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Abstract

An assessment is made of the rainout of airborne radioactive particles from a nuclear detonation with emphasis on the microphysical removal processes. For submicron particles the scavenging processes examined are Brownian and turbulent diffusion to cloud droplets. For particles larger than 1 μm radius, nucleation scavenging is examined. For various particle size and radioactivity distributions, it is found that from 27-99% of the radioactivity is attached to cloud droplets and subject to rapid removal by rain.

Introduction

The immediate dry fallout of radioactivity from a nuclear airburst is of little consequence due to the slow fall velocities of the aerosol particles produced. If the debris cloud encounters a raining system during the first several hours of its existence, however, the possibility exists for the rain to remove radioactivity from the air and deposit it on the ground in high concentrations.

A complete description of this phenomenon involves computing the debris-cloud trajectory, accounting for atmospheric diffusion and wind shear of the cloud, and modeling the interaction of the debris cloud with a rain cloud to determine the amount of radioactivity that enters the raining system and is removed. The dominant pathway for the transfer of small radioactive particles to rainwater is by their incorporation into cloud droplets followed by the removal of the particle-laden droplets through collision with raindrops. The efficiency of removal depends upon such factors as particle size, their propensity to become cloud-droplet-condensation sites, cloud-droplet size and concentration, flux of raindrops, external electric fields, droplet charge, particle charge, and the intensity of microscale turbulence inside the cloud. Other important considerations are the relative heights of the two clouds during interaction and the stage of development of the rain cell at onset of interaction. Finally, we need to know not only the concentration but the spatial contours of radiation deposited on the ground by raindrops falling from a moving cloud and being evaporated and transported by the winds all the while.

On the basis of atmospheric-diffusion calculations,[1,2] high levels of radiation on the ground are produced if all the radioactivity contained in a vertical column through the debris cloud center

is removed to the ground, this being true for several hours after detonation assuming no prior scavenging. Calculations using a convective rain-cloud model[3] show that the amount of debris that can enter a rain cloud is dependent upon the relative heights of the debris and rain clouds. The most efficient interaction occurs for debris located below the rain-cloud base, which can lead to an enhancement of the radioactivity contained in a vertical column over that predicted from atmospheric-transport considerations. Rainout thus becomes important only for low-yield devices that produce a low-altitude stabilized debris cloud.

According to the above considerations involving atmospheric-transport and rain-cloud modeling, the possibility exists for the deposition of significant amounts of radioactivity by rain. One further process can control the amount of radioactivity removed, that being the rate at which the particles are removed by a rain cloud. If the characteristic removal time for debris particles is long, compared to the lifetime of the rain cloud, the microphysical interactions that account for the transfer of particles into the rainwater limit the amount of debris removed. If the microphysical processes are very efficient, the amount of radioactivity is limited by some other aspect of the problem.

The present effort is directed toward determining the extent to which the scavenging of nuclear debris is controlled by microphysical processes. Basis for several important assumptions made in the course of the study is to arrive at a lower limit for the amount of debris removed under varying circumstances. An absolute lower limit is, of course, zero (e.g. it does not rain). For situations in which other controlling processes combine to admit the possibility of a scavenging event, however, microphysical processes can, in principle, limit the amount of debris removed. It is this lower limit that is under consideration here.

It is desirable to evaluate the microphysical processes independently of other controlling processes. The procedure used is to conceptually trace the history of a characteristic air parcel containing debris particles. Due to the small sizes of the particles, it is assumed that atmospheric- and rain-cloud-transport processes do not change the relative concentration of particles of each size, although the total number of particles per unit volume can change. The required result is to calculate the fraction of radioactivity removed from the parcel and transferred to cloud water. Radioactive decay and other processes that alter the total amount of activity in the parcel are neglected here.

Particle Size and Radioactivity Distribution

Debris particles from a nuclear air burst are formed as the fireball cools. The particles consist mainly of oxides of the dominant structural metals, usually iron and aluminum, with a comparatively small amount of fission and activation

products. The amount of fission products appears too small to influence appreciably either particle size or particle nucleation characteristics. There is evidence[4,5] to suggest that the debris-particle sizes are distributed log-normally.

For purposes of calculation the particle-size distribution is taken as log-normal and hence can be specified by assigning a geometrical mean radius, $\bar{r}$, and a geometrical standard deviation, σ. The distribution is written as

$$n(r, \bar{r}, \sigma) = C_0 (\sqrt{2\pi} \ln \sigma)^{-1} \exp\left[\frac{-(\ln r/\bar{r})^2}{2(\ln \sigma)^2}\right] \quad (1)$$

where C_0 is the total particle concentration at time of fireball stabilization and must satisfy:

$$C_0 = \int_{-\infty}^{\infty} n(r, \bar{r}, \sigma)\, d \ln r . \quad (2)$$

Defining the total mass of the condensed material as M, the volume of the stabilized debris cloud as V, and the mass density of the condensate as ρ, we can write:

$$\frac{M}{V} = \frac{4}{3} \pi \rho \int_{-\infty}^{\infty} r^3\, n(r, \bar{r}, \sigma)\, d \ln r . \quad (3)$$

By writing r^j as exp (j ln r), where j is a real number, we can show

$$r^j\, n(r, \bar{r}, \sigma) = \bar{r}^j \exp\left[\frac{(j \ln \sigma)^2}{2}\right] n(r, \bar{r}_j, \sigma) \quad (4)$$

where $\bar{r}_j = \bar{r} \exp (j \ln \sigma)^2$ and $n(r, \bar{r}_j, \sigma)$ is the log-normal function, Eq. (1), with $\bar{r}_j$ substituted for $\bar{r}$. Using this identity and the integration in Eq. (2), Eq. (3) can be written as:

$$C_0 = \frac{3M}{4\pi \bar{r}^3 \rho V} \exp\left[\frac{9}{2} (\ln \sigma)^2\right] . \quad (5)$$

Although this value for the particle concentration will not explicitly be used here, since we require only the fraction of particles in a unit volume that are scavenged, it is written for two reasons. First, in specifying the distribution by assigning a geometrical mean radius and geometric standard deviation, we must conserve mass which leads to Eq. (5). The total concentration is a strong function of both $\bar{r}$ and σ. Secondly, if we wish to account for atmospheric dissipation processes, other than microphysical processes that change the relative number of particles of different sizes, we can make the concentration given in (5) a function of time by letting the volume expand according to the appropriate transport criteria.

As the debris particles form and grow radioactive materials, mainly fission products, will tend to attach to the growing droplet. Those species that attach during the period of condensation will be more or less volume-distributed. Species that were volatile during the condensation stage, but which undergo radioactive decay to become refractory, will predominately be surface-distributed. The distribution of radioactivity within the particles should be a combination of both surface and volume distributions. For the present calculation the radioactivity will be considered either totally volume distributed or totally surface distributed and will be written

$$A(r) \propto r^k\, n(r, \bar{r}, \sigma) . \quad (6)$$

Here A(r) represents the activity per unit volume contained in particles of size

between ln r and ln r + d ln r, and k is taken as 2 or 3 for surface and volume distributions respectively. The total activity is proportional to

$$\int_{-\infty}^{\infty} A(r)\, d \ln r . \qquad (7)$$

Aerosol History

Following detonation of a nuclear device in the airburst mode, a visible water (ice) cloud is formed due to the rising of the buoyant mass of air. Some of the debris particles may serve as nucleation sites for the droplets. Since the induced water cloud exists for several minutes and is well mixed, the remaining debris particles are put in juxtaposition with the cloud droplets and have opportunity to attach to the droplets. (Attachment of debris particles to atmospheric aerosol particles or to each other is much slower than attachment to droplets due to the large surface area offered by the droplets.)

After the induced water cloud evaporates, providing it doesn't rain, the radioactive aerosol will consist of the particles that did not interact with droplets and the condensate of the evaporated droplets. The latter will be rich in activity due to nucleation and attachment of radioactive particles to the droplets. These enriched nuclei have much more mass than the particles that served as nuclei, having incorporated any impurities that managed to become attached to the droplets. They would probably be very efficient sites for condensation of water if again exposed to a humid or supersaturated environment.

At some later time the debris cloud may interact with a natural rain system. The enriched nuclei and any other debris particles that have suitable size and chemical characteristics will serve as condensation nuclei. The remaining debris particles will again have opportunity to attach to cloud droplets. When rain begins the droplets containing radioactivity may be incorporated upon collision with raindrops and fall to the ground.

In the present calculation it is assumed, as a lower limit for subsequent scavenging, that none of the debris particles serve as condensation nuclei in the induced water cloud. This is plausible in that the debris may well be initially contained in a toroid with the nucleation sites being atmospheric aerosols that are entrained in the rising air in and around the toroid. Due to the mixing that follows, it is assumed that the debris particles will all have opportunity to attach to the droplets by turbulent and Brownian diffusion. More efficient attachment that could occur due to electrical effects is not considered, again in the spirit of a lower limit.

After evaporation of the induced water cloud and subsequent transport downwind, it is assumed first that all enriched nuclei are activated in a natural cloud. In addition, any debris particle with radius larger than 1 μm is assumed to be a condensation site. That the enriched nuclei serve as condensation sites appears a good assumption, although no experimental data are available. Since the

debris particles are extremely wettable by water, those as small as 0.1 μm could be activated under natural cloud supersaturated conditions, but there is some question whether their surfaces undergo change during transport, so the 1 μm nucleation threshold is taken. (The possibility exists that soluble materials could attach to the debris particles during transit, lowering the nucleation threshold to 0.01 μm.)

Scavenging Rate

If we denote the concentration of particles of radius r at time t by c(r, t), the rate of decrease of the particles by attachment to cloud droplets by turbulent and Brownian diffusion[6] is:

$$\frac{d\,c(r,t)}{dt} = -\alpha\, c(r,t) \qquad (8)$$

where

$$\alpha = 4NV\pi R^2 \frac{(2 + Pe^{1/3})}{Pe}. \qquad (9)$$

$$Pe = \frac{2VR}{D_T + D_B}, \qquad (10)$$

and

- N = concentration of cloud droplets,
- V = droplet fall velocity,
- Pe = Peclet number (for both turbulent and Brownian diffusion),
- D_T = turbulent diffusion coefficient for particles of radius r,
- D_B = Brownian diffusion coefficient for particles of radius r.

The solution of Eq. (8) can be written

$$c(r,t) = c_0(r)e^{-\alpha t} \qquad (11)$$

where c_0 is the initial particle concentration. The fraction of particles of radius r that are removed can be written as:

$$f(r,t) = 1 - \frac{c(r,t)}{c_0(r)}. \qquad (12)$$

From Eq. (11) this becomes:

$$f(r,t) = 1 - e^{-\alpha t} \qquad (13)$$

If we characterize the particle attachment first in the induced water cloud following detonation by α_1, t_1 and the attachment in a natural cloud by α_2, t_2, the fraction of particles of radius r that becomes attached is:

$$f(r) = (1 - e^{-\alpha_1 t_1}) + (1 - e^{-\alpha_2 t_2})\, e^{-\alpha_1 t_1}$$

$$= 1 - e^{-(\alpha_1 t_1 + \alpha_2 t_2)}. \qquad (14)$$

The total fraction of radioactivity removed, accounting for particles of all sizes distributed log-normally as discussed above is:

$$F = \frac{\int_{-\infty}^{\infty} \left[1 - e^{-(\alpha_1 t_1 + \alpha_2 t_2)}\right] A(r)\, d\ln r}{\int_{-\infty}^{\infty} A(r)\, d\ln r} \qquad (15)$$

The removal rate α (Eq. 9) is a complex function of particle size, and there is no apparent way of rigorously evaluating the integral in Eq. (15) other than by numerical integration. However, for specific values of the important parameters, the fraction of particles removed from each size class (Eq. 14) can be approximated as:

$$f(r) = \begin{cases} K_1 & r \le 1\ \mu m \\ K_2 & 0.095\ \mu m \le r < 1\ \mu m \\ K_3/r^{0.74} & 0.016\ \mu m \le r < 0.095\ \mu m \\ K_4 & r < 0.016\ \mu m\ , \end{cases} \quad (16)$$

where $K_1 = 1$, $K_2 = 0.264$, $K_3 = 4.62 \times 10^{-2}$ $(cm)^{0.74}$, and $K_4 = 0.98$. For this approximation we have taken the concentration of cloud droplets to be $10^3/cm^3$ inside the induced water cloud and $300/cm^3$ inside the natural cloud. All droplets are assumed to be 10 μm in radius with a fall velocity of 10 mm/s. (For calculation of the turbulent diffusion coefficient we have assumed an atmospheric energy dissipation rate of 10 cm^2/s^3.) The particles are assumed to interact with the droplets for 15 min in both the induced cloud and the natural cloud. All debris particles greater than 1 μm in radius are assumed to be water condensation sites. Equation (15) now becomes:

$$F = \frac{K_1}{C_0}\int_{r=1\ \mu m}^{r=\infty} n(r, \bar{r}_k, \sigma)\, d \ln r$$

$$\times \frac{K_2}{C_0}\int_{r=0.095\ \mu m}^{r=1\ \mu m} n(r, \bar{r}_k, \sigma)\, d \ln r$$

$$+ \frac{K_3}{C_0 \bar{r}^{0.74}}\int_{r=0.016\ \mu m}^{r=0.095\ \mu m} n(r, \bar{r}_i, \sigma)\, d \ln r$$

$$+ \frac{K_4}{C_0}\int_{r=0}^{r=0.016\ \mu m} n(r, \bar{r}_k, \sigma)\, d \ln r \quad (17)$$

where i is equal to k - 0.74. These integrals can be readily evaluated on log-probability paper. Table 1 shows the results of Eq. (17) for several values of $\bar{r}$ and σ with the activity taken to be volume distributed, k = 3. Similar results are shown in Table 2 for a surface distribution of the activity, k = 2.

In order to calculate the total radioactivity removed we must account for atmospheric transport, radioactive decay, and the interaction of the debris cloud with a raining system. The above results could then be used to estimate the total amount of radioactivity incorporated in the cloud water. To predict the amount

Table 1. Percentage of activity attached to cloud droplets for various geometric mean radii, $\bar{r}$, and geometric standard deviations, σ, when the radioactivity is distributed according to particle mass.

Geometric mean radius, μm	Geometric standard deviation, σ		
	1.5	2	2.5
0.5	62	89	98
0.1	27	29	75
0.05	33	28	49
0.01	88	53	36

Table 2. Percentage of activity attached to cloud droplets for various geometric mean radii, $\bar{r}$, and geometric standard deviations, σ, when the radioactivity is distributed according to particle surface area.

Geometric mean radius, μm	Geometric standard deviation, σ		
	1.5	2	2.5
0.5	40	78	89
0.1	29	31	45
0.05	42	38	41
0.01	95	87	99

of activity removed to the ground, a reasonable estimate could be made using this information to calculate the specific activity of the water, then from the amount of rain at some point on the ground the total activity could be determined.

From the hydrodynamic considerations of raindrops accreting cloud droplets, it appears that about 90% of the droplets in a unit volume are removed after enough raindrops have fallen through the volume to account for 3 mm of rain. For rain rates greater than about 3 mm/h it takes only about 2 mm of rain to remove 90% of the original droplets. Under such conditions the fraction of the activity removed by cloud droplets becomes very nearly equal to the fraction of activity removed.

Conclusion

Upon consideration of microphysical precipitation-scavenging processes we have found that from about 30-100% of the radioactivity contained in aerosol particles from a free airburst is attached to cloud droplets and subject to rapid removal by rain under the following conditions:

1. The scavenging process for particles of radius greater than 1 μm is nucleation. For particles of radius less than 1 μm it is attachment by Brownian and turbulent diffusion of the particles to droplets, first in the detonation-induced water cloud of 10^3 droplets/cm^3 for 15 min and later in a natural cloud of 300 droplets/cm^3 for 15 min.

2. The particles are distributed log-normally with geometric mean radii ranging from 0.01 μm to 0.5 μm and geometric standard deviations ranging from 1.5 to 2.5.

3. The radioactivity is distributed either according to particle mass or particle surface area.

Precipitation scavenging of nuclear debris is a complicated phenomenon involving the related processes of atmospheric transport of the debris, interaction of debris and a rain system, and the microphysical scavenging processes. Only the latter process has been examined here and in a way that approaches a lower limit to the fraction of radioactivity scavenged. It appears that a significant fraction of the debris that becomes incorporated hydrodynamically into a raining system is scavenged.

References

1. J. B. Knox, T. V. Crawford, and W. K. Crandall, Potential Exposures from Low-Yield Free Air Bursts, Lawrence Livermore Laboratory, Rept. UCRL-51164 (1971).
2. W. K. Crandall, C. R. Molenkamp, A. L. Williams, M. M. Fulk, R. Lange, and J. B. Knox, An Investigation of Scavenging of Radioactivity from Nuclear Debris Clouds: Research in Progress, Lawrence Livermore Laboratory, Rept. UCRL-51328 (1973). (Rev. Oct. 1973).
3. C. R. Molenkamp, Numerical Modeling of Precipitation Scavenging by Convective Clouds, Lawrence Livermore Laboratory, Rept. UCRL-75896 (1974).
4. M. W. Nathans, R. News, W. D. Holland, and P. A. Benson, J. Geophys. Res. 75, 7559 (1970).
5. R. Heft, "The Characterization of Radioactive Particles from Nuclear Weapons Tests," Radionuclides in the Environment (American Chemical Society, Washington, D. C., 1970).
6. A. L. Williams, An Analysis of In-Cloud Scavenging Efficiencies, Lawrence Livermore Laboratory, Rept. UCRL-75897 (1974).

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