NUCLEAR SCIENCE AND TECHNIQUES **25**, 050301 (2014)

A fractionation model based on three lognormal particle size distributions

SHANG Jian-Bo (商建波),[1,2,*] SHI Quan-Lin (师全林),[2] WANG Qun-Shu (王群书),[1,2] ZHONG Zhen-Yuan (钟振原),[2] LEI Bei-Fang (雷北方),[2] LIU Jie (刘杰),[2] BAI Tao (白涛),[2] DAI Yi-Hua (代义华),[2] and LI Mou (李谋)[2]

[1]*Department of Engineering Physics, Tsinghua University, Beijing 100084, China*
[2]*Northwest Institute of Nuclear Technology, Xi'an 710024, China*

(Received December 25, 2013; accepted in revised form March 3, 2014; published online September 21, 2014)

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 μm 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.

Keywords: Fission products, Fractionation, Diffusion model

DOI: 10.13538/j.1001-8042/nst.25.050301

I. INTRODUCTION

A nuclear explosion generally produces five effects: blast and shock, thermal radiation, electromagnetic radiation, initial nuclear radiation, and residual nuclear radiation (fallout) [1]. Fallout plays an important role especially in ground nuclear explosion by creating a concentrated radioactive hazard causing potential damages to ecosystem in a relatively long period of time. Radioactive fallout must be evaluated to limit the ionization damages and provide guidance for formulating emergency plans in response to nuclear explosions. The total amount of radioisotopes and their distribution in particles of different sizes are determined by parameters of nuclear explosion, such as weapon yield, explosion height and soil type of ground zero. Therefore, computer simulation of the formation of radioactive particles in various explosions and quantitative analysis of the radioisotope distribution in different particles play an important role in fallout forecasting. It was found that the influence coefficient of fractionation in radioactive fallout is over 2 according to researches conducted in Air Force Institute of Technology (AFIT), but fractionation models are still important for better understanding of explosion phenomena and evaluation of validity of samples [2].

Freiling-Tompkins fractionation model (also known as Modified Radial Power Law), Miller's thermodynamic equilibrium model and Martin's diffusion based G-X model are typical fractionation models [3–5]. The G-X model was designed by Martin in 1980s, in which fallout particles are divided into two distinct types: glassy and crystalline. However, the total specific activity of glassy and crystalline particles calculated with the G-X model varies much more greatly than the results of test observation. This can be attributed mainly to the different particle position in the fireball. All the particles are drawn into fireball at an early time, and different locations in the fireball decide the specific activities because of rapid changes in temperature and fission product concentration in the fireball. Also, some fine particles may come into being from condensation of the vaporized soil on separate atoms/molecules. After the soil particles begin to solidify, soil particles of smaller diameters are still drawn into cloud by after-winds, and are entirely un-melted.

Considering the particle formation process, the fallout particles can be divided into three types: 1) particles from the device debris and vaporized soil, 2) particles representing completely or partially melted soil, and 3) particles representing soil never melted. Based on the particle size and formation process, the three types of particles are referred to as fine particles, glassy particles and crystalline particles, respectively. With this classification, a new fractionation model was constructed to describe the distribution of radioactive nuclides in these particles. Taking into account that effective volume and the mass fraction of fine particles cannot be determined clearly, influence of the result from effective volume and mass of fine particles was also examined.

II. METHOD AND PARAMETER SELECTION

A. Method

Calculation proceeds on one decay chain at a time when the fireball temperature is about 3500 K. After this point the isotopes are assumed to condense on surface of the particles according to Henry's law and diffuse into the fallout particles. The isotopes decay with the time and the temperature falls. The process is repeated until the temperature is close to the solidification temperature for the glass. After this point, the fission products in gaseous state will be distributed on surface of all types of particles. The amounts of the nuclides distributed in particles of different sizes and types are proportional to the total surface area of the particle group.

In the first stage, only agglomeration between fine particles should be considered. Since all the fine particles are small enough to be considered nuclides volume-distributed, if we

* Corresponding author, shangjianbo@nint.ac.cn

ignore the diffusion process in the fine particles, there is no relation between the distributions of fission products and the fine particle growth process. To simplify the model, we suppose that the fine particles of a constant mass at the first stage grow up mainly by agglomeration. The diffusion equation for spheres with only radial diffusion is

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

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

$$P_i = k_i x_i, \tag{2}$$

where k_i is the Henry Constant and x_i is concentration of fission product i at the particle surface.

By dividing the first stage into many small steps, at Step 1 we have the following mass balance by combing the diffusion equation, Henry's law and ideal gas law:

$$Y_{mi} = \frac{c_{i1} \bar{k}_{i1} V_1 M_i}{RT_1} + c_{i1} \sum_{s=1}^{p} \Delta_{i1}^{s} m_s + c_{i1} m_\nu, \tag{3}$$

$$\bar{k}_{i1} = k_{i1} \frac{M_i}{M_s}, \tag{4}$$

$$\Delta_{i1} = 1 - \frac{6}{\pi^2} \sum_{k=1}^{\infty} \frac{1}{k^2} \exp\left(\frac{-D_i k^2 \pi^2 t_1}{a^2} \right), \tag{5}$$

where subscript i denotes the fission product; Y_{mi} is its mass i in the first time period; c_{i1} is its surface layer concentration in the soil and k_{i1} is its Henry's Law constant, at temperature of Step 1; R is its gas constant; M_i is its mole weight; m_s is the mass of soil in particle size class s; p the number of particle size classes; m_ν the mass of fine particles; and t_1 is the first time period. On right side of Eq. (3), the first term is the amount of fission product i in gas phase, the second term is the amount of fission product i diffused into melted soil particles by solving diffusion equation, and the third term is the amount of fission product i diffused into fine particles formed mainly by vaporized soil.

The immediate precursor of isotope i which diffused into particles in time interval Δt_1 may have decayed into isotope i. The influence to diffusion of isotope i in Step 2 by the mentioned process can be ignored if the time step is small enough. The mass balance equation after the second time period is

$$\begin{aligned} Y_{i2} =& \frac{c_{i2} \bar{k}_{i2} V_2 M_i}{RT_2} + c_{i1} \exp(-\lambda_i \Delta t_1) \sum_{s=1}^{p} \Delta_{i,1,2}^{s} m_s \\ &+ c_{i1} \exp(-\lambda_i \Delta t_1) m_\nu \\ &+ [c_{i2} - c_{i1} \exp(-\lambda_i \Delta t_1)] \sum_{s=1}^{p} \Delta_{i,2,2} m_s \\ &+ [c_{i2} - c_{i1} \exp(-\lambda_i \Delta t_1)] m_\nu, \end{aligned} \tag{6}$$

where Y_{i2} is the mass of fission product i and c_{i2} is the surface layer concentration of i in the soil, in Step 2; λ_i is the decay constant of i; and λ_{i-1} is the decay constant of precursor of isotope i.

This calculation goes iteratively until the temperature is close to the solidification temperature for the glass. In the second stage, the distribution of fission product i in particles with different sizes and types is given by

$$Y_{msj} = Y_{m\nu} \cdot Surf_j \cdot f_s, \tag{7}$$

$$Surf_j = \frac{m_j \nu_j^{-\frac{1}{3}} \exp\left\{ \left(-2.5 \cdot \ln^2(\sigma_j) \right) \right\}}{\sum_{j=1}^{3} m_j \nu_j^{-\frac{1}{3}} \exp\left\{ \left(-2.5 \cdot \ln^2(\sigma_j) \right) \right\}}, \tag{8}$$

where Y_{msj} is the mass of isotope i on the surface of the s^{th} size and j^{th} type particles, $Y_{m\nu}$ the mass of gaseous istope i after stage 1, $Surf_j$ the total surface fraction of the j^{th} type, m_j the total mass of the j^{th} type particles, ν_j the mean volume of the j^{th} type particles and σ_j the standard deviation.

B. Particle size distribution

Heft suggested that nuclear debris might be described by a linear combination of several lognormal distributions [6], which are distributions for each type of particles generated by the burst. The subsurface mass distribution is given by

$$f(d) = \sum_{i=1}^{3} \left(\frac{\varphi_i}{\ln(\sigma_i) d \sqrt{2\pi}} \right) \exp\left[- \left(\frac{\ln(d) - \alpha_{0i}}{\ln(\sigma_i)} \right)^2 \right], \tag{9}$$

where φ_i is the percentage of mass for the i^{th} type of particles, σ_i is the standard deviation, and α_{0i} is the natural logarithm of average diameter.

Based on Delfic's lognormal distribution and Baker's airborne particle distribution, and with the knowledge that soil particles near ground zero of the Small Boy atom bomb are below 150 μm in diameter, the values of σ_i and α_{0i} in this research are given in Table 1.

TABLE 1. The values of σ_i and α_{0i} in this research

Particle types	α_{0i}	σ_i
Melted soil particles	0.407	4.0
Un-melted soil particles	4.000	2.0
Vaporized soil particles	0.200	2.0

C. Parameters selection

The main fireball parameters in this research are the volume and temperature during the fireball cooling to the solidification temperature of glass. The temperature is selected

from the Small Boy data. The fireball size varied little after the fireball breakaway, so the averaged fireball volume is 10^6 m^3/kt in this research.

Norman *et al*. did many studies to determine Henry's Law constants for those atomic species of interest in fallout research [7]. Norman and others contributed a lot in determining diffusion constants of various soil types. The soil type of interest here is the $CaO-Al_2O_3-SiO_2$ soil, which is representative of common soils. The values of Henry's Law constants and diffusion coefficients used in this research are listed in Ref. [3].

Fission yield data and radioactive decay data are required for fission product dynamic calculation. Independent fission yield data together with radioactive decay data were selected from ENDF BVII.1 [8].

III. RESULTS

A. Fractionation results

In a ground explosion, about 5000 t of earth per kt of yield are released into the atmosphere and, of that quantity, $180 \sim 200$ t are fused. The vaporized soil is $1.5 \sim 25$ t per kt. As the Small Boy is a nuclear explosion of low yield, 3 t vaporized soil particles and 1 kt crystalline particles are assumed and 200 t melted soil particles is determined [9].

To evaluate the model, the plots of $r_{89,95}$ as a function of particle size is useful. The ratio $r_{i,j}$ is defined as

$$r_{i,j} = f_i/f_j, \tag{10}$$

$$f_j = a_i/Y_i, \tag{11}$$

where a_i is the number of atoms of nuclide i in the sample corresponding to one fission and Y_i is the yield of nuclide i per fission. Log-log plots of $r_{i,95}$ versus $r_{89,95}$ values can be prepared and fitted with linear least squares to determine the correlation slopes.

The $r_{89,95}$ and the correlation slopes calculated by this model and G-X model are compared to the Small Boy data in Fig. 1 and Table 2. The cumulate activity-size distribution calculated by this model with 3 t vaporized soil particles and 1 kt crystalline particles is compared in Fig. 2 with the results using G-X and F-T models.

Figure 1 shows that our method better matches the Small Boy data above 100 μm. The Small Boy data are mainly above 20 μm, so the new method performs much better in calculating $r_{89,95}$. Fig. 2 shows that our results of the cumulate activity fraction of particles over 100 μm diameters, which mainly grounded in an area near ground zero after explosion, are between the results of the F-T and G-X models. But the logarithmic correlation slopes calculated with both models shows poor consistency with the Small Boy data, but the new model shows a poorer consistency in logarithmic correlation slopes of 131. Log-log plots of $r_{131,95}$ versus $r_{89,95}$ of G-X model in Ref. [3] are cited, and the linear fitting gives a slope

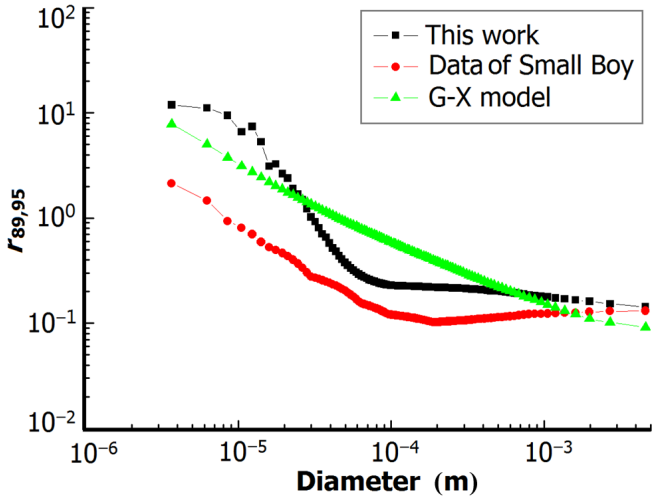

Fig. 1. (Color online) $r_{89,95}$ vs. particle size for different methods.

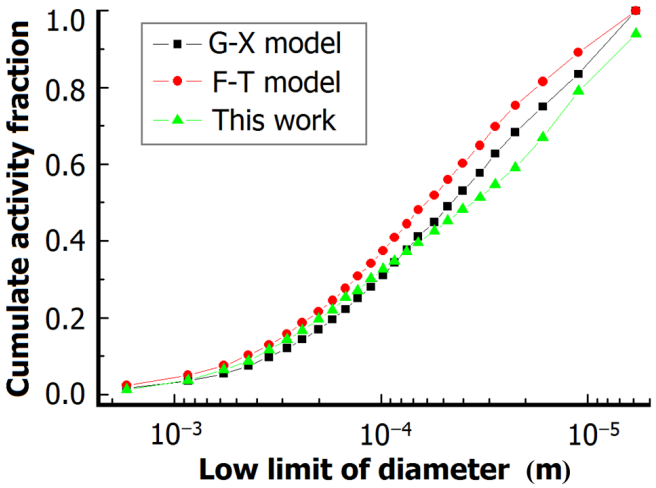

Fig. 2. (Color online) Cumulate activity-size distribution for different models.

TABLE 2. The values of σ_i and α_{0i} in this research

Atomic mass	Small Boy	G-X	This work
89	1.00	1.00	1.00
90	0.73	0.83	0.73
91	0.51	0.64	0.39
95	0.00	0.00	0.00
99	0.04	-0.07	0.09
131	0.84	0.58	0.36
132	0.90	0.61	0.67
136	0.65	1.39	1.35
137	1.19	0.91	1.02
140	0.58	0.34	0.34
144	0.03	-0.14	0.01

of about 0.40 which varies much with the value given in Table 2 and is nearly equal with result of this model. As we cannot find more information about how Martin fitted the plots and logarithmic correlation slopes vary little when we change

effective volume and fine particle mass fraction, we do not pay more attention to the logarithmic correlation slopes in later research. The current deviation of slopes of mass chain 91 and 140, where elements mainly affecting fractionation in the chain are mixed behavior elements whose distribution in fallout particles is appreciable gradient or some bulk loading, may suggest that diffusion constants used are overestimated.

B. Sensitivity analysis

1. *Model sensitivity to effective volume*

Since the fireball experiences a rapid temperature and concentration fall, the effective volume in which the condensation process is involved is smaller than the physical volume of the fireball. The $r_{89,95}$ and activity-size distribution were shown in Figs. 3 and 4, with 20% and 100% as the fraction of effective volume in the fireball.

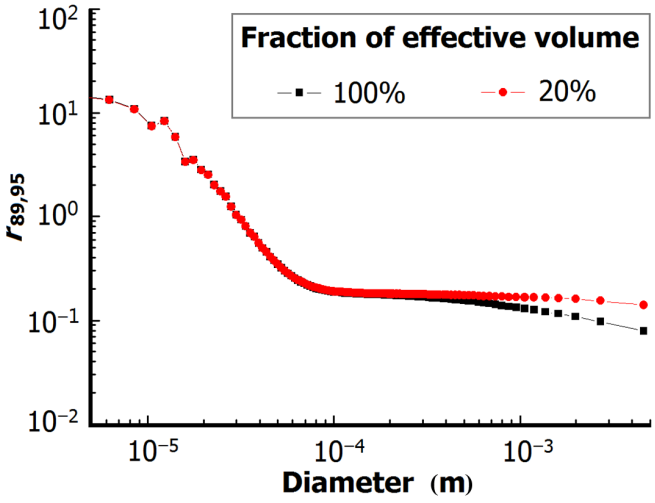

Fig. 3. (Color online) Dependency of particle size on $r_{89,95}$.

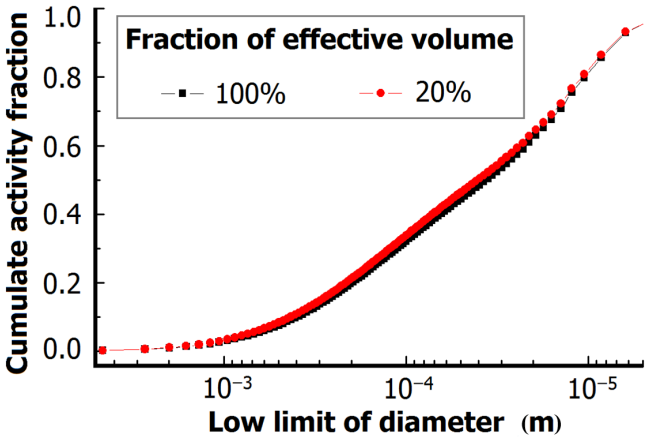

Fig. 4. (Color online) Cumulate activity-size distribution.

For particles of diameters over 1 mm, as shown in Fig. 3, the $r_{89,95}$ differ from each other for 20% and 100% fraction of effective volume, while the cumulate activity-size distributions remain unchanged. According to Henry's law and ideal gas law, the fraction of fission products condensed in particles or still in gaseous state changes when effective volume varies.

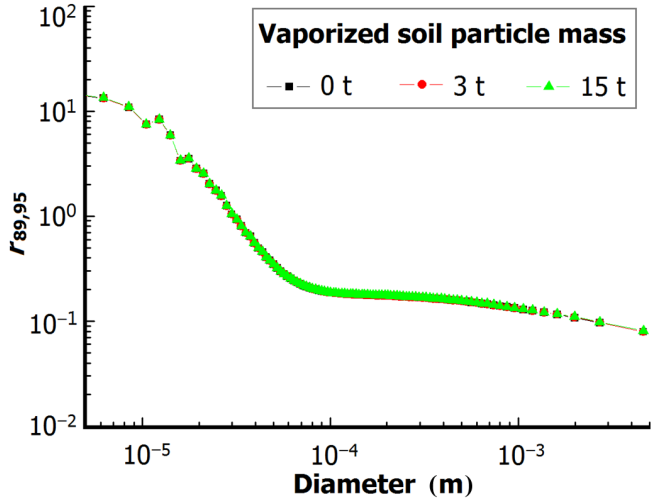

Fig. 5. (Color online) $r_{89,95}$ vs particle size for 0–15 t mass of vaporized soil particles.

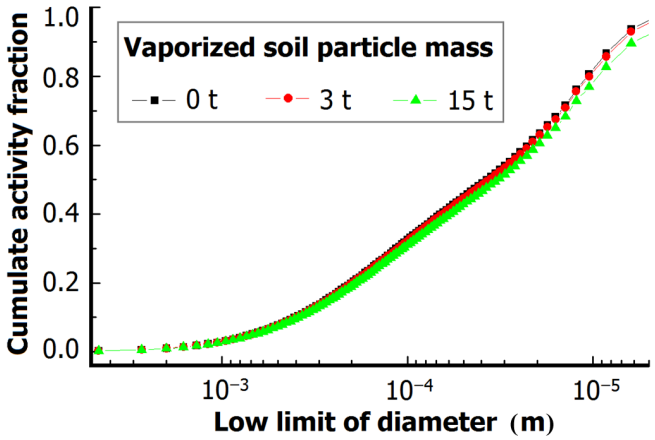

Fig. 6. (Color online) Cumulate activity-size distribution for 0–15 t mass of vaporized soil particles.

2. *Model sensitivity to mass of vaporized soil particles*

In surface nuclear explosions, fine particles may form in the process of condensation on separate atoms/molecules. But mass fraction of the particles cannot be determined due to uncertainty of vaporized soil mass and the condensation process competition between the fine particles and melted soil particles. About 40 t fine particles may form in an explosion of 1.65 kt weapon yield, but just 15 t fine particles was used as a maximum value because of the low yield and about 3 m explosion height. The $r_{89,95}$ and activity-size distribution were

calculated for vaporized soil particle masses of 0 t, 3 t and 15 t (Figs. 5 and 6). The results show that the mass of vaporized soil particles has no effect on the $r_{89,95}$–size relation and the cumulate activity–size distribution hardly changes with the vaporized soil particle mass, as melted and un-melted soil particles involved are much more than the vaporized soil particles.

IV. CONCLUSION

Large quantity of soil entrained into the fireball by blast and updraft winds is not melted. We can assume that the unmelted soil particles do not play an important role in the condensation process of fission product on the particles, but they offer large surface areas for the gaseous fission product after condensation of the melted soil. Based on the 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 the fission product distribution on particles of different sizes. The new method has a much better performance in calculating $r_{89,95}$, and the cumulate activity fraction for particles in diameters of over 100 μm is between the results calculated with the F-T and G-X models. Model sensitivity analysis shows that the $r_{89,95}$–size relation and the cumulate activity–size distribution are insensitive to both effective volume and mass of vaporized soil particles. It is concluded that in a nuclear explosion near surface, radioactivity is mainly distributed on the soil particles which have not been vaporized and $r_{89,95}$ may vary in larger particles when effective volume changes according to the Henry's law and ideal gas law.

[1] Glasstone S and Dolan P J. The Effects of Nuclear Weapons (Third Edition). Alexandria V A: US Department of Defense and US Department of Energy, 1977, 1-4.
[2] Trabalka J R and Kocher D C. Bounding Analysis of Effects of Fractionation of Radionuclides in Fallout on Estimation of Doses to Atomic Veterans. Fort Belvoir (USA): DTRA-TR-07-5, 2007.
[3] Martin C R. Ph.D. Thesis. Air Force Institute of Technology, 1983.
[4] Freiling E C. Science, 1961, **133**: 1991-1998.
[5] Freiling E C. Science, 1963, **139**: 1058-1059.
[6] Heft R E. Adv Chem Ser, 1970, **93**: 254-281.
[7] Norman J H. Henry's Law Constants for Dissolution of Fission Products in a Silicate Fallout Particle Matrix, San Diego (USA), GA 7058, 1966.
[8] McLane V. ENDF-102 Data formats and procedures for the evaluated nuclear data file ENDF-6. New York (USA), BNL-NCS-44945-01/04-Rev, 2001.
[9] Izrael Y A. Radioactive fallout after nuclear explosions and accidents. Argyll, Scotland, UK: Ampfield House, 2002, 9-10.