

## A fractionation model based on three lognormal particle size distributions (Postprint)

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### Abstract

In this paper, a new model is proposed to calculate distribution of fission products in particles of different sizes. The model sensitivity to the effective volume and mass of vaporized soil particles is examined. Compared with other fractionation models, the new method has a much better performance in calculating  $r_{89,95}$ , but the calculated cumulative activity fraction for particles in diameters over 100  $\mu\text{m}$  is in between the results using the F-T and G-X models. It is concluded that in a near surface nuclear explosion radioactivity is mainly distributed in soil particles which have not been vaporized, and according to the Henry's law and ideal gas law,  $r_{89,95}$  may vary in larger particles when effective volume of the fireball is changed.

### Full Text

### Preamble

#### A Fractionation Model Based on Three Lognormal Particle Size Distributions

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**Abstract:** This paper proposes a new model for calculating the distribution of fission products across particles of varying sizes. The model's sensitivity to the

effective volume and mass of vaporized soil particles is examined. Compared with other fractionation models, the new method demonstrates substantially improved performance in calculating  $r_{89,95}$ , though the calculated cumulative activity fraction for particles larger than 100  $\mu\text{m}$  falls between the results obtained using the F-T and G-X models. It is concluded that in a near-surface nuclear explosion, radioactivity is primarily distributed in soil particles that have not been vaporized, and according to Henry's law and the ideal gas law,  $r_{89,95}$  may vary in larger particles when the effective volume of the fireball changes.

**Keywords:** Fission products, Fractionation, Diffusion model

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## Introduction

A nuclear explosion produces five primary effects: blast and shock, thermal radiation, electromagnetic radiation, initial nuclear radiation, and residual nuclear radiation (fallout) [?]. Fallout is particularly significant in ground bursts, creating concentrated radioactive hazards that can damage ecosystems over extended periods. Evaluating radioactive fallout is essential for limiting ionizing radiation damage and guiding emergency response planning for nuclear explosions. The total quantity of radioisotopes and their distribution across different particle sizes depend on explosion parameters including weapon yield, burst height, and soil type at ground zero. Consequently, computer simulation of radioactive particle formation and quantitative analysis of radioisotope distribution across various particles are crucial for fallout prediction. Research at the Air Force Institute of Technology (AFIT) indicates that the fractionation influence coefficient in radioactive fallout exceeds 2, underscoring the importance of fractionation models for understanding explosion phenomena and validating sample representativeness [?].

Typical fractionation models include the Freiling-Tompkins model (also known as the Modified Radial Power Law), Miller's thermodynamic equilibrium model, and Martin's diffusion-based G-X model [?]. Developed by Martin in the 1980s, the G-X model categorizes fallout particles into two distinct types: glassy and crystalline. However, the total specific activity calculated for glassy and crystalline particles using the G-X model shows much greater variation than observed in test data. This discrepancy arises primarily from different particle positions within the fireball. All particles are drawn into the fireball at early times, and their specific activities vary by location due to rapid changes in temperature and fission product concentration. Additionally, some fine particles may form through condensation of vaporized soil on individual atoms or molecules. After soil particles begin to solidify, smaller-diameter particles continue to be drawn into the cloud by afterwinds and remain completely unmelted.

Considering the particle formation process, fallout particles can be divided into three categories: (1) particles from device debris and vaporized soil, (2) particles representing completely or partially melted soil, and (3) particles repre-

senting unmelted soil. Based on particle size and formation mechanism, these three types are referred to as fine particles, glassy particles, and crystalline particles, respectively. Using this classification, a new fractionation model was constructed to describe the distribution of radioactive nuclides among these particles. Since the effective volume and mass fraction of fine particles cannot be precisely determined, the influence of these parameters on the results was also investigated.

## Method and Parameter Selection

### Method

Calculations proceed one decay chain at a time when the fireball temperature is approximately 3500 K. After this point, isotopes are assumed to condense on particle surfaces according to Henry's law and diffuse into fallout particles. The isotopes decay over time as temperature decreases. This process repeats until the temperature approaches the solidification temperature for glass. Beyond this point, fission products in the gaseous state distribute across the surfaces of all particle types, with nuclide amounts proportional to the total surface area of each particle group.

In the first stage, only agglomeration between fine particles need be considered. Since all fine particles are small enough for nuclides to be considered volume-distributed, and ignoring diffusion within fine particles, there is no relationship between fission product distribution and the fine particle growth process. To simplify the model, we assume fine particles of constant mass grow primarily through agglomeration in the first stage. The diffusion equation for spheres with only radial diffusion is:

$$\left( \frac{\partial^2 c_i}{\partial r^2} + \frac{2}{r} \frac{\partial c_i}{\partial r} \right) = \frac{1}{D_i} \frac{\partial c_i}{\partial t}$$

where  $c_i$  is the concentration of fission product  $i$ ,  $t$  is diffusion time,  $r$  is the radial coordinate, and  $D_i$  is the diffusion constant. According to Henry's law, the partial pressure  $P_i$  of fission product  $i$  is:

$$P_i = k_i x_i$$

where  $k_i$  is the Henry's law constant and  $x_i$  is the concentration of fission product  $i$  at the particle surface.

By dividing the first stage into many small steps, the mass balance at Step 1 is obtained by combining the diffusion equation, Henry's law, and the ideal gas law:



## Particle Size Distribution

Heft suggested that nuclear debris could be described by a linear combination of several lognormal distributions [?], representing distributions for each particle type generated by the burst. The subsurface mass distribution is given by:

$$f(d) = \sum_{i=1}^3 \frac{\phi_i}{\sqrt{2\pi} \ln(\sigma_i) d} \exp \left[ -\frac{1}{2} \left( \frac{\ln(d) - \alpha_{0i}}{\ln(\sigma_i)} \right)^2 \right]$$

where  $\phi_i$  is the mass percentage for the  $i$ th particle type,  $\sigma_i$  is the standard deviation, and  $\alpha_{0i}$  is the natural logarithm of the average diameter.

Based on Delfic' s lognormal distribution and Baker' s airborne particle distribution, and considering that soil particles near the Small Boy test ground zero were below 150  $\mu\text{m}$  in diameter, the values of  $\sigma_i$  and  $\alpha_{0i}$  used in this research are given in Table 1 .

## Parameters Selection

The main fireball parameters in this research are the volume and temperature during cooling to the glass solidification temperature. The temperature is selected from Small Boy data. Since the fireball size varied little after fireball breakaway, the averaged fireball volume is  $10^6 \text{ m}^3/\text{kt}$  in this research.

Norman et al. conducted extensive studies to determine Henry' s law constants for atomic species relevant to fallout research [?]. Norman and colleagues also made significant contributions to determining diffusion constants for various soil types. The soil type of interest is CaO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> soil, which is representative of common soils. The Henry' s law constants and diffusion coefficients used in this research are listed in Ref. [?].

Fission yield data and radioactive decay data are required for fission product dynamic calculations. Independent fission yield data together with radioactive decay data were selected from ENDF/B-VII.1 [?].

## Results

### Fractionation Results

In a ground explosion, approximately 5000 tonnes of earth per kiloton of yield are released into the atmosphere, of which 180-200 tonnes are fused. The vaporized soil mass is 1.5-25 tonnes per kiloton. Since Small Boy was a low-yield nuclear explosion, 3 tonnes of vaporized soil particles and 1 kiloton of crystalline particles are assumed, with 200 tonnes of melted soil particles determined [?].

To evaluate the model, plots of  $r_{89,95}$  as a function of particle size are useful. The ratio  $r_{i,j}$  is defined as  $r_{i,j} = f_i/f_j$ , where  $f_j = a_i/Y_i$ ,  $a_i$  is the number of atoms of nuclide  $i$  per fission, and  $Y_i$  is the yield of nuclide  $i$  per fission. Log-log

plots of  $r_{i,95}$  versus  $r_{89,95}$  can be prepared and fitted with linear least squares to determine correlation slopes.

The  $r_{89,95}$  values and correlation slopes calculated by this model and the G-X model are compared with Small Boy data in Fig. 1 [Figure 1: see original paper] and Table 2. The cumulative activity-size distribution calculated by this model with 3 tonnes of vaporized soil particles and 1 kiloton of crystalline particles is compared in Fig. 2 [Figure 2: see original paper] with results from the G-X and F-T models.

Figure 1 shows that our method better matches the Small Boy data above 100  $\mu\text{m}$ . Since the Small Boy data are primarily above 20  $\mu\text{m}$ , the new method performs significantly better in calculating  $r_{89,95}$ . Figure 2 shows that our results for the cumulative activity fraction of particles larger than 100  $\mu\text{m}$  diameter, which mainly deposit near ground zero after the explosion, fall between the results of the F-T and G-X models. However, the logarithmic correlation slopes calculated with both models show poor consistency with Small Boy data, though the new model shows poorer consistency for the logarithmic correlation slope of mass chain 131. Log-log plots of  $r_{131,95}$  versus  $r_{89,95}$  from the G-X model in Ref. [?] are cited, and linear fitting yields a slope of approximately 0.40, which differs substantially from the value in Table 2 and is nearly equal to the result from this model. Since additional information about Martin' s fitting method is unavailable and the logarithmic correlation slopes vary little when effective volume and fine particle mass fraction are changed, further attention to these slopes was not pursued. The current deviation in slopes for mass chains 91 and 140, where elements primarily affecting fractionation are mixed-behavior elements with appreciable gradients in particle distribution or some bulk loading, may suggest that the diffusion constants used are overestimated.

### Sensitivity Analysis

**Model Sensitivity to Effective Volume** Since the fireball experiences rapid temperature and concentration declines, the effective volume involved in the condensation process is smaller than the physical fireball volume. The  $r_{89,95}$  values and activity-size distributions are shown in Figs. 3 [Figure 3: see original paper] and 4 [Figure 4: see original paper] for effective volume fractions of 20% and 100% of the fireball.

For particles larger than 1 mm diameter, as shown in Fig. 3,  $r_{89,95}$  differs between the 20% and 100% effective volume fractions, while the cumulative activity-size distributions remain unchanged. According to Henry' s law and the ideal gas law, the fraction of fission products condensed on particles or remaining in the gaseous state changes when the effective volume varies.

**Model Sensitivity to Mass of Vaporized Soil Particles** In surface nuclear explosions, fine particles may form through condensation on individual atoms or molecules. However, the mass fraction of these particles cannot be

determined precisely due to uncertainties in vaporized soil mass and competition in the condensation process between fine particles and melted soil particles. Approximately 40 tonnes of fine particles may form in a 1.65 kt explosion, but only 15 tonnes were used as a maximum value due to the low yield and approximately 3 m burst height. The  $r_{89,95}$  values and activity-size distributions were calculated for vaporized soil particle masses of 0 t, 3 t, and 15 t (Figs. 5 [Figure 5: see original paper] and 6 [Figure 6: see original paper]). The results show that vaporized soil particle mass has no effect on the  $r_{89,95}$ -size relationship, and the cumulative activity-size distribution changes negligibly with vaporized soil particle mass, as the melted and unmelted soil particles involved are far more abundant than vaporized soil particles.

## Conclusion

Large quantities of soil entrained into the fireball by blast and updraft winds remain unmelted. We can assume that unmelted soil particles do not play an important role in the condensation process of fission products onto particles, but they provide large surface areas for gaseous fission products after condensation on melted soil. Based on Small Boy data near ground zero, Delfic' s lognormal distribution, and Baker' s airborne distribution, the particle size distribution is characterized and a new model is proposed to calculate fission product distribution across particles of different sizes. The new method performs significantly better in calculating  $r_{89,95}$ , and the cumulative activity fraction for particles larger than 100  $\mu\text{m}$  diameter falls between results calculated with the F-T and G-X models. Model sensitivity analysis shows that both the  $r_{89,95}$ -size relationship and cumulative activity-size distribution are insensitive to effective volume and vaporized soil particle mass. It is concluded that in a near-surface nuclear explosion, radioactivity is mainly distributed on soil particles that have not been vaporized, and  $r_{89,95}$  may vary in larger particles when effective volume changes according to Henry' s law and the ideal gas law.

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