63 - 3 - 2

FRACTIONATION II.
ON DEFINING THE SURFACE DENSITY OF CONTAMINATION

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ABSTRACT

This report presents a technical basis for defining surface

density of fallout contamination when the contaminating debris is

fractionated. Specifically, it recommends that the exposure dose

rate from fractionated debris be expressed as a contamination surface

density multiplied by the sum of three terms. The first term is the

exposure dose rate contribution of refractorily behaving fission-

product nuclides per unit contamination surface density. The second

term is a similar quantity for volatilely behaving fission-product

nuclides. The third term expresses the contribution of the induced

activities.

i

PREFACE

This series of reports presents and discusses the effects of radionuclide fractionation in nuclear-bomb debris. The first part (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 non-representative of the detonation products taken as a whole." It showed how the radionuclide compositions of fractionated samples could be correlated empirically by logarithmic relations. The present part uses these relations as the basis of a technical discussion of contamination density as applied to fractionated nuclear debris. The third part will present a theoretical foundation for the observed logarithmic correlations of Part I. It will use this as a simplified means of estimating fractionation as a function of particle size and the partition of product radionuclides among local, intermediate, and world-wide fallout. The fourth part will extend the calculations to show how fractionation-correlation parameters can be used to estimate the exposure dose rate from nuclear debris with various degrees of fractionation. It will serve to illustrate the proposals made in the present part.

INTRODUCTION

Those engaged in the study of fallout contamination from nuclear debris frequently express the surface density of contamination in terms of "kiloton-equivalents of debris per square mile." This term is particularly useful in discussing exposure dose rate, and when applied to unfractionated debris (1) it is easily defined with precision (2). However, the presence of unfractionated nuclear debris is the exception rather than the rule, and the concept of "kiloton-equivalent of debris" becomes undefined when the debris is fractionated. That this is so becomes evident when one considers that "kiloton-equivalent of debris" is actually a variation of the concept, "fraction of the device." As explained in Reference 1, "fraction of the device" becomes undefined in the presence of fractionation. It is best replaced with the fraction of some component of the debris, either of some residual, pre-shot material or of some product of the nuclear processes which occurred. Because of fractionation, the fraction of one component per unit area at a given location will not necessarily equal the fraction of any other. The difficulty is overcome by choosing one component as a reference and relating the abundance of the other components to it.

2

The purpose of this report is to present the technical background for modifying the "kiloton-equivalent of debris" concept for fractionated fallout and to define the modified concept in simple terms. The material will be presented from the viewpoint of exposure dose rate. Since the "kiloton-equivalent" is, by definition, 10^{12} calories or a "teracalorie", we will use the latter term throughout the remainder of this report.

TECHNICAL DISCUSSION

Basic Considerations

The radiological hazard from a deposit of fallout is generally expressed as exposure dose rate (D) and arises from the photons produced by radioactive decay and by various interactions of the radiation with matter. The radionuclides responsible are predominantly beta-gamma emitters. The beta particles can interact with nuclei to form photons known as Bremsstrahlen, but these contribute only a few percent to the total dose rate and will not be considered further. The gamma-photons may be internally converted to characteristic X rays, or they may interact with environmental material by elastic scattering, photoelectric effect, Compton scattering, or pair production. Regardless of subsequent alterations, the primary emissions of radiological significance are the gamma photons and characteristic X rays, which are energetically discrete and virtually independent of the environment.

3

Their relation to activity, spectra, and exposure dose rate can be described as follows. The terminology, symbols and units used are summarized at the end of the discussion.

Consider a homogeneous source of N_j atoms of type j disintegrating according to the radioactive decay law

$$A_j = -\dot{N}_j = \lambda_j N_j \qquad (1)$$

where A_j is the disintegration rate (activity) and λ_j is the total radioactive decay constant. The photons emitted will have discrete energies (E_i). An average of N_{ij} photons with energy E_i will be produced per disintegration. The (total) photon emission rate for these atoms is $A_j \sum_i N_{ij}$ and the rate of energy emission in the form of photons is $A_j \sum_i N_{ij} E_i$. The distribution as a function of energy of the fraction of the total number of photons emitted which fall into each energy group is called the photon spectrum.

Now consider the source material as a contaminant distributed over a soil-air interface. The quantity of radioactive material of type j per unit area at any location on the interface is the surface density of contamination (σ_j) at that location. The exposure dose rate sensed by a detector at any point in this system will be due to the photon spectrum at that point and this will differ from the primary spectrum in several ways: it will no longer be discrete, it will be degraded in energy, and will depend upon both the environment and the source-detector geometry. Each photon in this secondary spectrum will produce a contribution to the dose which will depend upon its energy.

4

The calculation of dose rate from the nature of the source and from the source-detector arrangement for even one type of nuclide is very complicated and requires the introduction of several reasonable simplifications.

Calculation of Exposure Dose Rate From a Homogeneous, Distributed Source Composed of a Single Nuclide.

A reasonably accurate simplifying device for calculating exposure dose rate from a distributed source is found in the dose-rate conversion factor. Assume that the homogeneous contaminant is distributed on the interface so that the contamination density varies gradually with location and, as far as any given detector position is concerned, may be considered everywhere equal to the contamination density σ_j at the closest point of the interface. Primary photons of energy E_i now produce a contribution to the dose rate D_i which is proportional to the photon emission rate and therefore to $\sigma_j \lambda_j$. The proportionality factor (d_i) is a function of the energy and will be called the photon dose-rate conversion factor for the given arrangement.

$$D_i = d_i \, N_{ij} \, \sigma_j \, \lambda_j \tag{2}$$

A nuclide dose-rate conversion factor (d_j) is defined similarly in terms of the total dose rate (D_j) from all nuclides of type j. Whereas D_i is proportional to the photon emission rate per unit area, D_j will be proportional to the disintegration rate per unit area:

5

$$D_j = d_j \sigma_j \lambda_j = \sigma_j \lambda_j \sum_i d_i N_{ij} \qquad (3)$$

Therefore

$$d_j = \sum_i d_i N_{ij} \qquad (4)$$

To illustrate dose-rate calculations for fallout, it is customary to specify the following standard set of conditions: the detector is located 3 feet in air above an infinite, smooth, uniformly contaminated plane of unspecified soil material which is impenetrable to the radioactive fallout; the time is 1 hour after detonation with reference to radioactive decay, but is an infinite time after detonation with reference to the amount of material fallen out. An asterisk will be used to indicate dose rates and dose-rate conversion factors for standard conditions. Thus

$$D_j^* = d_j^* \sigma_j \lambda_j = \sigma_j \lambda_j \sum_i d_i^* N_{ij} \qquad (5)$$

Extension to Homogeneous, Representative Sources Composed of a Single Mass Chain.

As mentioned earlier, the radiologically important radionuclides produced in nuclear detonations are predominantly beta-gamma emitters, although some neutron emission occurs at early times. Since the nuclidic mass does not change in beta-gamma emission, fission products and induced activities group into isobaric chains[*], each with a constant total number of atoms. Although some 90 mass chains result from fission, only a few of these contribute significantly to the dose rate at any given time. They can be treated as follows.

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[*] Nuclides with equal mass numbers but different nuclear charge are called isobars.

6

Let Y_k be the average number of fission-product or induced atoms of mass k produced per fission. Then the fraction of atoms with mass k which exist as type j at any given time is a perfectly definite quantity which we will call $G_{jk}(t)$. The number of atoms of type j per fission existing at any time is $Y_k G_{jk}(t)$. If a total number of fissions F has occurred, the total number of j-type atoms present at time t is $FY_k G_{jk}(t)$. Let us represent the number of atoms of mass chain k per unit area by $\sigma_k Y_k$. The number of atoms of type j per unit area is now $\sigma_j = \sigma_k Y_k G_{jk}(t)$. Here σ_k is the number of fissions which produced the quantity of chain k present per unit area at the location considered. Now the total contribution of the kth chain to the dose rate $\left(D_k\right)$ is obviously the sum of the contributions of the individual members.

$$
\begin{aligned}
D_k &= \sum_j D_j \\
&= \sum_j d_j \sigma_j \lambda_j \\
&= \sigma_k Y_k \sum_j d_j G_{jk}(t) \lambda_j
\end{aligned}
\tag{6}
$$

The quantity $d_j G_{jk}(t) \lambda_j$ is the dose rate contribution from isobar j per atom of chain k at time t. Its sum over j is the dose rate contribution of the k chain per atom of mass k per unit area. It will be convenient to designate this sum $\bar{D}_k(t)$ and write

$$
D_k = \sigma_k Y_k \bar{D}_k(t)
\tag{7}
$$

7

The total contribution from all chains is

$$D = \sum_k D_k$$

$$= \sum_k \sigma_k \, Y_k \, \bar{D}_k \, (t) \tag{8}$$

If the debris is representative, all σ_k's are equal and we can write

$$D = \sigma \sum_k Y_k \, \bar{D}_k \, (t). \tag{9}$$

Fractionated Debris

The above formulas can reflect fractionation in two different ways. The first is by requiring a different value of σ for each mass chain. The second is by requiring a factor for each nuclide to correct Y_k. The second way reflects the fractionation of isobars from one another. It is predominant in the early stages, when debris is formed, and is responsible for much of the fractionation observed. For example, the fractionation of Xe^{140} and Cs^{140} from Ba^{140} produces the volatile behavior observed for the mass-140 chain. However, subsequent growth and decay convert this intrachain fractionation into interchain fractionation, so that by one hour after detonation we are justified in neglecting intrachain fractionation and concentrating on the variation of σ_k.

The variations in σ_k are most easily handled by taking one mass chain as a reference and relating σ_k for all other mass chains to that for the reference chain. Now in any nuclear event, a large number of mass chains do not fractionate from the mass-95 chain. Abundant

data exist for the mass-95 chain composition in fallout from previous nuclear tests. It is one of the most refractorily behaving mass chains and one least subject to secondary fractionation. It has a high and relatively constant fission yield which can be determined in two independent ways by analyzing for either Zr^{95} or Nb^{95}. The half lives of these nuclides are such that they can be determined long after fission. For these reasons the mass-95 chain is chosen as the primary reference chain. Then σ_{95} becomes the measure of contamination density. It is the number of mass-95 chain equivalent fissions (i.e., the number of fissions required to produce the number of mass-95 atoms found) per unit area at a given location.

The fractionation of other chains can be expressed by ratios of the type

$$r_{k,95} = \sigma_k / \sigma_{95} \tag{10}$$

The total dose can now be written

$$D = \sigma_{95} \sum_k Y_k \, r_{k,95} \, \bar{D}_k \, (t) \tag{11}$$

Empirical correlations of fractionated debris (1) and theoretical considerations (3) both lead to relations among the ratios of the type

$$\log_{10} r_{k,89} = a_k + b_k \log_{10} \left(\frac{1}{r_{89,95}} \right) \tag{12}$$

Here $r_{89,95}$* is the fractionation ratio and is a measure of the degree

*The fractionation ratio $r_{89,95}$ is the ratio of the number of fissions required to produce the amount of the mass-89 chain found in a sample to the number of fissions which would be required to produce the amount of the mass-95 chain found in the same sample.

9

of fractionation at the location of interest. The quantities a_k and b_k are empirically determined by the intercept and slope of a log-log plot of $r_{k,89}$ vs. $1/r_{89,95}$. The quantity a_k can be theoretically determined from b_k if the particle-size distribution is known (4). For dose-rate calculations, its departure from zero can be neglected when the particle size distribution is unknown, and we can write the above in a more convenient, approximate form

$$r_{k,95} \approx (r_{89,95})^{1 - b_k} \tag{13}$$

For important contributors to the radiological field, the quantity b_k is almost always between 0 and 1 in value and its magnitude can be estimated by an empirical rule (1). Equation (11) can then be written

$$D = \sigma_{95} \sum_k Y_k (r_{89,95})^{1 - b_k} \bar{D}_k (t) \tag{14}$$

The various mass chains can be divided into three groups. The first group consists of those mass chains which do not fractionate significantly from the mass-95 chain. Examples are the mass 99, 103, 106, and 144 through 161 fission-product chains and the activities induced in the weapon components. For these refractorily behaving chains, $b_k \approx 1$ and their contribution to the dose rate is

$$D_R = \sigma_{95} \sum Y_k \bar{D}_k (t). \tag{15}$$
$$\text{quasi-refractory}$$
$$\text{chains}$$

The other fission-product mass chains will exhibit values of b_k which depart appreciably from unity. In local fallout, they will be depleted relative to the mass-95 chain. The contribution of these volatilely behaving chains can be written

$$D_v = \sigma_{95} \; \Sigma \; (r_{89,95})^{1-b_k} \; Y_k \; \dot{D}_k \, (t). \qquad (16)$$
$$\text{quasi-volatile} \\ \text{chains}$$

Finally, since they do not have gaseous precursors, and have shown no evidence of volatile behavior, the induced activities can be assumed not to fractionate from the refractorily behaving activities (i.e., σ_k can be assumed equal to σ_{95}). The induced activity chains can also be considered to have yields Y_k, the yield of the kth chain being simply the ratio of the number of atoms of that chain produced by the detonation to the total number of fissions in the detonation. (The value, however, is not easily established with certainty.) Thus we can write their contribution as

$$D_I = \sigma_{95} \; \Sigma \; Y_k \; \dot{D}_k \, (t) \qquad (17)$$
$$\text{induced} \\ \text{chains}$$

The total exposure dose rate can now be written

$$D\,(t) = \sigma_{95} \left[\Sigma \, Y_k \, \dot{D}_k(t) \; + \; \Sigma \, Y_k \, (r_{89,95})^{1-b_k} \, \dot{D}_k \, (t) \; + \; \Sigma \, Y_k \, \dot{D}_k \, (t) \right]. \quad (18)$$
$$\text{quasi-refractory} \qquad\qquad \text{quasi-volatile} \qquad\qquad \text{induced} \\ \text{chains} \qquad\qquad\qquad \text{chains} \qquad\qquad\qquad \text{chains}$$

This equation says that the exposure dose rate from fractionated debris can be expressed as a contamination surface density multiplied by the sum of three terms. The first is the exposure dose rate contribution of the refractorily behaving fission-product elements per unit contamination surface density. The second term is a similar quantity for the volatilely behaving fission product radionuclides. The third term expresses the contribution of the induced activities. For standard conditions the expression is

$$D^* = \sigma_{95} \left[\Sigma\, Y_k \dot{D}_k^* + \Sigma\, Y_k \left(r_{89,95}\right)^{1-b_k} \dot{D}_k^* + \Sigma\, Y_k\, \dot{D}_k^* \right] \quad (19)$$

Units

This section lists the primary quantities of interest in the preceding discussion, together with their designations, and a recommended system of units for consistent use of the equations. Care has been taken to avoid the use of cumbersome units such as "curies" and "gamma curies".

Symbol	Physical Quantity	Units
D (D_i, D_j, D_k, D_R, D_v, D_I)	exposure dose rate	roentgens per second
A	disintegration rate	disintegrations per second
N_j	number of atoms	atoms

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* Although the term "chain" is used, most induced activities decay to stable nuclides in one step.

N_{ij}	photons per disintegration	photons per disintegration
λ	decay constant	per second
E	energy	Mev
σ (σ_j, σ_k)	contamination surface density	σ_j atoms per cm^2 σ_k fissions per cm^2
$\sigma_j \lambda_j$	just as the combined quantity $N\lambda$ (atoms sec^{-1}) is commonly given the unit "disintegrations per second," so the combined quantity $\sigma\lambda$ (atoms cm^{-2} sec^{-1}) has the unit "disintegrations sec^{-1} cm^{-2}."	
d (d_i, d_j, $\dot{D}_k$ (t))	dose rate conversion factors	d_k roentgens per (photon/cm^2) d_j roentgens per (disintegration/cm^2) $\dot{D}_k$(t) (roentgens/sec) per(atom/cm^2)
Y	total chain yield	atoms per fission
G	chain fraction	dimensionless
r	fractionation factor	dimensionless

SIMPLIFIED PRESENTATION

To most effectively dispel misunderstandings about fractionation and its effect on dose rates requires that a technical exposition of the complexities involved be supplemented by dissemination of the information.

The following presentation attempts to achieve maximum simplicity
for the general user of such information with the minimum loss of rigor.

In this presentation we first recommend use of the following
alternative system of units for large scale considerations. They are
obtained from those in the previous paragraph by simply substituting
teracalorie for fission and square mile for square centimeter. The
only quantities affected, together with their alternate units, are
the following

σ   teracalorie per square mile

d   (roentgens/photon) per (teracalorie/square mile)

Y. atoms per teracalorie

If these substitutions are made in the treatment, Equation 19 would
still apply, as in the detailed discussion. Again, the units of curies
and of gamma curies have been successfully avoided.

With these minor modifications, the considerations of the fore-
going section can be summarized for a scientific (but non-specialized)
audience as follows:

   If, at any time after detonation, all the debris from

   a nuclear burst of W teracalories fission yield were uniformly

   spread over W square miles, the resulting dose rate three feet

   above the plane at one hour can be shown to be of the order of a

   roentgen per second. In this situation each radionuclide is said

   to be present at the **same** contamination surface density of "one

14

teracalorie equivalent of that nuclide per square mile"
for the burst in question, even though the quantities of
these radionuclides would differ from nuclide to nuclide
by orders of magnitude when expressed in terms of atoms
or disintegrations per second. In a real deposit of
debris, say from a land-surface burst, the debris will be
fractionated, and the individual radionuclides will be
present at different "teracalorie equivalents" per square
mile. However, even here a large fraction of the fission-
product nuclides will be present at any location in nearly
the same number σ of teracalories per square mile. One of
these nuclides is Zr^{95}, which we will use for a reference
nuclide, and so we will designate this value of σ by σ_{95}.
In addition, the activities induced in the weapon com-
ponents and the soil can be considered to be present at
the same number σ_{95} of teracalorie equivalents per square
mile. Other nuclides will have contamination surface
densities that depart significantly from σ_{95}. For the most
part, these will be fission products, and in local fallout
their contamination surface densities will be less than σ_{95}.
One of these, Sr^{89}, will be taken as a reference of such
volatile behavior. Its contamination surface density at
any point, σ_{89}, can then be used to define the degree of
fractionation ϕ at any point.

15

$$\phi = \log_{10}(\sigma_{89}/\sigma_{95}) \tag{20}$$

It has been found that nuclides of mass chain k have contamination levels related to σ_{95} by expressions such as

$$\log_{10} \sigma_k \approx \log_{10} \sigma_{95} + C_k \phi \tag{21}$$

where C_k is a constant depending on k. The values of σ_{95}, σ_{89} and ϕ will all vary from point to point in the fallout pattern, but at any point their values will remain essentially constant with time if the pattern is not disturbed. Once the values of C_k are established for each mass chain of interest, the resulting exposure dose rate can be expressed, even for fractionated fallout contaminant, by summing the contributions to the exposure dose rate from all the contributors.

Finally, the adoption of this definition of contamination level should do much to clarify the effects of fractionation on dose rates from fallout, to obviate false conclusions arising from a misunderstanding of fractionation effects, and to provide a sound foundation for the treatment of associated problems.

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III. Title.

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fractionated debris be expressed as a contamination surface density multiplied
by the sum of three terms. The first term is the exposure dose rate contribu-
tion of refractorily behaving fission-product nuclides per unit contamination
surface density. The second term is a similar quantity for volatilely behaving
fission-product nuclides. The third term expresses the contribution of the
induced activities.

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