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CALCULATION OF THE CONCENTRATION OF ANY RADIONUCLIDE DEPOSITED ON THE GROUND BY OFFSITE FALLOUT FROM A NUCLEAR DETONATION

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Lawrence Livermore Laboratory

CALCULATION OF THE CONCENTRATION OF ANY RADIONUCLIDE DEPOSITED ON THE GROUND BY OFFSITE FALLOUT FROM A NUCLEAR DETONATION

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ABSTRACT

A method is described to calculate the dry deposition on the ground of any radionuclide in offsite fallout from a nuclear weapons test. The method depends upon the calculation of a source term (microcuries per square meter of fission and activation products), the use of concentration-to-exposure-rate conversion factors, and normalization to a measured external exposure rate. Fractionation is treated explicitly by elimination of a fraction of the refractory elements from the source term. Calculations are in agreement with available measurements.

1. INTRODUCTION

The considerable work on the mechanism of formation and properties of fallout has used data from Pacific Ocean events (coral island or sea water emplacement) and from excavation experiments (Fr70, Kl64). However, about 90% of the Nevada and Utah population's exposure to radioactive fallout from nuclear weapons testing in Nevada during the years 1951 through 1957 was due to 16 devices (An81) detonated on steel towers 300-700 feet above the ground. No data from shots emplaced on steel towers at the Nevada Test Site have been compared to the accepted model because the only data extant were taken for another reason, i.e., to study the uptake of radionuclides by animals living in the fallout pattern (La66). Only some of the data were reported, thus preventing their use in checking the fallout model or these calculations. The Nevada Operations Office of the U.S. Department of Energy has recently acquired the laboratory notebooks containing more of the data. This work describes the comparison of all these data with results derived from the fallout composition model.

In this work I have developed a method to calculate the ground concentration ($\mu Ci/m^2$) of each radionuclide from the external gamma exposure rate given by the fallout pattern. The effects of fractionation are included. The results are compared with field data and interpreted according to the accepted model of fallout formation (Ad60). The calculations agree in detail with the field data; thus, the fractionation behavior of offsite fallout from tower shots within 160 miles of ground zero is well characterized. Uranium and plutonium

data are omitted from this report to keep the results unclassified. This omission is not a significant loss, because the natural uranium content of the desert soil is about 10^6 times that received from fallout and the plutonium concentration in the soil has been measured and reported (Bl77, Ha76).

2. FRACTIONATION

The phenomenon of fractionation is due to both chemical and physical separation of the radionuclides. Chemical separation occurs in the first few minutes. At zero time, everything in the vicinity of the device is vaporized. As the fireball cools to about 3000°C the FeO and soil components form liquid droplets in which the refractory elements dissolve (Ad60). The fireball cools to the melting point of soil and iron oxides, ~1500°C, in about 20 sec (Mi63, St58). The solidified droplets contain the refractory elements, while the volatile elements and their radioactive daughters remain in the gas phase. In 6-8 minutes, when the cloud has cooled to ambient temperature, ~-50°C, the volatile elements (except for Kr and Xe) and their daughters condense on the surface.

Since 1959 our group at Lawrence Livermore National Laboratory, under P. C. Stevenson, has interpreted fractionation phenomena in terms of a two-chemical-phase model of fallout. Heft (He70) and Miller (Mi60) have also used this model. In this work I refer to the two phases as the volatile phase and the refractory phase. The volatile

phase consists of those elements in the gas phase when the liquid droplets solidify. The refractory phase consists of those elements in the liquid droplets when they solidify. The volatile/refractory ratio is the ratio of the amounts of volatile phase to refractory phase, denoted by r(A) for mass chain A.

Consider for the moment only the material vaporized by the detonation. The condensate is in the form of small spheroids and the refractory nuclides are assumed to be distributed uniformly throughout the volume of the droplet (amount varies as r^3). The volatile nuclides are assumed to be distributed uniformly over the surface (amount varies as r^2). The volatile/refractory ratio associated with a given particle will vary as r^{-1}: the smaller the particle, the larger the proportion of volatile phase.

This ideal picture becomes complicated because spheres of debris can attach to each other or to unmelted dust particles which have been entrained by the cloud. The result is a mixing of spherical and irregular particles. This mixing should tend to reduce the strong dependence of the volatile/refractory ratio on the size of particles collected in the field.

The solid particles in the cloud of nuclear debris fall to earth at different velocities and from different altitudes. This process has the effect of separating the debris by particle size along the path of the cloud. Thus, the refractory phase and the volatile phase are partially physically separated, a process taking hours or days

Offsite fallout is characterized by an excess of volatile phase, because the smaller particles tend to be deposited farther from ground zero. The calculation of fractionation effects for offsite fallout is straightforward according to the picture above: remove some of the refractory phase in the calculation. The composition of each phase is determined by the volatility of elements at 1500°C (Appendix 1). The fission product elements of a given mass number are either all volatile or all refractory, except the mass chains 91, 140 and 141. The refractory fractions of these latter chains have been determined at 20 sec postshot (Appendix 2, Sec. A2) and these mass chains partitioned accordingly. The computer program allows removal of any fraction of the refractory phase.

3. CALCULATIONS

The purpose of these calculations is to relate the ground surface concentration ($\mu Ci/m^2$) of every radionuclide to the external gamma-ray exposure levels (mR/hr) from the fallout pattern as a function of time and fractionation behavior. The results of the calculation are the contributions of each of 152 fission products and each of 25 neutron-induced nuclides to the surface deposition and to the external gamma-ray exposure levels. These values and the sums of all values at each decay interval are computed from zero time to 50 years postshot. The entire calculation is outlined in this section and is described in detail in Appendix 2.

The calculation proceeds as follows:

- For each shot calculate the proper distribution of its fission products.
- Define refractory and volatile mass chains and refractory fractions of mass chains 91, 140 and 141.
- Allow for fractionation by removing a fraction of the refractory phase.
- Calculate the curies of each fission product present as a function of time and the total for each decay time.
- Calculate the mR/hr due to each fission product present as a function of time and the total for each decay time.
- Calculate the curies of each neutron-induced nuclide allowing for fractionation and the total for each decay time.
- Calculate the mR/hr of each neutron-induced nuclide and the total for each decay time.
- Sum the total fission product and total neutron-induced nuclide curies.
- Sum the total fission product and total neutron-induced mR/hr.
- Normalize to an external gamma-ray exposure rate of 1 mR/hr at 12 hr postshot by dividing every value by the total mR/hr at 12 hr postshot.
- Fit the total normalized $\mu Ci/m^2$ and mR/hr by the method of least squares to the form

$$\sum_{i=1}^{11} a_i e^{-\lambda_i t}$$

4. RESULTS

Fallout patterns were routinely measured on all shots detonated at the Nevada Test Site from 1951 through 1958. The quantity $(\mu Ci/m^2)/(mR/hr)$ was measured on the fallout from 13 tower shots and three balloon shots from 1953 through 1957 (Ba58, La66, Ra54). In 1957, fallout from four tower shots and one balloon shot were collected, sorted by particle size, and each size fraction analyzed radiochemically (La66). The analytical results from the tower shots provide a detailed check of the calculations described herein.

DECAY OF FALLOUT

The calculated external gamma-ray exposure rates* are shown in Fig. 1. Fractionation affects the shape of the decay very little, but losing half of the refractory elements reduces the relative exposure rate by ~12% and losing all but one tenth reduces the rate by ~25% over the first six weeks.

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* According to H. L. Beck, (Be80) the concentration of fallout varies exponentially with soil depth, z; $C = C_0 e^{-\alpha z}$. He defines relaxation length as $1/\alpha$. All calculations in this work use a relaxation length of 0.16 g/cm^2 unless otherwise specified. See Appendix 2.

Figure 2 shows that the calculated decay for fallout from shot Harry is little different from that presented by Glasstone and Dolan which is "a reasonable average for situations in which the fallout activity arises mostly from fission products." (Gl77, pp. 392, 393, 450)

The best in-situ fallout decay curves were measured in operation Plumbob in 1957 (Di57). The decay of Smoky debris was followed for the longest time. This calculation and data from shot Smoky (Di57) agree well within the precision of the measurements, 10% (Fig. 3). The data taken after 100 hr are generally lower than the calculated values. This may be due to the effects of weathering or because radiation levels under 10 mR/hr were measured with an instrument different from that used at levels over 10 mR/hr.

Data for the fallout patterns were normally taken 6-30 hr postshot and brought to a common time, (Sh59) 12 hr postshot, by the decay law $t^{-1.2}$. Dunning (Du58) proposed a prescription for the decay of fallout which is $t^{-1.2}$ decay for the first week after a detonation, $t^{-1.3}$ for the second week and $t^{-1.4}$ thereafter. Figure 4 shows good agreement between this calculation, the early decay law, and Dunning's prescription.

NUCLIDES ACCOUNTING FOR A MAJOR FRACTION OF EXPOSURE

The 5 mass numbers giving the highest external gamma-ray exposure rates were determined for each of 10 decay times from the calculations of fractionated and unfractionated Harry and Smoky debris. The fission products were the same as those found in the calculations of

unfractionated debris by Freiling, Crocker and Adams (Fr64), which did not include neutron-induced nuclides. In the calculations of the compositions of Harry and Smoky fallout, only two neutron-induced nuclides contributed more than 1% to the external gamma-ray exposure, namely, ^{239}Np from 1 to 15 days, up to 20%, and ^{60}Co from 2 to 15 years, up to 10%. A check of the Nevada Test Site data for 25 Events (tower shots, balloon shots, and air bursts) showed (though not for every Event) that contributions of more than 1% to the total gamma-ray levels were due to only three neutron-induced nuclides, ^{60}Co, ^{237}U and ^{239}Np.

CURIES OF FISSION PRODUCTS PER KILOTON OF FISSION YIELD

From information given by Glasstone and Dolan (Gl77, p. 390), one can deduce that 1 kiloton of fission produces about 1.5×10^7 curies of fission products 24 hr postshot. The calculation in this work should be considered as an update. The calculations are summarized in Table 1.

Table 1. Calculated fission products (Ci/kt) of five Nevada Test Site tower shots, 12 and 24 hr postshot.

Shot	12 hr postshot	24 hr postshot
Annie	2.84×10^7	1.12×10^7
Bee	2.82×10^7	1.17×10^7
Galileo	2.72×10^7	0.98×10^7
Harry	2.87×10^7	1.10×10^7
Smoky	2.79×10^7	1.25×10^7
Average	$(2.81 \pm 0.06) \times 10^7$	$(1.12 \pm 0.10) \times 10^7$

VALUES OF $(\mu Ci/m^2)/(mR/hr)$

From information given in Glasstone and Dolan (Gl77, pp. 390, 394), one can deduce the value of approximately 90 $(\mu Ci/m^2)/(mR/hr)$ at 24 hours postshot 1 m above 1 kt of unfractionated, unshielded fission products spread uniformly over a 1 km^2 plane surface. The relevant calculation was repeated for the debris from the five shots listed in Table 1, incorporating the latest values of the correction factors, (Be80) $\mu Ci/m^2$ to mR/hr. The results gave an average value of 126 $\pm$ 3 $(\mu Ci/m^2)/(mR/hr)$ at 24 hr postshot 1 m above unfractionated, unshielded fission products spread uniformly over an infinite plane surface. This new value is considerably more accurate and precise than the one deduced from the information given in Glasstone and Dolan (Gl77, pp. 390, 394). The marked differences in neutron spectra and fissioning species of these five shots affect this quantity very little, because the vast majority of the external gamma-ray exposure is due to the fission products at the peaks of the distribution curve. The yields of these nuclides change little with changing fissioning nuclides and neutron spectra.

Values of $(\mu Ci/m^2)/(mR/hr)$ were measured from the fallout of 13 tower shots and 3 balloon shots (Ba58, La66, Ra54). The values for the tower shots were calculated with the assumption that only half of the refractory elements were present. This assumption lowers the value of $(\mu Ci/m^2)/(mR/hr)$ by about 7%. The debris from the balloon shots was assumed to be unfractionated. Surface roughness effects were simulated by a relaxation length of 0.16 g/cm^2. The results are shown in Table 2.

Table 2. Measured and calculated values of $(\mu Ci/m^2)/(mR/hr)$ for offsite fallout from Nevada Test Site tower and balloon shots, 12 hr postshot.

Shot Name	Year	Measured	Calc.
Ray	1953	280 ± 100	149
Simon	1953	160 ± 30	137
Tesla	1955	130 ± 60	139
Apple 1	1955	160 ± 110	132
Met	1955	60 ± 30	132
Apple II	1955	100 ± 40	132
Boltzmann	1957	100 ± 50	130
Priscilla[a]	1957	28 ± 14	139
Hood[a]	1957	22 ± 4	139
Diablo	1957	120 ± 50	136
Shasta	1957	130 ± 50	137
Smoky	1957	120 ± 50	133
Galileo	1957	110 ± 60	147
Fizeau	1957	100 ± 30	130
Newton[a]	1957	19 ± 9	145
Whitney	1957	140 ± 50	140

[a]Balloon shot.

The calculations agree with the rather imprecise measurements for all the data from tower shots except for shots Ray and Met. The measurements on fallout from balloon shots are lower than the calculated figures by a factor of about five to eight. There is no single nuclide of the peak fission products or major activation product that can produce such a result. Therefore, I conclude that the counting samples used to measure $\mu Ci/m^2$ from all balloon shots contained only about 20% of the amount of radionuclides that should have been present, and, consequently, the samples are not representative of the surface deposit.

FRACTIONATION OF TOWER SHOT DEBRIS

Fallout collected from four tower shots in 1957 was sorted into particle size fractions by wet sieving (La66). Before radiochemical analysis, the beta disintegration rate for each fraction was determined for $^{89,90}Sr$, ^{91}Y, ^{95}Zr, $^{103,106}Ru$, ^{136}Cs, ^{140}Ba, and $^{141,144}Ce$. The results for each size fraction were reported as percentage of the total beta activity in that size cut due to each nuclide, but the fraction of the total beta activity of each size cut was not reported in La66. Therefore, these published data could not be used for comparison with calculations of the radionuclide composition of undisturbed fallout. The original data sheets were recently made available by the Nevada Operations Office of the Department of Energy. I synthesized the radionuclide composition measured for each undisturbed fallout sample from the information in these data sheets

and La66 and calculated each radionuclide's atom ratio to ^{95}Zr at zero time. The ratios of ^{89}Sr, ^{91}Y, ^{136}Cs and ^{141}Ce to ^{95}Zr were plotted against the ^{140}Ba to ^{95}Zr ratio (Fig. 5).

In this work I have made the following assumptions:

- Volatile elements do not fractionate from each other.
- Refractory elements do not fractionate from each other.
- The composition of offsite fallout can be represented by the volatile phase and a fraction of the refractory phase.

It follows directly from these assumptions that the atom ratio of a volatile mass chain to a refractory mass chain (e.g., ^{95}Zr) is a linear function of any other such volatile mass chain atom ratio to the same refractory mass chain. The line that plots this function must pass through the point representing unfractionated debris. The data show this linear behavior and the ^{91}Y, ^{136}Cs, ^{140}Ba, and $^{141,144}Ce$ agree, within experimental error, with the values calculated for unfractionated debris (solid circles in Fig. 5). However, the ^{89}Sr data are low by a factor of about five. Therefore, no conclusions have been drawn from these data. Ruthenium data have been omitted because they appear to be low, probably because of the intractable chemical behavior of that element.

It should be noted that Freiling (Fr61, Fr63) has shown that such ratios in fractionated debris are power functions, not linear functions of each other. His ratios range over an order of magnitude above and below the unfractionated value. The Nevada tower shot data, on the other hand, are close to the unfractionated values, and constitute a special case in which the simplifying assumptions lead to a linear relation.

The volatile/refractory ratio of a fallout sample can be deduced from the ratios of the amount of volatile or partly volatile fission product mass chains to the amount of a refractory fission product mass chain.

The number of atoms, A, of a given mass chain in a sample is

$$A = R(A) + V(A),$$

where $R(A)$ = number of atoms of mass chain A refractory at 20 sec postshot,

$V(A)$ = number of atoms of mass chain A volatile at 20 sec postshot.

Dividing by the number of atoms, R, of a mass chain containing only refractory members, and transposing, we obtain

$$\frac{V(A)}{R} = \frac{A}{R} - \frac{R(A)}{R} \quad .$$

Refractory masses do not fractionate; therefore,

$$\frac{V(A)}{R} = \frac{A}{R} - \left[\frac{R(A)}{R}\right]_u \quad ,$$

where the subscript u denotes the ratio calculated for unfractionated debris. The value of $[R(A)/R]_u$ and $[V(A)/R]_u$ are known from the fissioning nuclides and neutron spectrum of the detonation (Ne73). Dividing the above equation by $[V(A)/R]_u$ gives $r(A)$, the volatile/refractory ratio for mass chain A.

$$r(A) = [V(A)/R]/[V(A)/R]_u = \left\{\frac{A}{R} - \left[\frac{R(A)}{R}\right]_u\right\}/[V(A)/R]_u = 1/f$$

The quantity $r(A)$ is the reciprocal of the quantity, f, the fraction of refractories present in the sample and is equivalent to Edvarson's

(Ed59) f_{A-95} only when the mass chain A is 100% volatile, e.g., ^{136}Cs. Equations for specific nuclides are in Appendix 3.

The values of r(91), r(136), r(140), and r(141) generally agree, within experimental error, in the 25 samples measured. These values are averaged for each sample; the averages appear in Table 3. The values of r(A) are plotted against radial distance from ground zero (Fig. 6) and arrival time (Fig. 7). Clearly, r(A) increases with time and distance from ground zero. The value of r(A) = 2 (half refractories present) can be used to characterize the nuclide composition of the offsite fallout within 160 miles from ground zero.

The relative amount of volatile phase increases with distance, indicating that the volatile phase is relatively enriched on the smaller particles, even though there was a considerable amount of unmelted dust entrained in the cloud. This correlation is consistent with the concept that the refractory nuclides are contained in the bulk of the particles formed by condensation and the volatile nuclides are deposited on the surface. As time and distance from ground zero increase, the size of the fallout particles decreases.

Values of r(A) for each size fraction were averaged for each shot. The average values are shown in Table 4 and in Fig. 8.

Table 3. Volatile/refractory ratio (volatile to refractory phase relative amounts) in fallout at various distances from Nevada Test Site tower shots.

Shot	Miles from G.Z.	Arrival time (hr)	Vol./ref. ratio
Diablo	16	3.2	1.2 ± 0.2
	20	3.6	1.3 ± 0.3
	40	5.1	1.6 ± 0.3
	60	6.7	1.7 ± 0.3
Shasta	15	0.7	1.3 ± 0.1
	25	2.7	1.9 ± 0.2
	25	1.6	1.5 ± 0.2
	32	1.7	2.0 ± 0.4
	34	2.3	1.6 ± 0.1
	35	1.4	2.8 ± 0.1
	41	4.1	2.2 ± 0.5
	42	3.0	2.5 ± 0.3
	43	4.8	2.1 ± 0.1
	44	2.8	1.7 ± 0.2
	44	3.6	2.1 ± 0.2
	45	4.7	2.3 ± 0.3
	47	2.0	2.1 ± 0.2
	81	5.2	2.1 ± 0.1
Smoky	5	0.2	1.1 ± 0.4
	25	1.1	1.5 ± 0.5
	48	3.2	1.6 ± 0.5
	65	4.2	2.1 ± 0.5
	99	4.6	2.1 ± 0.2
	158	5.6	3.5 ± 1.3

Table 4. Volatile/refractory ratio (volatile to refractory phase relative amounts) as a function of particle size in fallout from Nevada Test Site tower shots.

Particle size (μm)	Tower shot		
	Diablo	Shasta	Smoky
500-1000	--	--	1.5 ± 0.5
350-500	--	1.6 ± 0.6	1.5 ± 0.3
297-350	1.2 ± 0.1	1.6 ± 0.9	1.4 ± 0.5
250-297	1.3 ± 0.1	1.4 ± 0.5	1.8 ± 0.4
210-250	1.5 ± 0.4	1.6 ± 0.4	1.4 ± 0.3
177-210	1.3 ± 0.1	1.5 ± 0.4	1.6 ± 0.5
149-177	0.4 ± 0.4	1.5 ± 0.4	1.9 ± 1.0
125-149	1.2 ± 0.2	1.9 ± 0.6	2.1 ± 1.2
105-125	1.4 ± 0.7	1.8 ± 0.5	1.5 ± 0.4
88-105	1.2 ± 0.2	2.1 ± 0.6	2.4 ± 0.9
44-88	1.4 ± 0.3	1.7 ± 0.3	2.3 ± 0.8
<44	1.7 ± 0.1	2.2 ± 0.4	2.5 ± 0.4

The pertinent physical properties of fallout (Gl77, p. 411) from a tower shot are summarized as follows:

- Size--largest dimension between a few micrometers and a few hundred micrometers.

- Shape--both spheroids and irregular, larger particles with small spheroids attached.
- Distribution of radioactivity--some particles radioactive throughout, some particles radioactive only on or near the surface.

These properties indicate that after the liquid particles solidify, they begin to agglomerate with the irregularly shaped dust particles and with each other. The daughters of the nuclides in the gas phase are continually being deposited on the surface of all particles until the particles fall out of the gas cloud. The refractory elements in a given fallout sample are acquired partially by agglomeration, but the volatile elements all are acquired by condensation on a solid surface. The spheroidal particles in a sample should show a strong inverse relation between r(A) and particle size. On the other hand, the irregular particles in a sample, having been sieved, contain variable (indeterminate) relative amounts of solid particles and gaseous condensate. Therefore, in a sample containing both spheroidal and sieved irregular particles, the relation of r(A) to particle size will depend on the relative amounts of spheroidal and irregular particles in the sample.

The inverse relation of r(A) and particle size is easily seen in the Smoky data. There is less of a trend in the Shasta data and still less in the Diablo data.

5. CONCLUSIONS

A method is described whereby the concentration of any radionuclide deposited on the ground can be derived from the exposure rates shown in the fallout pattern; fractionation is included specifically. Field data are consistent with calculations. The radionuclide composition of fallout within 160 miles of ground zero can be adequately described as having all the volatile nuclides and half of the refractory nuclides present in unfractionated debris. The calculated surface concentrations ($\mu Ci/m^2$) of volatile nuclides are as accurate as the meter readings, which are accurate to $\pm$20%. The calculated surface concentrations of refractory nuclides are accurate to $\pm$30%.

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APPENDIX 1

Table A1. Division of elements and masses important to fallout.

Volatile at 1500°C		Refractory at 1500°C	
Elements			
Ge	Cd	Be	Nb
As	In	Na	Ba
Se	Sn	Mn	Rare Earths
Br	Sb	Fe	Th
Kr	Te	Co	U
Rb	I	Cu	Np
Mo	Xe	Sr	Pu
Tc	Cs	Y	Am
Ru	W	Zr	Cm
Rh	Au		
Pd	Pb		
Ag			
Mass Numbers			
75 - 90		7 - 74	
101 - 139		92 - 100	
181 - 203		142 - 180	
		204 - 242	

APPENDIX 2

DETAILS OF THE CALCULATION

The purpose of these calculations is to relate the surface concentration ($\mu Ci/m^2$) of every radionuclide significant to the external gamma-ray exposure levels (mR/hr) as a function of time and fractionation behavior.

A. Fission Products

A total of 152 fission products and 25 neutron-induced nuclides were included in the computerized calculations described stepwise below.

1. Unfractionated fission product distribution

$$FY(A) = \sum_i FY(A)_i \cdot a_i \quad ,$$

where $FY(A)$ = fission yield of mass chain A for a given event,

$FY(A)_i$ = fission yield of mass chain A from ^{233}U, ^{235}U, ^{238}U or Pu by fission spectrum or 14 MeV neutrons (Ne73) (table in computer program),

a_i = fraction of total fissions due to a given fission type (input data).

2. Fractionated fission product distribution for offsite fallout. For offsite fallout, remove some of the refractory phase (input data).

   Fractionated fission yield = FY(A) • b(A),

   where b(A) = 1.0 for volatile mass chains 75-90 and 101-139.

   b(A) = fraction of refractory elements present, f, (input data), for mass chains 92-100 and 142-180.

   b(A) = 0.75 + 0.25f for mass 91

   0.7 + 0.3f for mass 140

   0.33 + 0.67f for mass 141.

   The independent fission yields of the 91, 140 and 141 chains change with fission split only by small amounts under the conditions of this study; therefore, the above assumptions were made for all shots.

3. Relative fractionated fission product distribution

   The decay matrix (see 4 below) was calculated by the ORIGEN computer program (RSIC79) written at Oak Ridge National Laboratory to calculate the formation and decay of radionuclides in reactor fuel elements. Results of that calculation were the number of curies of each fission product from a 1-sec run at a power level of 4.185 MW or 10^6 calories or 10^{-6} kt or 1.43×10^{17} fissions in a fuel

element composed of 1 kg each of ^{235}U, ^{238}U, and Pu. The fission product yield distribution was not that of a nuclear device, so the values in this decay matrix were corrected by the factor R(A).

$$R(A) = \frac{FY(A) \cdot b(A)}{FYO\ (A)} \quad ,$$

where R(A) = relative fractionated fission yield

FYO(A) = fission yield used by the ORIGEN computer program.

4. Curies of fractionated fission products

The ORIGEN computer program followed the decay of each of the 152 fission products from zero time to 50 years postshot. There were 30 decay intervals, 10 from zero time to 21 hours, 10 from zero time to 300 days, and 10 from zero time to 50 years. The elements of this matrix were then multiplied by 10^6 to give the correct values for each fission product for 1 kt of fission yield. This new matrix was incorporated into the computer program for this work. (See $D(A)_t$, below.) The curies of each fractionated fission product as a function of time was given by

$$CI(A)_t = D(A)_t \cdot R(A) \cdot KT,$$

where $CI(A)_t$ = curies of mass chain A (fractionated) present

as a function of time, t, for a given shot,

$D(A)_t$ = curies of mass chain A for 1-kt fission yield as a function of time, calculated by the ORIGEN computer program with the ^{233}U, ^{235}U, ^{238}U and $^{239,240}Pu$ data omitted (table in computer program).

KT = kilotons of fission yield (input data).

5. External gamma-ray exposure rates

Assume that the fallout is distributed uniformly over 1 square kilometer. The external gamma-ray exposure rate for each individual nuclide measured 1 meter above ground level is given by

$$MR(A)_t = CI(A)_t \cdot C(A),$$

where $MR(A)_t$ = mR/hr measured 1 meter above ground level due to mass chain A as a function of time.

C(A) = conversion factor (Be80), $(mR/hr)/(Ci/km^2)$, (table in computer program), the value is defined by specifying the relaxation length* (input data).

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* According to H. L. Beck, (Be80) the concentration of fallout varies exponentially with soil depth, z; $C = C_0 e^{-\alpha z}$. He defines relaxation length as $1/\alpha$.

6. Fractionated neutron-induced nuclides, curies

Each neutron-induced nuclide was put into the original ORIGEN calculation as 1.43×10^{17} atoms or one atom produced per fission. The resulting curies appeared in the decay matrix referred to in sections 3 and 4 above. The curies of fractionated, neutron-induced nuclides was calculated by

$$I(A)_t = (A/F) \cdot b(A) \cdot D(A)_t \cdot KT \ ,$$

where $I(A)_t$ = fractionated curies of mass A as a function of time,

A/F = atoms per fission of mass A (input data),

b(A) = 1.0 for volatile masses 181-203,

b(A) = fraction of refractory elements present, f, (input data) for refractory masses 7-74 and 204-242--note that there are no data for any volatile nuclide in these mass ranges,

$D(A)_t$ = curies of mass A as a function of time, assuming a production of 1 atom per fission.

7. Fractionated neutron-induced nuclides, mR/hr

$$IMR(A)_t = I(A)_t \cdot C(A)$$

where $IMR(A)_t$ = mR/hr due to mass A measured 1 m above the ground as a function of time.

8. Normalization to external gamma-ray exposure rate of 1 mR/hr at 12 hr postshot

The contours of the fallout pattern are drawn with the field readings corrected to a time 12 hr postshot (Di57). Therefore, all values, curies and mR/hr, are normalized to an exposure rate of 1 mR/hr at 12 hr postshot. The total exposure rate is given by

$$T(t) = \sum_A MR(A)_t + \sum_A IMR(A)_t ,$$

where $T(t)$ = Total mR/hr as a function of time, assuming debris is uniformly spread over 1 km^2.

Normalized values are calculated by the equations that follow.

Normalized mR/hr of each fission product:

$$NMR(A)_t = MR(A)_t/T(12),$$

where $T(12)$ = Total mR/hr at H + 12 hours assuming debris is uniformly spread over 1 km^2 (normalization factor).

Normalized mR/hr of each neutron-induced nuclide:

$$NIMR(A)_t = IMR(A)_t/T(12).$$

Normalized mR/hr of total external gamma-ray exposure rate:

$$NMR_t = T(t)/T(12).$$

Normalized fission product curies:

$$NCI(A)_t = CI(A)_t/T(12).$$

Normalized neutron-induced nuclide curies:

$$NI(A)_t = I(A)_t/T(12).$$

Normalized total curies:

$$NCI_t = \sum_A CI(A)_t + \sum_A I(A)_t \ /T(12) \ .$$

9. Least squares fit of totals

The computer program generates an input file for a second computer program which fits, by the method of least squares, the value of the normalized mR/hr and total curies to the following expression:

$$\sum_{i=1}^{11} a_i e^{-\lambda_i t}$$

The values of λ_i are fixed and the values of the 11 a_i are fitted to the data.

APPENDIX 3

EQUATIONS FOR VOLATILE/REFRACTORY RATIO FOR SPECIFIC NUCLIDES

The ^{136}Cs is 100% volatile 20 seconds postshot.

$$r(136) = \left[\frac{A(136)}{R(95)}\right] / \left[\frac{A(136)}{R(95)}\right]_u$$

The 91 chain, A(91), is 25% refractory 20 seconds postshot.

$$r(91) = \left\{\frac{A(91)}{R(95)} - 0.25\left[\frac{A(91)}{R(95)}\right]_u\right\} / 0.75\left[\frac{A(91)}{R(95)}\right]_u$$

The 140 chain, A(140), is 30% refractory 20 seconds postshot.

$$r(140) = \left\{\frac{A(140)}{R(95)} - 0.3\left[\frac{A(140)}{R(95)}\right]_u\right\} / 0.7\left[\frac{A(140)}{R(95)}\right]_u$$

The 141 and 144 chains are reported together in the field data (La66), i.e., β dpm in the cerium fraction at 30 days postshot. The 144 chain is 100% refractory with the ^{144}Ce and ^{144}Pr in equilibrium. Therefore, we must subtract twice the calculated $^{144}Ce/^{95}Zr$ ratio from the measured ratio.

$$r(141) = \left\{\frac{A(Ce)}{R(95)} - \left[0.67\,\frac{A(141)}{R(95)} + \frac{2A(144)}{R(95)}\right]_u\right\} / 0.33\left[\frac{A(141)}{R(95)}\right]_u$$

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FIGURE CAPTIONS AND TABLE TITLES

FIG. 1. Relative external gamma-ray exposure rate vs. time calculated for shot Harry.

FIG. 2. Comparison of the relative external gamma-ray exposure rates calculated for shot Harry with those reported by Glasstone and Dolan (Gl77, pp. 392, 393, 450).

FIG. 3. Comparison of the relative external gamma-ray exposure rates calculated for shot Smoky with field measurements of Smoky fallout.

FIG. 4. Comparison of the relative external gamma-ray exposure rates calculated for shot Smoky with Dunning's prescription (Du58).

FIG. 5. Zero time atom ratios of volatile fission product mass chains to ^{95}Zr in offsite fallout from Nevada tower shots Diablo, Shasta and Smoky. The solid circles represent unfractionated debris.

FIG. 6. Volatile/refractory ratio (volatile to refractory phase relative amounts) in the fallout from tower shots Diablo, Shasta and Smoky as a function of distance from ground zero.

FIG. 7. Volatile/refractory ratio (volatile to refractory phase relative amounts) in the fallout from tower shots Diablo, Shasta and Smoky as a function of arrival time.

FIG. 8. Volatile/refractory ratio (volatile to refractory phase relative amounts) in the fallout from tower shots Diablo, Shasta and Smoky as a function of (particle size)$^{-1}$.

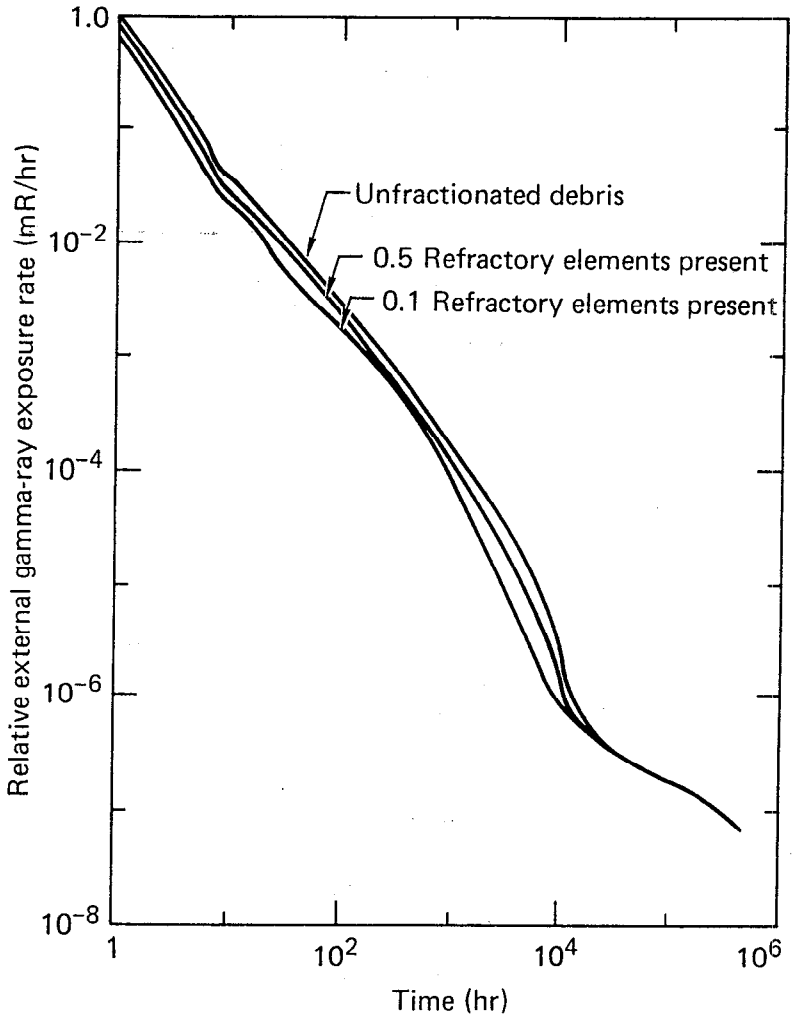
Table 1. Calculated fission products (Ci/kt) of five Nevada Test Site shots, 12 and 24 hr postshot.

Table 2. Measured and calculated values of (μCi/m^2)/(mR/hr) for offsite fallout from Nevada Test Site tower and balloon shots, 12 and 24 hr postshot.

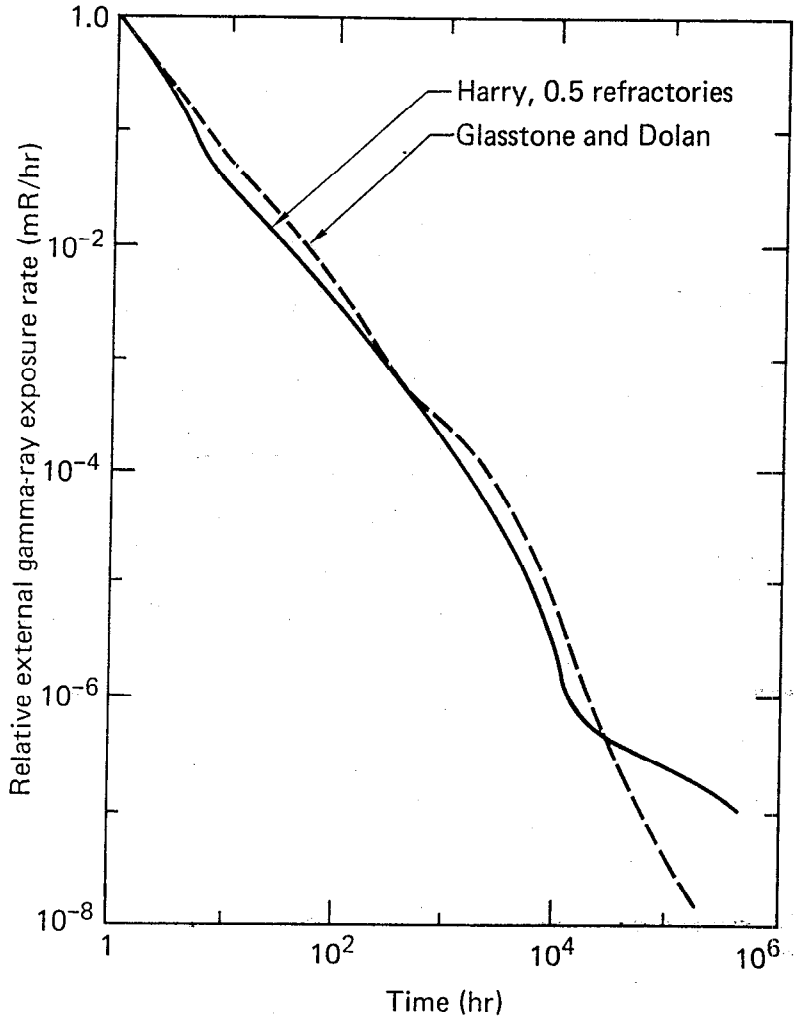
Table 3. Volatile/refractory ratio (volatile to refractory phase relative amounts) in fallout at various distances from Nevada Test Site tower shots.

Table 4. Volatile/refractory ratio (volatile to refractory phase relative amounts) as a function of particle size in fallout from Nevada Test Site tower shots.

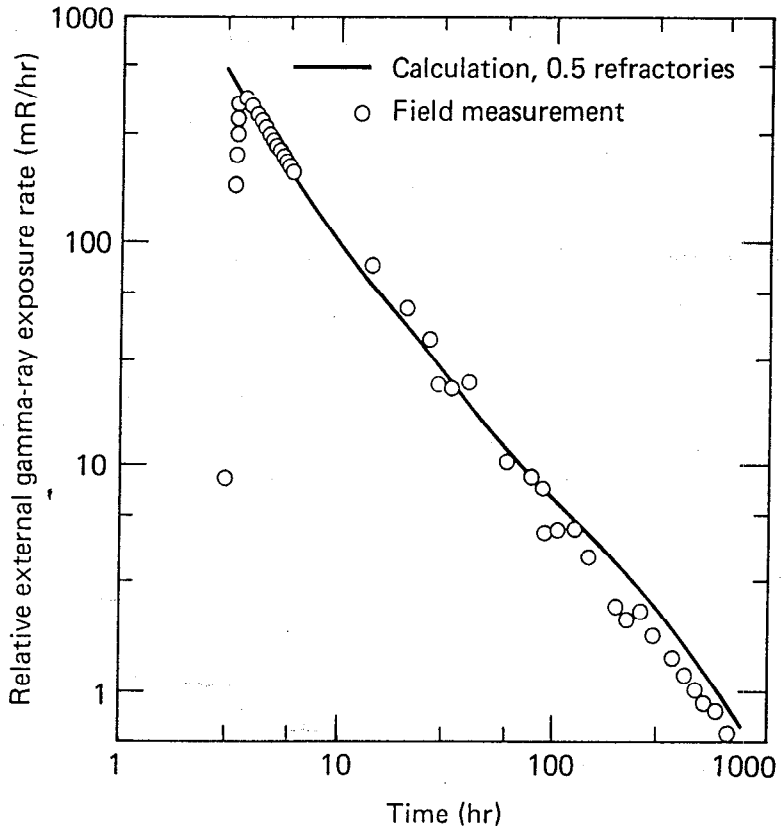
Table A1. Division of elements and masses important to fallout.


1.0
10^{-2}
10^{-4}
10^{-6}
10^{-8}
Relative external gamma-ray exposure rate (mR/hr)
Unfractionated debris
0.5 Refractory elements present
0.1 Refractory elements present
1
10^2
10^4
10^6
Time (hr)

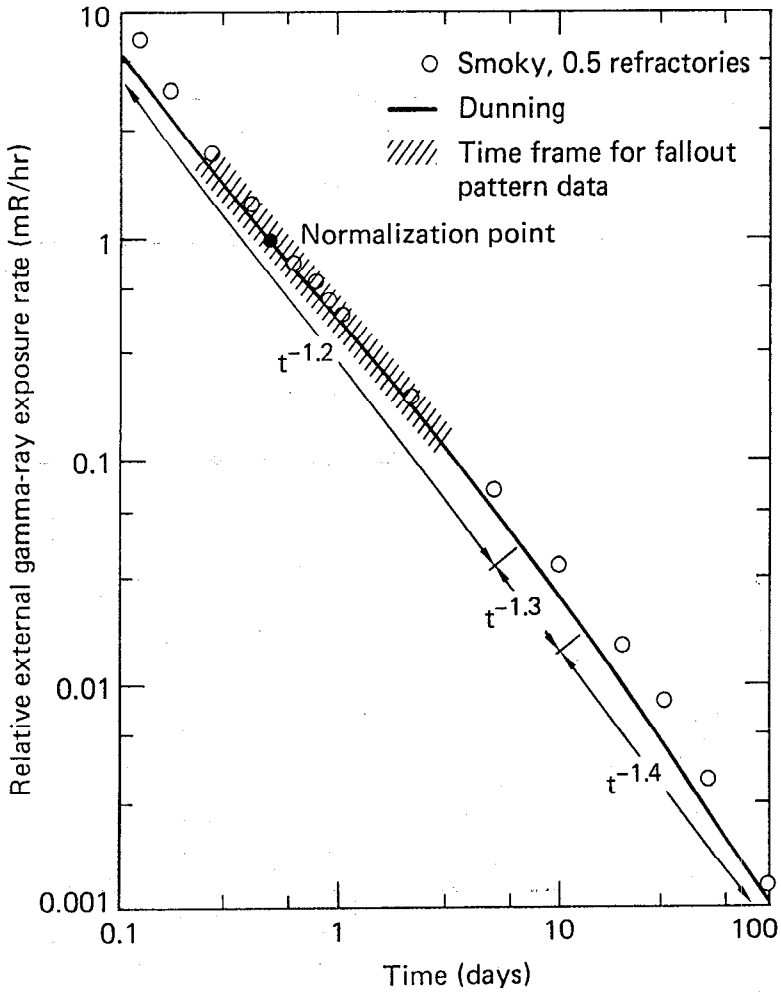
Hicks - Fig. 1


1.0
10^{-2}
10^{-4}
10^{-6}
10^{-8}
Relative external gamma-ray exposure rate (mR/hr)
Harry, 0.5 refractories
Glasstone and Dolan
1
10^2
10^4
10^6
Time (hr)

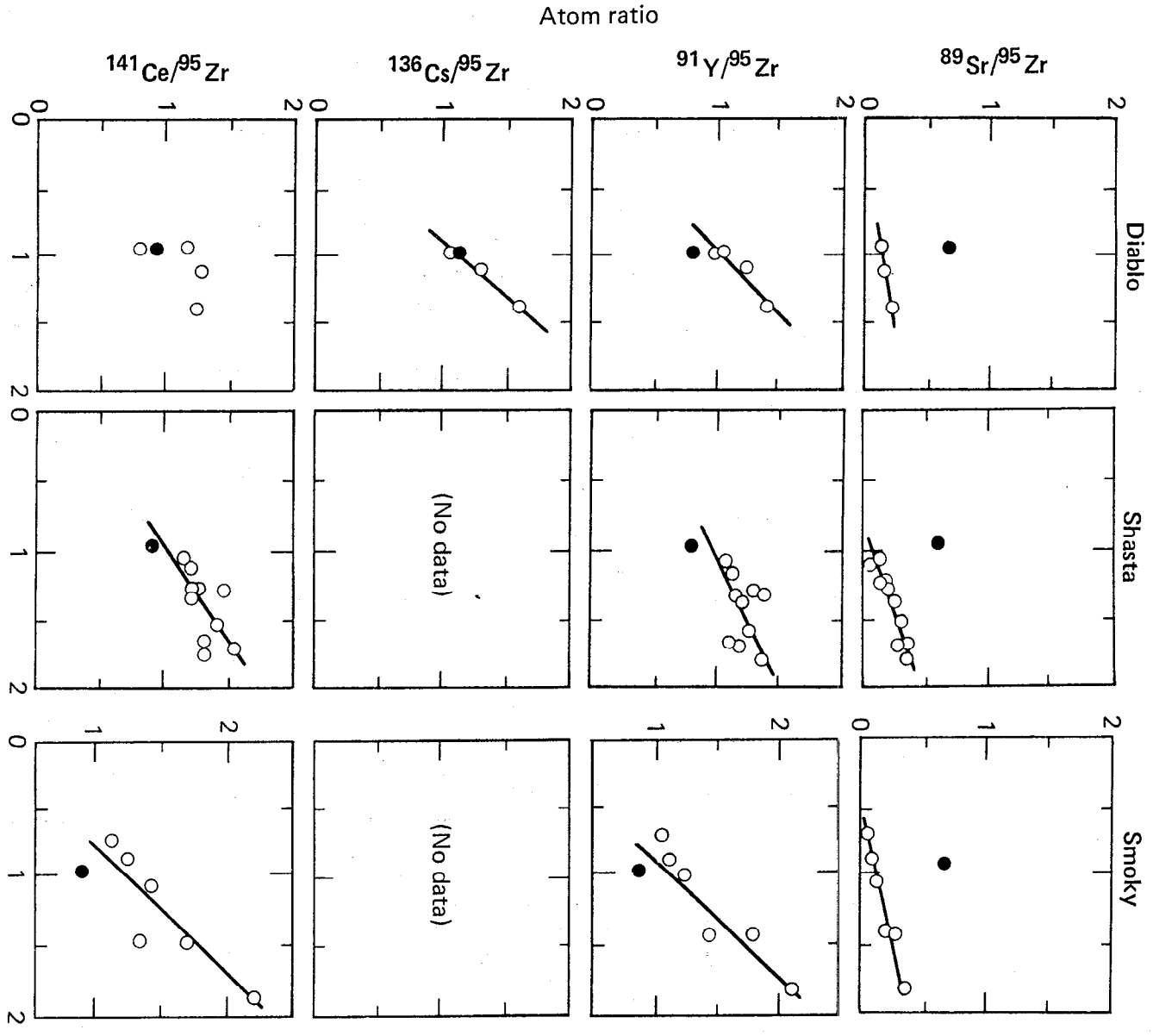
Hicks - Fig. 2


Calculation, 0.5 refractories
Field measurement
Relative external gamma-ray exposure rate (mR/hr)
1000
100
10
1
1
10
100
1000
Time (hr)

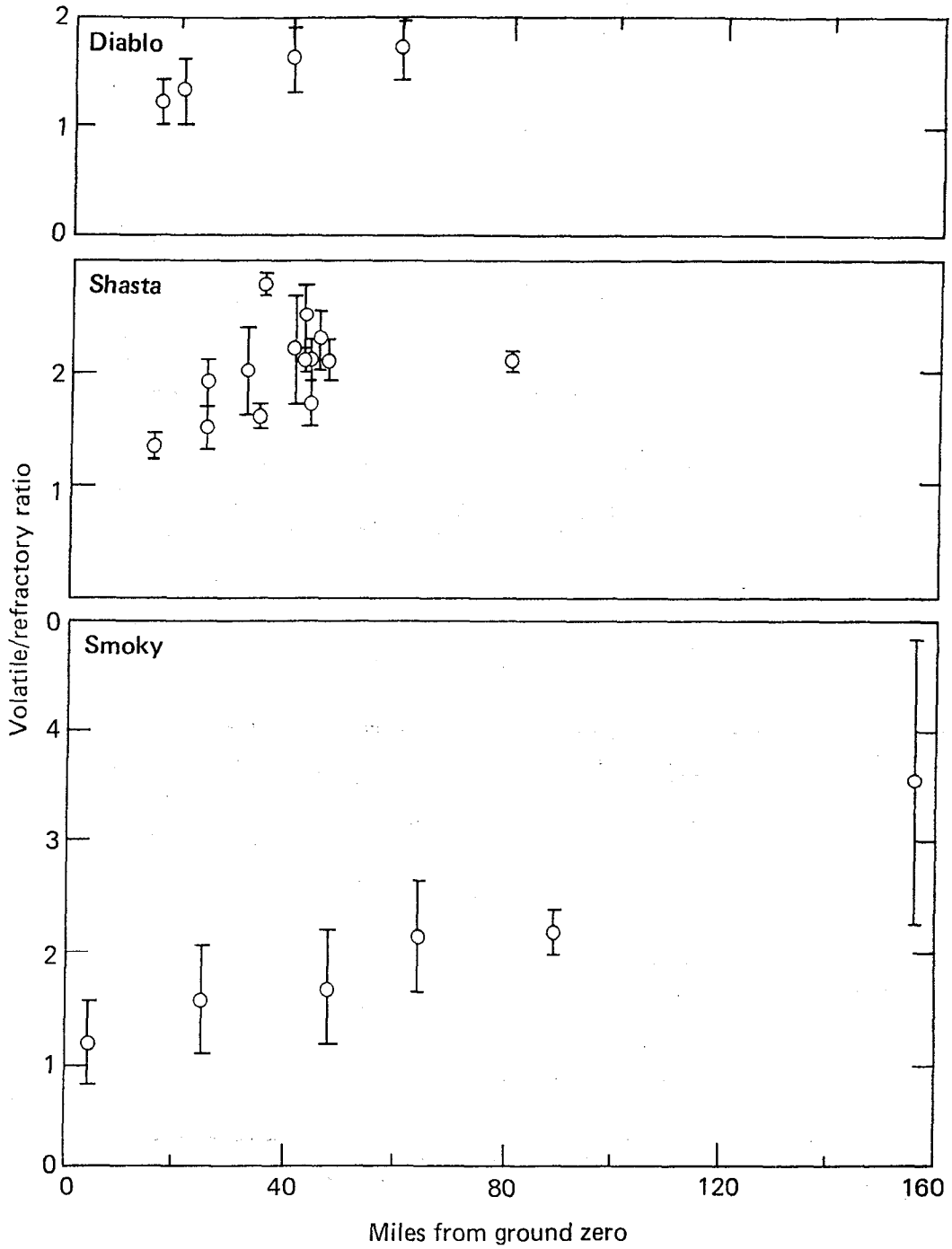
Hicks - Fig. 3


Smoky, 0.5 refractories
Dunning
Time frame for fallout pattern data
Normalization point
$t^{-1.2}$
$t^{-1.3}$
$t^{-1.4}$
Relative external gamma-ray exposure rate (mR/hr)
10
1
0.1
0.01
0.001
0.1
1
10
100
Time (days)

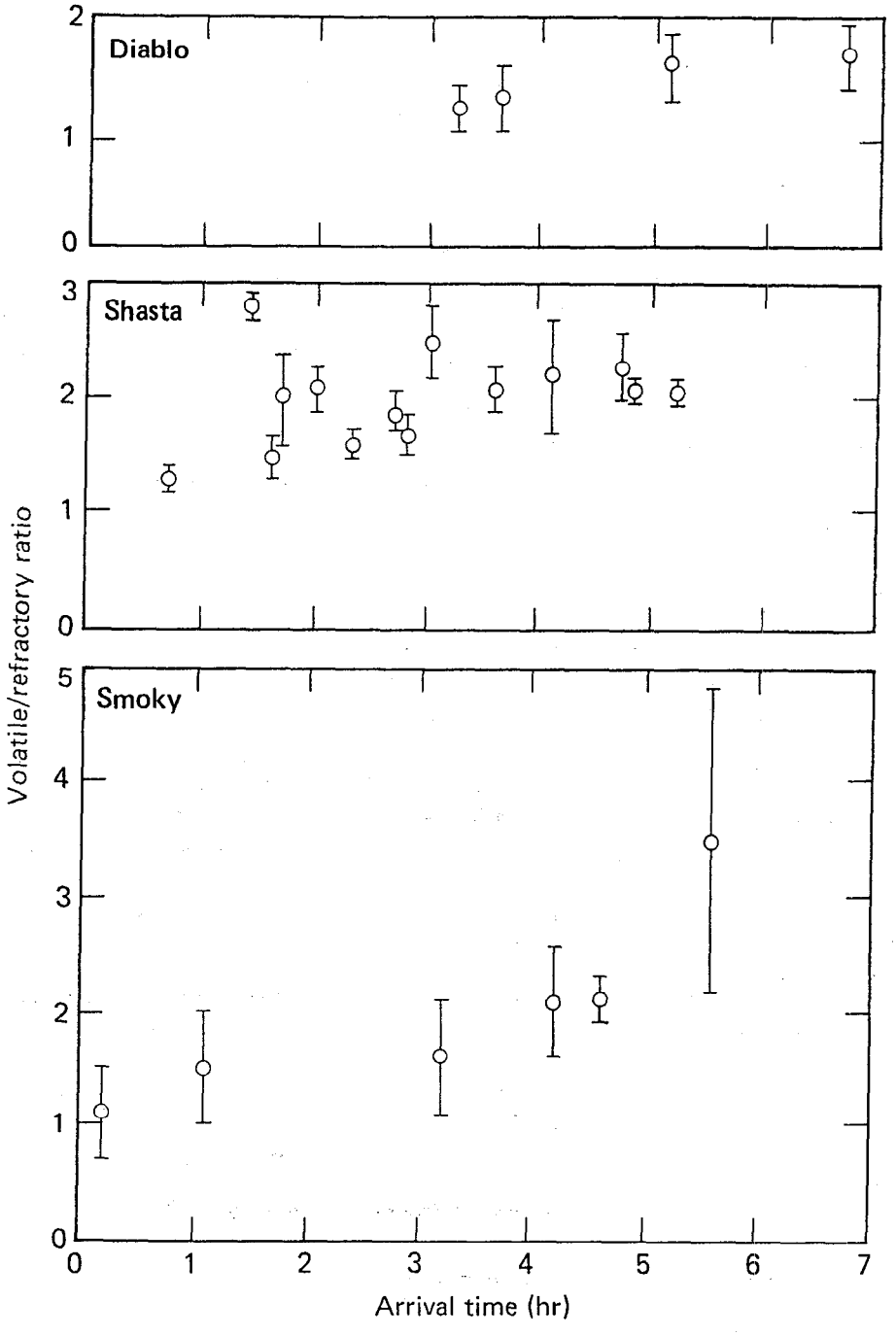
Hicks - Fig. 4


Atom ratio
$^{89}Sr/^{95}Zr$
$^{91}Y/^{95}Zr$
$^{136}Cs/^{95}Zr$
$^{141}Ce/^{95}Zr$
Diablo
Shasta
Smoky
(No data)
(No data)
$^{140}Ba/^{95}Zr$ atom ratio

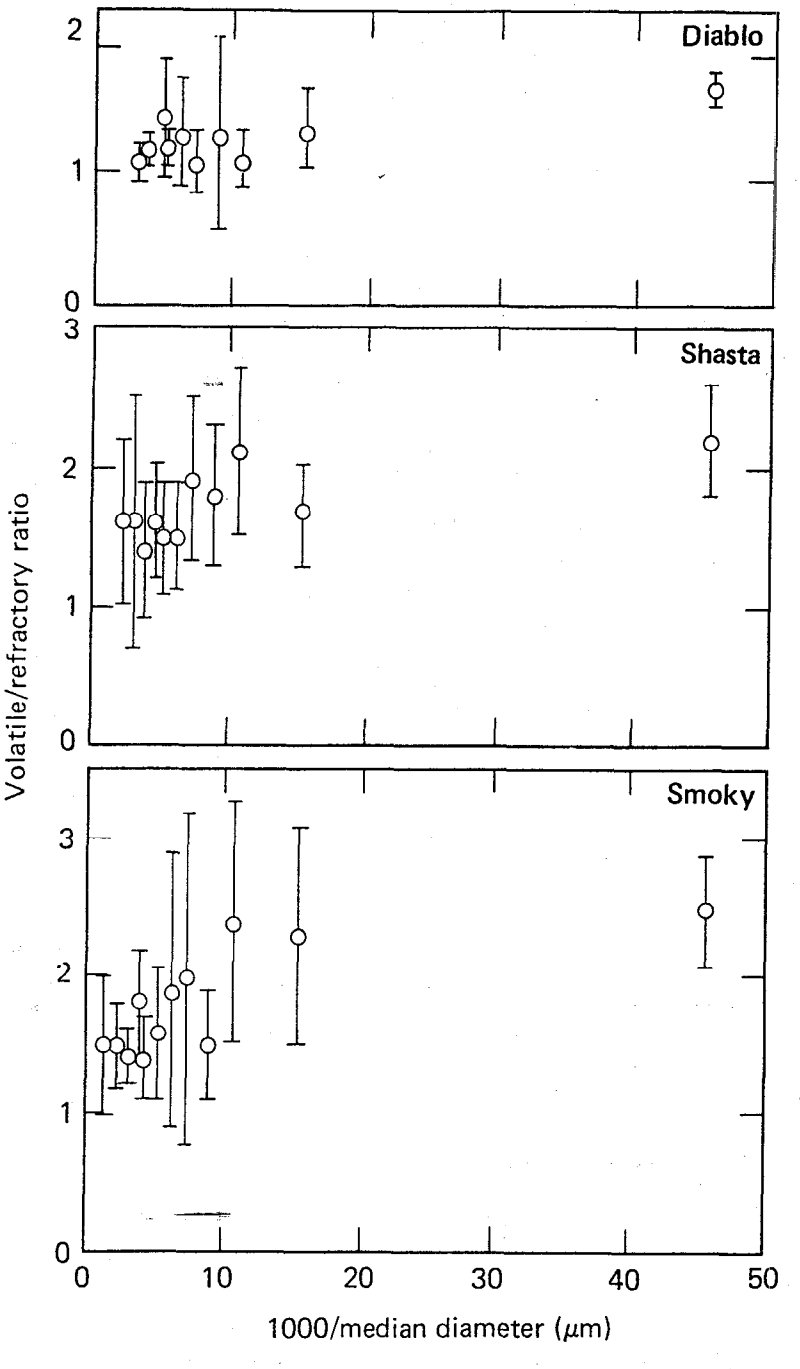
Hicks - Fig. 5


Diablo
Shasta
Smoky
Volatile/refractory ratio
Miles from ground zero
0
40
80
120
160

Hicks - Fig. 6


Diablo
Shasta
Smoky
Volatile/refractory ratio
Arrival time (hr)
0
1
2
3
4
5
6
7

Hicks - Fig. 7


Diablo
Shasta
Smoky
Volatile/refractory ratio
1000/median diameter (μm)
0
1
2
3
10
20
30
40
50

Hicks - Fig. 8