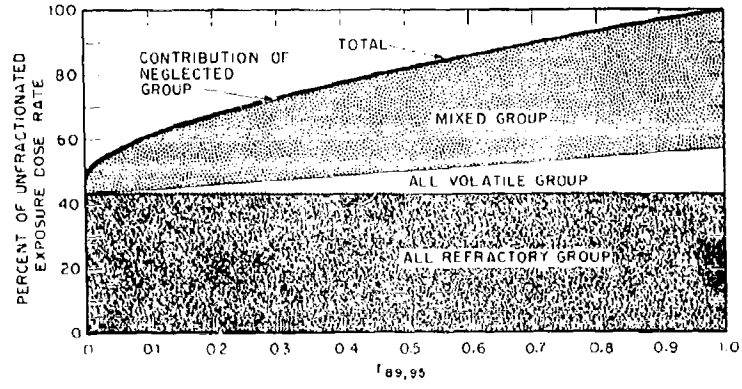

Fig. 9 Relative Exposure-Dose Rate From Volatile-Depleted Fission Products as a Function of Fractionation Ratio. Same geometry as Fig. 1. (23.8 h, 41-sec. solidification, Present's Theory)

to the fractionation ratio. In order to include the contribution of induced activities, one need only increase the contribution of the refractorily behaving group by the amount appropriate to the device and environmental conditions of interest.

Estimated Decay and the Way-Wigner Rule

By assuming that, at reasonably early times unfractionated mixed fission products behave as a statistical assembly of emitters with decay constants linearly related to the fifth power of the disintegration energy, Way and Wigner[11] derived a $t^{-1.2}$ dependence for the gross beta decay. The rule has been found useful in estimating the exposure-dose rate decay of mixed fission products,[12] but has become the subject of a popular fallacy, namely, that departures from the rule indicate fractionation, while correspondence to the rule indicates representativity. To illustrate the effect of fractionation on the decay of exposure dose rate, Table 7, lists the ratio of the 1.12-hr exposure-dose rate to the 23.8-hr exposure-dose rate as calculated for the various conditions listed here and from the $t^{-1.2}$ rule. The calculations show that, rather than produce a departure from the $t^{-1.2}$ rule, it is possible that, in the absence of induced activities, fractionation could promote correspondence to the rule. The main point here is to beware of superficial generalizations based on such a complicated property of nuclear debris as gross decay rate.

TABLE 7

Ratio of 1.12-hr Exposure-Dose Rate to 23.8-hr Exposure Dose Rate for Various Conditions

$r_{89,95}$	6-sec Solidification		41-sec Solidification	
	Present	Glendenin	Present	Glendenin
1/3	46	46	43	42
1/10	41	41	38	38
1/30	40	40	36	36
1/100	40	39	37	36
0	32	34	37	36

Unfractionated value ($r_{89,95}=1$): 53

Way-Wigner value $(1.12/23.8)^{-1.2}$: 39

DISCUSSION

Although the calculations described here are primarily intended to illustrate the application of a new definition of surface density of contamination, the results also illustrate the magnitude of the variation in exposure-dose rate to be expected from fractionation, and the sensitivity of the results to several important variables. During and subsequent to the preparation of this report, more up-to-date and realistic input data was and is becoming available. This data includes total chain and independent fission yields for various fission processes, improved dose-rate conversion factors from new decay-scheme information, and fractionation-correlation parameters from silicate-surface bursts. More detailed calculations will be made when the acquisition of this input and a computer program for its application have been completed.

For the present, the predicted dose-rate from local fallout for a given degree of fractionation does not appear sensitive to the independent fission-yield data used as input. The sensitivity to total yield is significant only at extreme degrees of fractionation. The predicted effect of fractionation varies considerably with the time at which exposure-dose rate is estimated.

Many other fractionation effects remain to be dealt with. These may be of much greater magnitude and show much greater sensitivity to independent fission-yield data and solidification or condensation time than does the effect on exposure-dose rate from local fallout in land-surface burst debris. These include: the partition of radionuclides between local, intermediate and world-wide fallout (as treated by a theoretical rather than a semi-empirical method); the effect on transient dose and dose-rate; the exposure dose-rate from debris which is enriched in volatilely-behaving mass chains; fractionation in venting underground and underwater bursts, on land surfaces or particle surfaces that show different correlation properties than those used here, or in tower and low air bursts.

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APPENDIX A

COMPARISON OF OUR NOTATION WITH THAT OF OTHER WORKERS

The following comparison of notation will help the reader compare work published by different investigators on the subject of radionuclide fractionation in nuclear debris.

The symbols used to describe fractionation in this series of reports are as follows:

F	Total number of fissions occurring in a nuclear event
A_i	Total number of atoms of chain i, either fission product or induced activity, occurring in the event
Y_i	Total yield of chain i in the event. $Y_i = A_i/F$
a_i	Number of atoms of chain i in a given sample as determined by radiochemical analysis and corrected to the time the event occurred. The same symbol has also been used for a correlation parameter (see b_i below).
f_i	The number of fissions in the sample according to the analysis for chain i, hence, the mass-i chain equivalent fissions. $f_i = a_i/Y_i = a_i F/A_i$
$r_{i,j}$	The fractionation coefficient for the i and j chains. $r_{i,j} = f_i/f_j$
$r_{89,95}$	The fractionation ratio. A measure of the departure of a given samples' radiochemical composition from representativity.
Φ	The degree of fractionation. $\Phi = \log_{10} r_{89,95}$

b_i The slope of the regression curve correlating log $r_{i,89}$ with $-\phi$. $1 - b_i$ is the slope for correlating log $r_{i,95}$ with ϕ. $\log_{10} r_{i,95} = a_i + (1 - b_i)\phi$.

$F_R(t)$ The fraction of atoms in a given mass chain which are in a refractory (condensible) form at the time t under the fireball or cloud conditions prevailing at that time.

Edvarson, Low and Sisefsky (A1) use f_{A-95} which they call a "fractionation factor," to indicate the fractionation of nuclide A from the mass-95 chain. This would be equivalent to our $r_{A,95}$, if we used A, rather than i, to indicate the mass chain.

Mamuro, et al., (A2) also call this same quantity a fractionation factor, but give it the symbol f.

Miller's notation (A3) is more complex. He uses a variety of symbols, which he calls "fractionation numbers," in apparently the following way. On page 51 of his report, he uses $r_o(A)$ in a manner equivalent to $r_{A,95}$, although he defines it on page xxviii more like our F_R. The quantity R^A_{99} on page 62 of his report, when referred to thermal neutron fissions from U^{235} as a reference event, is the familiar $R^{99}(A)$ value, frequently used by radiochemists and diagnosticians. When referred to a given nuclear burst as the reference event, however, it becomes $r_{A,99}$ in our notation or f_{A-99} in Edvarson's. On pages 107 and 108, $r_o(A,t)$ is our $F_R(t)$, while $r^{99}(A)$ is again $r_{A,99}$.

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3. Radiological contamination.
4. Radioactive fallout.
5. Density.
6. Surface area.
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