# 8 Fallout Models—Past, Present and Future

**R. D. SMALL**

## 8.1 NUCLEAR TESTS AND THE FIRST FALLOUT MODELS

Most of the energy released by a nuclear explosion is partitioned among blast, thermal radiation, and electromagnetic effects; together they account for all the damage and many of the immediate casualties. Some of the energy associated with these strong effects is involved in the formation and growth of radioactive particles. Although fallout of such particles contributes to immediate casualties, it is also a threat that persists for tens of years. In fact, fallout still continues after the atmospheric tests conducted by the USA, USSR, and UK until 1963, France until 1974 and China until 1980 (De Geer, 1994).

Despite the fact that over time nuclear fallout can cause more casualties than immediate effects, permanently contaminate large areas, and persist for many years, fallout models formulated in the 1960s before the development of modern supercomputers are still in use. As authors of this chapter have shown, such composite models can serve well to assess some past events—as long as the events are reasonably similar to the model's data base and the meteorological conditions are not too different. This is not possible in all cases; as often as not, weapon and weather conditions fall outside the empirical data range. There are two critical limitations: the first involves particle growth in nuclear fireballs; the second, nuclear cloud dynamics and the interaction with actual weather systems.

Aerosol dynamics are traditionally the weak point in weapon environment codes. Emphasis (and millions of dollars) on fireball and transport hydrodynamics has led to increasingly sophisticated analyses and codes. Weapon regime aerosol research, though, has lagged. At present, none of the current models robustly predict radioactive particle size distributions, accurately partition the radioactivity, or even reasonably account for processes controlling the formation of particles.

Most aerosol subroutines or deposition codes borrow heavily from conventional microphysics technology, which works fine (most of the time) for ambient environments and natural processes. From the nuclear aerosol

*Nuclear Test Explosions: Environmental and Human Impacts*
Edited by Sir F. Warner and R. J. C. Kirchmann

perspective, however, there are critical limitations: nuclear aerosols are likely to pass through widely varying thermodynamic regimes outside normal aerosol environments; and, nuclear aerosols are multispecies with morphologies and properties unlike normal atmospheric aerosols. Each compromise—especially in high-energy weapon environments—profoundly reduces solution fidelity. Recent developments in microphysics, plume, cloud and weather codes have yet to be applied to nuclear fallout. Except in a few select cases, approximations that were acceptable for assessing fallout from hundreds or thousands of weapons in a general exchange, fail when precision is needed.

In the 1950s several modellers noted that fallout depends directly on the particle size distribution of radioactive particles, which in turn depends on the size distribution of non-radioactive particles as well as vapours of the bomb casing, entrained dust, and water (Stewart, 1956; Adams *et al.*, 1960). Although the microphysics processes accounting for particle formation and growth were recognized, the high-resolution solutions for nuclear cloud sweep-up (dust) concentrations, size distributions as well as temperature, velocity and turbulence fields required to complete a first principle model were beyond computer capabilities at that time; they have become available only in the last few years.

The technical problem is easily summarized: N(100) kt of dust from several kilometres around the burst are drawn into a nuclear fireball that radiates N(10)–N(100) kt of thermal energy; added to that mix is N(10) kt of water, and N(0.1) kt of smoke. Each of these materials has special properties that influence the growth of radioactive particles and the subsequent fallout cloud dynamics.

In the mix of hundreds of kilotons of material added to the nuclear fireball, the weapon adds roughly 100 kg of radioactive material. The first task of a fallout model is to determine how it forms, attaches, grows through diffusion processes and coagulates with other particles. The partition of radiation both to large and submicron particles and the interactions with smoke, dust, and water in the nuclear cloud determines immediate, intermediate- and long-term fallout dose. No model does this at present.

Nuclear clouds are embedded in natural weather systems and deposition is either by settling of radioactive particles or episodic deposition by rain. Weather controls deposition, but there is an important connection or feedback with the nuclear aerosol. The special physical and chemical properties of the nuclear cloud determine the partition between interstitial dry particles and those captured by water. Both influence deposition and most importantly the timing of rain formation. A little earlier and some areas are spared fallout, while other areas are heavily dosed. The degree that a nuclear aerosol influences deposition depends on the type of weather system it is embedded in; impacts for deep and shallow convective systems, occult deposition, urban area clouds, and dry settling are different.

Smoke adds a large number of submicron particles that are (mostly) nucleation centres for water (Pittock *et al.*, 1989). Should those particles dominate, a nuclear fog could form thus delaying the formation of large particles (that fallout rapidly). Dust is just the opposite, creating large particles that fallout rapidly leading to heavily dosed regions. At the other end of the spectrum, high burst heights entrain little if any material into the nuclear cloud; and submicron radioactive particles form that settle very slowly, reaching the ground months or years later.

Model builders used nuclear test data from atmospheric test programmes conducted on three continents in the 1960s to construct empiricisms that prescribe the size distribution of radioactive particles. This approach compensated for limited computer power and the corresponding poor resolution of fireball hydrodynamics solutions. Several approximate models were developed (Pittock *et al.*, 1989). The advantage here is that real data are used in assessments; the glaring disadvantage is that it is very risky to extrapolate the empiricisms to other yields, soil conditions, urban targets, and meteorologies.

One to two orders of magnitude difference in fallout occurs depending on whether the particle size distribution (PSD) is given by commonly used log-normal, $r^{-3}$ or $r^{-4}$ distributions. In fact, new microphysics models (Small *et al.*, 1994), now show that there may not be a natural distribution that fits all burst conditions and yields. Nevertheless, as long as weather conditions are similar and target conditions match those in Nevada, Semipalatinsk, Novaya Zemlya and the Pacific, the 1960's generation of models provides reasonable fallout assessments. The next generation models promise a family of far more versatile models.

## 8.2 CREATION OF RADIOACTIVE PARTICLES

Both immediate and long-term fallout depend initially on molecular and submicron scale interactions. There are other factors, such as height of burst, yield, bomb construction and the mass entrained, that matter a great deal, but the important fallout physics occurs at a microscale. The fast formation of large particles or condensation of bomb vapours on large entrained particles account for immediate effects and fallout patterns around and downwind of targets. The very small particles—those $< 1$ $\mu$m—can account for one-half or more of the radioactivity and are responsible for fallout long after the burst. Such particles, initially lofted to high altitudes, are the continuing global legacy of atmospheric bursts (De Geer, 1994). Moreover, it is the very small (submicron) particles that most readily percolate, via many pathways, through the environment, with a pervasive impact on human health and ecological systems (Warner and Harrison, 1993).

The energy balances associated with particle evolution are complex and act through telescoping scales. The balance of heat and work in the expanding fireball regulates the formation and growth of particles. Part of the fireball thermal radiation vaporizes dust lifted off the ground by mechanical (blast) forces and entrained into the rising fireball by winds generated by the decaying buoyancy. The vaporized dust is a source of potential energy that is only released as the vapour nucleates and grows by condensation. From several kilometres around the burst, as much as $10^9$ to $10^{11}$ g of dust are drawn into the fireball; and over tens of seconds, $10^{12}$ to $10^{14}$ calories are radiated from the rising cloud. The evolution of particles at molecular and micron scales is shaped by balances of kilotons of energy and mass. Fallout over continents depends on the evolution of submicron particles. The physics is entwined in many scales and the details are important.

The early nuclear cloud models crudely approximated the formation of particles. The PSDs are based on sweep-up formulations and modified at coarse resolution either by simplistic microphysics formulations that omit major particle interactions or by empiricisms borrowed from natural cloud models. Neither approach faithfully accounts for all the interactions responsible for particle growth in nuclear clouds. Furthermore, particles smaller than 1 $\mu$m are usually neglected entirely. This is a critical flaw; such particles play a major role in the formation of radioactive particles. The consequence of approximating the microphysics is extreme uncertainty in the calculated PSD and, by extension, low confidence in fallout predictions or assessment of radioactive clouds.

Particle formation in nuclear clouds takes place in a fast thermodynamic environment. Not only do particles evolve in the rapidly cooling fireball, but they also quickly grow during the very fast rise to high stabilization altitudes. There are many paths: some particles form in the early seconds as the fireball plasma cools and the gas converts initially to molecular clusters and finally after successive waves of condensation to multicomponent solids; other particles grow by condensation on nuclei entrained by the rising cloud and by coagulation. Because of the large thermodynamic gradients any process can be reversed. Particles grown in one area of the cloud can be evaporated or 'broken' apart in high-temperature regions. Moreover, soil, smoke, or organic particles entrained by the cloud ablate when suddenly fluxed into the vortex core. Such processes continue for tens of seconds after the burst.

The microphysics is greatly influenced by the amount of entrained (swept-up) material. A number of factors determine how much material is added, but it mostly depends on the height of burst. Close to the ground, mass equal to one-third of the weapon yield is brought into the cloud; for burst heights greater than 120 m/kt$^{-1/3}$ only the bomb debris contributes to the microphysics.

The rising fireball also entrains low-level humid air and the water vapour condenses and freezes in a highly surpersaturated environment during the fast rise to high altitude. Stabilization in the stratosphere is not unusual. The nuclear cloud water microphysics may differ considerably from natural cloud processes. The rise is at much higher velocity, leading to greater supersaturation and a much altered balance of nucleation, condensation, and coagulation.

Moreover, the very high particle concentrations established by incipient particle formation, smoke particles added by immediate nuclear fires and the fireball processing of entrained dust provide an excess of condensation sites for the available moisture. In such conditions it is unlikely that normal cloud parameterizations apply.

More than one material is involved in nuclear cloud calculations: the radioactive bomb debris (which decays with time), entrained materials such as $SiO_2$, $Al_2O_3$, smoke, organic ground litter, and water. Particles may be either pure or mixed with varying fractions of the primary materials. Of special concern is the very small mass $N(10^5\ g)$ of radioactive material (plus neutron activated material) in a cloud with $N(10^{11}\ g)$ of dust and other materials. The radioactive mass is negligible compared with the entrained mass, but for fallout it is the meaningful quantity. Moreover, the radioactive material which is transformed from a plasma to a frozen solid can serve as nuclei for other condensing vapours, can itself condense on other particles, or can remain as a pure radionuclide.

There are several processes peculiar to nuclear cloud microphysics calculations. Radioactive decay produces high-energy electrons and concentrations of ion pairs; they rapidly recombine but nevertheless can accelerate nucleation, condensation, coagulation and even the breakup of large agglomerates. Turbulence forced by large shear and large temperature gradients increases coagulation rates and mixes the small amount of radioactive material throughout the cloud. Because of the very large temperature gradients, thermophoresis is important near the vortex core. Despite the fact that these and other effects may occur only for a short time and possibly only in a small area of the fireball, they may be important in determining the formation and growth of particles and the partition of radioactivity across the size distribution.

New models, enabled by the massive increase in computing power, couple high-resolution hydrodynamic solutions of the nuclear fireball with solutions of the microphysics general dynamic equation (GDE) for multispecies aerosols (Small *et al.*, 1994). The new hydrocode capabilities provide the detailed thermodynamic (pressure and temperature) and velocity–turbulence maps needed to 'drive' a microphysics calculation of radioactive particle formation and growth. Such calculations accounting for the first 10–60 s following a burst require several hours of CRAY simulation time.

Solution of the GDE describes the change in concentration of particles ($n$) of species $\kappa$ by the fundamental microphysics processes of nucleation, coagulation, and condensation (evaporation), ablation and breakup:

$$\frac{\partial}{\partial t}n^{\kappa}(v,t) = \sum_{\lambda}\left\{\frac{1}{2}\int_0^{v}E_{\kappa,\lambda}(\xi, v-\xi)n^{\kappa}(\xi,t)n^{\lambda}(v-\xi,t)d\xi - n^{\kappa}(v,t)\int_0^{\infty}E_{\kappa,\lambda}(\xi,v)n^{\lambda}(\xi,t)d\xi\right\}$$

$$-\frac{\partial}{\partial t}(n^{\kappa}(v,t)g(v,t)) + S^{\kappa}(v,t) - n^{\kappa}(v,t)R(v,t)$$

The integrals in the GDE represent the formation of particles of volume $v$ by collision of small particles and depletion in this size class by collision of particles of volume $v$ with any other particle. $E_\kappa$ is a collection kernel that prescribes collision rates for different forcings. The rates relate either to velocity differences caused by shear, turbulence, Brownian motion, or different settling velocities, or to phoretic forces due to gradients in temperature, concentration, or charge.

The third term accounts for growth by condensation, $g(v, t)$ distinguishes evaporation and ablation, because solid particles suddenly immersed in a high-temperature environment lose mass (independent of the difference in vapour pressures). The last two terms are source and loss terms. Sources include nucleation of particles from supersaturated vapour, evaporation of component(s) from mixed particles leaving a pure particle, and breakup of very large particles by aerodynamic or electric forces. Losses can occur as a result of condensation of vapour on pure particles, coagulation of different species particles, and complete evaporation of particles.

The GDE regulates the competition for vapour and balances the competing microphysics processes such as nucleation, ablation, condensation, evaporation, coagulation and breakup; it is solved in particle radius space and thus seamlessly melds with driver models providing the fireball–cloud thermodynamic field. The GDE balances all the effects together, thereby apportioning pure and mixed particles the correct growth. Solutions specify particle concentration $n^\kappa(v, t)$ for each species ($\kappa$) tracked from the nuclear plasma to definition of respirable and deposited radioactive products. The formulation is nonlinear, underscoring that approximations based on a simple superposition of effects are bound to be incorrect.

The next generation of fallout models combining high-resolution fireball–nuclear cloud solutions with interactive solution of the microphysics GDE and advanced cloud transport models will provide superior assessments of nuclear events and impacts on environment and humans.

## 8.3 WEATHER AND FALLOUT

In the troposphere, weather determines the fate of the radioactive particles or nuclear aerosol. Winds and thermals advect, moisture scavenges and precipitation rains or washes aerosols (fallout) from the atmosphere. The influence is not entirely one way, however; aerosols control cloud formation and thus influence the radiation budget, which in part drives the weather, and most importantly when, where and how much precipitation occurs. The feedback is important and underscores the interactive nature of fallout physics. The interaction of nuclear aerosols and weather is especially important because departure from 'normal' aerosol properties means that rainout and washout patterns are significantly changed. Here, the influence of smoke, dust, and entrained water play a critical role.

The path is direct. Particle morphology, concentration and most importantly chemical properties determine the probability of nucleating water. Multispecies (nuclear aerosol, dust, water, salt, pollutants) microphysics processes are highly competitive, with the largest soluble particles capturing the available moisture and other particles remaining dry in interstitial air. Compared with particles nucleating water, dry particles have very low washout rates. If the nuclear particle is soluble, fallout can be high.

This simple physics accounted for the highly non-uniform deposition pattern of Chernobyl fallout (Warner and Harrison, 1993). Shallow short-lived convective systems deposited much of the $^{137}$Cs/$^{134}$Cs in small areas of Sweden, Wales, and Belarus (fallout near Gomel was 1500 Bq m$^{-2}$). Although the updraft–precipitation pattern was effective in rainout and washout of Cs, much of the interstitial radionuclide aerosol was vented through the top of the system. Other hot spots near hill tops resulted from increased deposition of soluble radionuclides in feeder–seeder cloud systems formed by orographic enhancement. Fallout of the dry components was hemispheric, but low dose and innocuous (although unneeded).

Fallout models currently approximate the aerosol–weather interaction using conventional cloud microphysics. Although such approaches can estimate particle growth, which influences settling speed, they are poor predictors of episodic rain or scavenging—both of which are important for accurate fallout assessments.

There are many advantages to using implicit engineering formulations (such as those based on Kessler (1969) type bulk parameterizations or (more recently) on semispectral Berry–Reinhardt parameterizations (Chaumerliac and Rossett, 1989)). They are keyed to easily calculated bulk measures of the aerosol; they execute rapidly, require little storage and generally are simple to apply. The detail is often impressive, with some versions determining 21 types or forms of particles all based on a few 'bulk' parameters.

The only difficulty is that most of the time, the prescriptions are of questionable accuracy. They can be correct in some cases, especially if the embedding weather duplicates the average conditions the empiricism was built for; generally they are not. Comparisons at different times and by different groups have shown that deviations from actual conditions can be quite large (Soong, 1974; Shiino, 1983; Lee and Hong, 1987; Kogan, 1991). This is not surprising because average weather conditions are usually a poor assumption—poor for cloud prediction over a target, and likewise poor for prediction of winds and rain in specific (battlefield, target, or vacation) areas.

Similarly, bulk microphysics formulations based on *average aerosols* (despite impressive pedigree) poorly approximate particle growth in 'non-average' clouds, cloud development over urban areas, dry deposition, rainout, washout, occult deposition, not to mention chemical changes caused by particle ageing and solubility. The latter is especially relevant as it controls an important pathway for agent integration in terrestrial and aquatic systems and thus influences lethal dose and persistence (Warner and Harrison, 1993).

Nuclear aerosols as a class impose additional constraints on using 'regular' microphysics approximations, because wartime aerosols are not regular atmospheric aerosols. They have different concentrations, morphologies, properties, and behave differently in the atmosphere.

Particle concentration along with the size distribution is a key parameter in all implicit systems. Rate processes such as growth by condensation and coagulation are prescribed as long as concentrations and distributions reasonably represent the empiricism. Greater concentrations that may lead to fog or marine layers rather than accelerated growth of cloud particles (not all clouds rain) generally are not well modelled. Similarly, engineering approximations rarely if at all recognize different balances caused by rapid changes in supersaturation (characteristic of high-energy nuclear clouds). Small *et al.* (1994) showed that such rapid changes in nuclear aerosols lead to episodic nucleation, formation of submicron particles, and eventually multimode size distributions. Departure from average conditions may imply a considerable uncertainty in the eventual size distribution and ultimate rainout, washout, or dry deposition—or simply put, uncertainty in the fallout. Moreover, such uncertainties carry forward and, for example, influence estimates of toxic resuspension.

Particle morphology is an important topic that remains to be included in fallout models. Departures from spherical geometry lead to different growth rates for condensation or evaporation (saturation vapour pressures are lower) and for coagulation (cross-sections and aerodynamic properties change). Both processes control particle development and thus fallout velocity; coagulation influences in-cloud growth as well as capture by raindrops or washout. Uncertainty in those quantities translates directly to uncertainty in the location and intensity (dose) of fallout. Moreover, uncertainty in chemical and physical agent properties impacts calculation of infiltration and migration through pervious surfaces and consequently ecological impacts and decontamination requirements. Growth rates for non-ideal particles is a current area of emphasis in the aerosol community, and extensions of classical microphysics theory could be developed for non-spherical geometries.

Chemical and photic ageing are suitable for treatment in explicit first principle models because changes generally occur in one species and the dynamics of multispecies interactions are changed. Treatments that prescribe process rates for nucleation, condensation/evaporation, and coagulation based on bulk parameters, such as concentration and saturation ratios, have difficulty modifying those rates to reflect chemical or photic change in surface properties. In such cases, accuracy and fidelity depend not only on the microphysics empiricism (and predicting whether the aerosol is shielded by clouds, exposed to sunlight, etc.), but also on whether the appropriate physics is even included.

Similar issues apply to chemical processes, such as surface ageing that changes particles' affinity to water (hydrophobic to hydrophilic), phase changes for elements such as $^{131}$I (three forms are possible—gaseous, particulate and methyl), exchangeable form and solubility of elements such as $^{137}$Cs and $^{90}$Sr.

An immediate jump in capability can be implemented for nuclear aerosols by including new explicit, multispecies, microphysics capabilities with current weather (stochastic or deterministic) algorithms. Uncertainties of using 'normal' aerosol approximations for nuclear aerosols will be eliminated. Moreover, explicit calculation of nuclear size distribution, properties, and changes by chemical, photic and microphysics processes ties in with and provides all the right information for human and ecosystem impact models.

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