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INVESTIGATIONS OF THE DOSE TO MAN FROM
THE WET DEPOSITION OF NUCLEAR AEROSOLS

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INVESTIGATIONS OF THE DOSE TO MAN FROM
THE WET DEPOSITION OF NUCLEAR AEROSOLS*

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I. INTRODUCTION

The U. S. Atomic Energy Commission has for a number of years had a distinct interest in improving the understanding and predictive capability regarding the scavenging of nuclear aerosols from the atmosphere and assessment of the resulting dose-to-man. The motivation for this has originated from questions pertaining to reactor safety and analysis of the fate of nuclear aerosols produced by nuclear explosive systems in various modes of detonation. Research in the Atmospheric and Geophysical Sciences Division at the Lawrence Livermore Laboratory has been concerned with the development of submodels which simulate the processes that take place (a) from the creation of the stabilized cloud, (b) through the period of self-diffusion of the cloud of aerosol, (c) to the three-dimensional regional transport of the aerosol to the scavenging site, (d) the precipitation interaction model, (e) dispersion of raindrops containing the scavenged nuclear aerosol, and (f) construction of a demographic radiation

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hazard model. The evolving submodels addressing these subproblems have been applied in actual assessments to estimate the dose-to-man from the wet deposition of nuclear aerosols. It is the purpose of this paper to summarize the status of development of some of these submodels, to report on the highlights of our assessment activities, and to discuss the uncertainties of our dose-to-man assessments from nuclear aerosol wet deposition in the hope that the identification of these uncertainties and their articulation might lead to information exchange in this regard.

Because of the generally excellent operating record of nuclear safety associated with the power reactor program, there exists little definitive information on the fate of aerosols of this origin in response to scavenging processes in the atmosphere. However, nuclear testing has provided us with evidence, however fragmentary, that rapid scavenging of nuclear aerosols does indeed occur. Examples include the wet deposition "hot-spots" at Troy, New York (1953), at Fargo ND (48 hours after the Diablo Event), and in the Snake River Valley (27 hours after the Cabriolet-Plowshare cratering shot). It is this evidence along with certain clues from investigations of global fallout that has led to a re-examination of the radiation hazard that might be expected from the early scavenging of nuclear aerosols from low yield nuclear explosive systems.

Since most of the present body of data available is concerned with the fate of nuclear aerosols produced from atmospheric tests - balloon shots, tower shots, or venting underground shots, we will focus our attention on nuclear aerosols produced by explosive systems, and in particular, those in the free airburst mode. It is our view that analysis of the free airburst mode are crucial in that the particles so formed are less than 10

microns in diameter and hence remain airborne as a reservoir of fission products and radioiodines whose removal is largely associated with wet deposition. The conventional or traditional view of nuclear aerosols produced by the free airburst mode is as follows, the hazard of close-in exposure from low yield free airburst has generally been regarded as negligible because no particles of sufficient size to fall out are generated. This conclusion may well be true as long as wet deposition processes are excluded. However, in real-world situations involving nuclear safety one may not have the option of excluding wet deposition. It therefore becomes necessary to assess quantitatively the consequences of wet deposition of the radioactive inventory during, for instance, the first 24 hours after such a postulated detonation. In an early study (1971), we examined the potential close-in whole body exposure from external gamma radiation and the thyroid exposure through the forage-cow-milk pathway for hypothetical 1, 10 and 100 kt fission airbursts. Our results indicate that the complete scavenging of the 1 kt cloud, the scavenging of the lower 10% of the 10 kt cloud, or the scavenging of the lower 1-2% of the 100 kt cloud would give infinite external gamma exposures of a few hundred to a thousand rems at distances of 100 kilometers downwind of ground zero. If wet deposition should occur a thousand kilometers downwind without previous precipitation scavenging along the cloud trajectory, the potential external gamma exposure is in the order of 1 rem. Results also indicated that large potential thyroid exposures to man by way of the forage-cow-milk pathway from wet deposition of radioiodine 131 could also occur. The parameters determining the radioactive inventory, the transport speed at the cloud center, and atmospheric diffusion coefficients that govern the

distribution of radioactivity in space at the time the scavenging event is postulated to occur are shown in Table 1. More detailed information on the potential surface exposures are given in Table 2 for these postulated yields and scavengable fractions as a function of downwind distance. It should be noted that these early estimates are applicable on the cloud centerline and indicate the peak value of the exposure; they also assume very rapid scavenging processes that might be associated with convective rain systems.

In view of the magnitude of the potential hazard indicated above, it is frequently asked why there is not more experimental information bearing on this potential hazard. Discussions with U. S. meteorologists providing guidance during atmospheric testing indicate that one of the guidelines adhered to was that no precipitation scavenging along the cloud trajectory should occur following the test for at least a 24 hour period. The fact that no data exist on this wet deposition hazard attest to the success of the advice provided. The data points cited earlier for Troy, New York; Fargo, ND and the Snake River Valley in Idaho involve times exceeding 24 hours, but nonetheless indicate that wet deposition can occur at rates that far exceed dry deposition. In fact the estimates that we have made in the Cabriole and Diablo tests indicate that the full vertical integral of radioactivity airborne at the time of the scavenging event was deposited on the ground at the receptor areas indicated. It should be pointed out that those tests were not free airbursts and the particle size distribution of the debris was probably significantly different from free airbursts. Hence, their extrapolation to conditions for free airbursts is

TABLE 1. Input cloud parameters for exposure calculations.

Yield (kt)	Center height (m)	Thick- ness ^a (m)	Radius ^a (m)	Gross ^b fission (pCi)	¹³¹ I (pCi)	Wind speed (km/hr)	Atmospheric dissipation (ergs/g sec)		Vertical diffusivity (cm ² /sec)	
							Slow diffusion	Fast diffusion	Slow diffusion	Fast diffusion
1	2,840	1760	920	4.44×10^{20}	1.4×10^{17}	39.6	0.5	3	1000	10,000
10	7,000	3060	2400	4.44×10^{21}	1.4×10^{18}	70.2	1	5	1000	10,000
100	11,700	5340	6000	4.44×10^{22}	1.4×10^{19}	72.0	0.7	3.5	1000	10,000

^aCorresponds to two standard deviations from cloud-center concentration.

^bAfter H+1 hr.

TABLE 2. Potential surface exposure
and downstream exposure rates.

Yield (kt)	Fraction deposited (%)	Distance downwind (km)	Infinite whole-body external gamma dose (rem)	Exposure to child's thyroid due to forage-cow- milk pathway (rem)	External gamma exposure rate at time of arrival (R/hr)
1	100	10	25,000	200,000	25,000
		100	400-1200	5000-15,000	35-100
		1000	0.3-1.0	5-15	<0.01
10	10	10	5000	30,000	700
		100	1000-1500	10,000-15,000	150-200
		1000	1-4	20-60	0.015-0.05
100	1	10	800	5500	1200
		100	300-350	3500	45-55
		1000	1.5-5	25-70	0.02-0.06

not justified. In this regard we must look further, perhaps to the clues regarding wet deposition of global fallout.

Much of the nuclear debris or aerosol material placed in the stratosphere has resulted from high yield free airbursts. It is this tracer material that has been subjected in the natural atmospheric system to processes of transport and removal mechanisms of all scales. By examining the fate of this material we may be able to deduce clues concerning the behavior of nuclear tracer material pertinent for the present problem. It is known that the stratospheric residence time for such material is about 6 to 18 months depending upon the height of injection and the season of the year. Whereas the tropospheric residence time is much shorter, possibly from a few days to 30 days with wet deposition removal being the primary mode of depleting this material. The effective deposition velocity associated with dry deposition is small, probably less than 1 cm/sec while that from wet deposition is 20 cm/sec or higher. Hence, the rates of wet removal are probably at least 2 orders of magnitude higher than those for dry removal processes from the tropospheric reservoir. It should also be noted that since 20 cm/sec is considerably smaller than the terminal fall velocity associated with raindrops, there is some other process than the settling rate of raindrops that is rate determining. It is our suspicion that the rate determining processes are within the natural cloud during the interaction of the debris with the cloud water. In any event, wet removal processes appear to be the primary reason that nuclear aerosols are removed from the troposphere efficiently.

The key question in rainout assessment is - Can the same processes operate sufficiently fast to make the close-in wet deposition of nuclear

aerosols a problem of significance in regard to radiation hazard? All of our research to date, even allowing that limitations and uncertainties regarding some of the crucial inputs do exist, leads us to answer the latter question in the affirmative. However, in so answering this question, self criticism of this answer leads us to points of qualification, namely it should be pointed out that the concentrations of debris in the global fallout removal mechanisms are lower than those we are considering, that the aged nuclear aerosols may have acquired surface attached hygroscopic material and that the presence of peripheral material around test devices, even though free airbursted, may make the particle size distribution of the aerosol different or subject to selective fractionation. Beyond these, many of our uncertainties in rainout assessments do in fact stem from our imperfect knowledge of the detailed mechanisms by which the atmosphere cleanses itself.

2. THE STRUCTURING OF THE RAINOUT PROBLEM FOR SOLUTION

It is now instructive to discuss the manner in which the rainout problem may be separated into six submodels. These models will be presented in conceptual form only with the necessary inputs mentioned.

2.1 The Stabilized Cloud Model - following a vent or nuclear detonation a stabilized cloud of nuclear aerosols is formed in the boundary layer of the atmosphere, the troposphere, or the stratosphere depending on the energy of release. By "stabilized" cloud it is generally meant that the vertical dimensions of the cloud are by and large fixed at the time of stabilization. At this time the desired information is the initial distribution of the aerosol in space, the particle size distribution of the aerosol, the specific activity as a function of particle size and surface properties of the particles in regard to their chemical composition and the manner in which that affects nucleation rates for water vapor or other removal mechanisms.

2.2 The Cloud Self Diffusion Stage. After the stabilized cloud is formed there is a residual field of decaying turbulence within the cloud during which the initial distribution of the aerosol at several minutes is modified by the presence of turbulence. The details of this particular stage are poorly understood at this time, but the stage probably only lasts on the order of some 20 minutes.

2.3 The 3-Dimensional Transport Diffusion Model. During the period of time from stabilization until the aerosol cloud is advected to the scavenging site, for example from a half hour to 10 to 15 hours down range, there is a requirement to know the 3-dimensional distribution of the aerosol in space. Hence there is a requirement to calculate the regional 3-dimensional transport of the aerosol including anisotropic diffusion processes and to include the effects aerosol aging during this time period.

2.4 The Scavenging Interaction Model. The scavenging interaction model in principle would include the description of the time history of the 3-dimensionally distributed aerosol and the interaction of that aerosol with the natural cloud system and the time dependent rates of the various removal mechanisms which may be functions of dynamic cloud evolution properties. In addition, we may need to include competition or interaction with natural aerosol components which might become part of the aerosol during its downwind transport or might become entrained into the natural cloud system along with the nuclear aerosol. Such a model will probably need to be so structured that the nuclear aerosol remaining in the dry air, that associated with cloud water, and that associated with rainwater must have spatial structure and time dependency in a conservation sense.

2.5 The Rainwater Dispersion Model. During the interaction of the nuclear aerosol with the natural cloud system the nuclear debris associated with the rainwater component is that which will reach the ground surface quickly. In so doing, depending on the wind structure in 3-dimensions, and the rainwater drop spectrum there can be a dispersion of the debris during its fall to the ground. This aspect of the problem needs to be treated, in principle, by a reasonably sophisticated raindrop dispersion model that is driven by outputs of the interaction model. However, in perhaps the most pertinent rainout scenario, that involving a convective cloud system, there is observational evidence that the precipitation shafts are largely vertical and that this model can be a very simple one - a vertical precipitation column.

2.6 The Demographic Radiation Model. Given the spatial and time dependent ground level wet deposition (the net result of the previous five submodels), it is desirable to have a demographic radiation hazard model which converts that deposition into dose rate or integrated dose from the external gamma radiation field, including the effect of shielding and/or enhanced shielding due to the infiltration of the particles into the surface material on which it is deposited.

Recognizing that our knowledge and the required inputs to each of these submodels is imperfect at this time, we will nonetheless proceed to describe some of the model developments and some of our rainout assessment results in the following sections.

3. APPLIED RESEARCH HIGHLIGHTS

It is now pertinent to present some of the applied research highlights relevant to the six submodels included in the structure of our rainout assessment capabilities.

a. Modeling of the Nuclear Aerosol Formation.

The predominant factor governing the rate of scavenging of aerosol particles is their size. Because of a lack of reliable experimental data on the size distribution of debris particles from a free airburst, a calculation based on homogeneous nucleation theory is used to deduce the expected size distribution assuming there is only one predominant condensing material.

As a nuclear fireball expands and cools to temperatures below 2000 K certain materials condense by the formation of small liquid droplets; the rate of condensation depends upon the temperature and supersaturation of the condensing material. Once droplets are formed they grow by diffusion of the material to the droplet surface and, if the droplet number concentration is high enough, by coagulation of droplets. Calculation of the supersaturation as a function of time permits a further calculation of the number of droplets as a function of droplet radius. A rather surprising result is that for all cases examined the concentration of droplets is too low for droplet coagulation to be important in shaping the size distribution.

A size distribution calculated using the model is shown in Figure 1. Over a fairly wide range the size distribution is symmetric indicating the distribution is approximately log-normal. The abrupt cutoff at the small particle sizes reflects the fact that late in the nucleation period small droplets evaporate due to decreasing supersaturation. The most important conclusion reached thus far is that the geometric standard deviation or sharpness of the peak of the size distribution is very sensitive to the surface energy of the condensing material, materials with high surface energies having more sharply peaked distributions. Most of the debris particles have sizes on the order of $0.1 \mu\text{m}$.

Before the calculations from this model can be considered realistic several improvements are required. First, there are usually at least two major condensing species, both of which must be considered. The major difference between the first and later condensing materials is that the later materials will probably condense on the particles formed when the first phase condensed, and hence at a much lower supersaturation. Second, the possibility that the first material condenses on ions rather than homogeneously must be considered.

b. Removal Rates for the Relevant Scavenging Mechanisms.

For a debris particle to be deposited by rain it is necessary that the particle be incorporated in or attached to a raindrop that falls to the ground. There are several mechanisms potentially active in clouds that can lead to scavenging of debris particles

Fig. 1. Droplet size distribution. Plotted on the ordinate is $dN(r)/d \ln r$ where $N(r)$ is the number of droplets with radius less than r/cm^3 .

by raindrops, and the rate of collection as a result of each mechanism depends largely on the size of the debris particle. In addition, there are two-step processes that occur at significant rates which also lead to deposition of debris on the ground. Two-step processes involve the scavenging of debris particles by cloud droplets of negligible terminal velocity and the subsequent inertial capture of the droplet by a falling raindrop transporting the debris to the ground. Since the inertial capture of cloud droplets which are larger than $10\text{ }\mu\text{m}$ generally occurs quite rapidly in precipitating clouds, the rate limiting step in the two-step processes is the rate of scavenging of debris particles by cloud droplets.

Inertial capture of droplets or debris particles by falling raindrops occurs efficiently in clouds when the droplets or particles are of micron sizes as shown by Curve I in Figure 2. However, there are very few debris particles resulting from a free airburst of such large sizes, so that direct collection of debris is usually negligible. It is also apparent in Figure 2 that cloud droplets which are typically $5\text{--}10\text{ }\mu\text{m}$ in radius are scavenged very quickly by raindrops.

Brownian motion of small particles can lead to significant scavenging of debris by cloud droplets and to a lesser extent by raindrops. Curve II in Figure 2 gives the rate of capture due to Brownian motion; it is apparent that this mechanism is only efficient for particles with radii less than $10^{-2}\text{ }\mu\text{m}$, but theory suggests that there is little debris in particles of these small sizes.

Previously we have shown that freshly formed debris particles have sizes close to $0.1 \mu\text{m}$, that is, in the size range where neither inertial capture nor Brownian motion are significant scavenging mechanisms. There are processes active in clouds that can effect scavenging of particles in this size range; however, each one generally requires a special set of circumstances.

It is observed in the atmosphere that when air becomes slightly supersaturated with water vapor, small droplets form on aerosol particles and grow quickly to a size of several microns. These aerosol particles have acted as condensation nuclei and are enclosed by the droplets that have formed around them. Experiments indicate that soluble particles larger than about $10^{-2} \mu\text{m}$ and insoluble but wettable particles larger than about $10^{-1} \mu\text{m}$ are normally activated in the atmosphere as condensation nuclei. This process is indicated by the dashed and dotted curves in Figure 2, where the small debris particles are assumed to act as condensation nuclei, grow quickly as cloud droplets to sizes of $5\text{-}10 \mu\text{m}$, and are inertially captured by falling raindrops. This process is only effective, however, for those debris particles that pass through a region of the cloud where slight supersaturation exists. How long it takes for an aerosol particle to be transported through such a region is really the limitation on the rate of this removal mechanism.

Debris particles that have not served as condensation nuclei but are present in a cloud can become attached to droplets by any of several mechanisms.

Fig. 2. Available information on removal or rainout coefficients as a function of particle size. A rain rate of 1 cm/hr and a maximum drop radius of 0.2 mm is assumed. Curves I and II, are described in the text.

Thermo- and diffusiophoretic forces can lead to scavenging of small particles by nearby droplets. For evaporating hydrometers the net phoretic force is attractive and scavenging can be enhanced. The rate, however, tends to be rather slow, and it is very likely that the droplets will evaporate completely before reaching the ground. For growing droplets, the net force on a particle due to thermo- and diffusiophoresis can be either towards or away from the droplet and is generally of small magnitude.

Small scale turbulence within a cloud can lead to acceleration of debris particles relative to nearby droplets or drops leading to enhanced scavenging. This mechanism is often dismissed as unimportant, but there is no theoretical argument showing that this is correct.

Electrical forces are very likely of great importance in altering scavenging by attachment. Although it has been shown using the best available data that attachment due to electric charges on natural aerosol particles and hydrometeors and the normal electric fields in non-thunderstorm clouds occurs at rates normally considered too slow, significant attachment rates can also be calculated within the uncertainty of the parameters. The fact that debris from a free airburst is radioactive completely alters the electrical phenomena within clouds since the particles are charged by α and β decay and the ion production rate may be orders of magnitude increased in comparison to the normal atmosphere.

Most of the available field data, which uses non-radioactive tracers, indicate that particles in the $0.1 \mu\text{m}$ size range are efficiently scavenged by precipitation.

c. Modeling of the Three-Dimensional Distribution of the Nuclear Aerosol.

In this section we describe an emerging validated capability for calculating the three-dimensional distribution of a nuclear aerosol after its down range transport and diffusion to the site of scavenging. By way of introduction and summary it can be said that during the early lifetime of a nuclear aerosol cloud, that is while the cloud may have an over-kill capability (e.g. a capability to deliver doses far in excess of say 150 rem), any process that spreads the cloud over a larger area, in fact, leads to cases of greater potential hazard than those previously estimated in UCRL-51154. The parameters that we consider and vary over reasonable ranges in order to examine their effect on rainout deposition patterns include vertical wind shear, time of on-set of rain, intensity of rain, duration of rain and the removal rate of the scavenging mechanism selected (10^{-3} per sec or 10^{-4} per sec). In the future as more is learned about precipitation interaction scenarios and about the spatial distribution removal rates with the natural clouds it would be quite possible that it would be appropriate to examine other removal rates.

1. Description of ADPIC

ADPIC is a hybrid Lagrangian Eulerian 3-dimensional particle-in-cell code for calculating the transport and the anisotropic diffusion of a pollutant from its source to its temporal and regional distribution at arbitrary times. ADPIC represents a convenient mathematical framework in which to include the following effects on a regional basis: speed and directional wind shear; occurrence of calms; space variable surface roughness; wet and dry deposition; radioactive decay; gravitational settling; space and time dependent eddy diffusion parameters; and single or multiple sources of continuous or instantaneous nature. ADPIC solves the 3-dimensional advection diffusion equation in flux conservative form using a pseudo-velocity technique for a given regional mass consistent advection field in 3-dimensions and in time. In this method the Lagrangian particles represent the activity distribution and concentration associated with the nuclear aerosol within the structure of an Eulerian grid (the governing equations are shown in Figure 3). The chief advantages of this ADPIC approach are (a) the artificial diffusion inherent in purely Eulerian finite difference codes is practically eliminated and (b) the Lagrangian coordinates representing mass or activity can be tagged so that the age of each particle is known at all times. Two versions of ADPIC have been developed: one version uses a fixed regional

Advection-diffusion equation:

$$\frac{\partial \chi}{\partial t} + \vec{U}_A \cdot \nabla \chi = \nabla \cdot K_{ij} \nabla \chi$$

Assume:

$$\nabla \cdot \vec{U}_A = 0 \quad (\text{Incompressibility})$$

Define:

$$\vec{U}_D = -K_{ij} \frac{\nabla \chi}{\chi} \quad (\text{diffusivity velocity})$$

Pseudo-velocity form of diffusion-advection equation:

$$\frac{\partial \chi}{\partial t} + \nabla \cdot (\vec{U}_p \chi) = 0$$

$$\text{where } \vec{U}_p = \vec{U}_A + \vec{U}_D$$

and $\vec{U}_p$ is the pseudo velocity vector
 $\vec{U}_A$ is the advection velocity vector
 $\vec{U}_D$ is the diffusivity velocity vector
 K_{ij} is the diffusivity tensor
 χ is the concentration

Fig. 3. Governing equations for ADPIC.

Eulerian grid so that complex surface boundary conditions may be incorporated in the solution; the second version uses an expanding grid that travels with the center of the pollutant cloud over flat terrain and automatically expands with the expanding cloud. The latter version is applicable to sensitivity studies in which a large number of parameters are varied. The grid spacing and the eddy diffusivity velocity algorithm used in ADPIC for the 2-dimensional case is shown in Figure 4.

Each ADPIC time cycle (Δt) consists of an Eulerian and a Lagrangian part. As shown in Figure 5, in the Eulerian part, the diffusivity velocity vector $\vec{U}_d$ and the pseudo velocity vector $\vec{U}_p$ are calculated as functions of the concentration field χ , the eddy diffusivity tensor K_{ij} and the mass consistent regional flow field $\vec{U}_a$. In the Lagrangian part, the new position vector, $\vec{R}$, for each Lagrangian particle is calculated from its old position vector plus the displacement of the particle in the pseudo-velocity vector field $\vec{U}_p$. It should be mentioned that the advective vector field $\vec{U}_a$ could be furnished externally by a regional mass consistent objective analysis code, a regional predictive meteorological code, an urban heat island simulation code, or a hydrodynamic model of a convective system. We have, of course, not achieved all of these objectives at this time, and will report the results when utilization of this code includes the $\vec{U}_a$ input derived from regional objective analysis codes as a higher level of sophistication.

Fig. 4. Grid spacing and eddy diffusivity velocity algorithm used in ADPIC (two-dimensional example).

ADPIC time cycle:

Eulerian part:

$$\bar{U}_D = -K_{ij} \frac{\nabla x}{x}$$

$$\bar{U}_P = \bar{U}_A + \bar{U}_D$$

Lagrangian part:

$$\bar{R}_{new} = \bar{R}_{old} + \bar{U}_P \cdot \Delta T$$

Fig. 5. ADPIC time cycle.

It is perhaps pertinent to explain briefly in a simple 1-dimensional example the way in which the ADPIC formulation eliminates the fictitious diffusion plaguing the purely Eulerian finite difference approach. In the upper portion of Figure 6, we show the fate of a pollutant in a 3 cell simulation of diffusive transport where all of the pollutant is in the first cell initially ($T=0$). At the end of a small time step ($T=1$) a portion of the pollutant in the first box has been distributed to the second cell and, in like manner, at the end of the second time step ($T=2$), a portion of the pollutant has been transported to the third cell. During successive time steps pollutant originating in the first cell is transmitted through the grid "too fast" because the pollutant in any cell with an adjacent empty cell is instantaneously shared. In contrast to this, the particle-in-cell treatment of diffusion moves each particle with the vector velocity $\vec{U}_p$ which incorporates the diffusive transport. The particles are seen in the lower part of Figure 6 to move in a more physically consistent manner.

The verification studies of the ADPIC solutions against closed form analytical solutions are summarized in Table 3. The results of these validation studies indicate that the standard deviation of the pollutant distribution was reproduced by ADPIC with a maximum error of about 5% and a mean error of about 2% in the standard deviation for the known solution. The importance

Fig. 6. Simplified illustration of the elimination of the fictitious diffusion in ADPIC.

TABLE 3. ADPIC verification against closed Gaussian solutions.

For initially Gaussian, instantaneous, and continuous sources, ADPIC has been found to agree with analytical solutions to the diffusion-advection equation to within 5% of the standard deviation for the following cases:

<u>Case</u>	<u>Description</u>
1	Instantaneous source; constant-K diffusion.
2	Instantaneous source; scale-dependent $K(t)$ diffusion.
3	Instantaneous source; constant-K diffusion in simple vertical speed shear $U=U(z)$, $V=W=0$.
4	Continuous source; constant-K diffusion.
5	Continuous source; constant-K diffusion; advection= $U=2$ m/s.
6	Continuous source; constant-K diffusion; advection= $U=10$ m/s.

of this result in the several validation studies is that the ADPIC code represents a reasonably accurate calculation framework for attacking regional transport and diffusion problems in multi-dimensional space for transient regional flows. Further, the accuracy of the method is sufficiently high that the residual errors are normally small in comparison to our knowledge of the source term. The quality of the ADPIC solutions are primarily given by a knowledge of the temporal spatial regional flow fields and the spatial distribution of the eddy diffusion processes. (Some of the validation results for the ADPIC code are discussed in detail in UCRL-75731.)

Figure 7 displays the relative particle concentration per cell at the nuclear aerosols cloud center as a function of time for both the analytical solution and for the test case of scale dependent diffusion versus that calculated by ADPIC; the indicated excellent performance of the ADPIC code is shown in this inter-comparison.

We now present some of the results of the application of ADPIC to the investigation of effects of noninstantaneous wet deposition and the effect of shear on the wet deposition radiation patterns that would result from a postulated 1 kt fission aerosol cloud. The parameters utilized in this study are shown in Table 4. Figures 8 and 9 show the isopleth of infinite whole body dose from the external gamma field deposited under various conditions. Problem A is for instantaneous deposition, no vertical wind shear, and mean time of deposition of 900 seconds;

Fig. 7. Scale-dependent diffusion—plot of relative particle concentration per cell at cloud center vs time.

TABLE 4. Description of the 1-kt, fission problem.

Number of ADPIC cells = $20 \times 20 \times 20$	
Number of particles = 11,900	
Initial cloud center height = 3000 m	} at stabilization time H + 600 s
Initial cloud thickness ^a = 1860 m	
Initial cloud radius ^a = 950 m	
Yield = 1 kt at H + 1 h	
Radioactive decay $\propto t^{-1.2}$ (t in h)	
Mean advection velocity = 11 m/s	
Speed shear, $dU/dz = 0.0015 \text{ s}^{-1}$	
Directional shear, $dV/dz = 0.001 \text{ s}^{-1}$	
Horizontal scale-dependent diffusion, $K_h = \epsilon^{1/3} (\sigma_0^{2/3} + \frac{2}{3} \epsilon^{1/3} t)^2 \text{ m}^2/\text{s}$ with $\epsilon = 10^{-4} \text{ m}^2/\text{s}^3$	
Vertical diffusion, $K_z = 0.1 \text{ m}^2/\text{s}$	
Rain rates, $\Lambda = \infty, 0.001 \text{ s}^{-1}$ and 0.0001 s^{-1}	
Rainout over the entire cloud	
Rainout times ^b = 0-1800 s and 3600-5400 s	

^aCorresponds to two standard deviations from cloud center.

^bT=0 corresponds to stabilization time H + 600 s.

Problem A: $\Lambda = \infty$; no shear, total vertical integral at $T = 900$ s, isopleths for 1500, 150, 15, 1.5 rad_∞ .

Problem B: $\Lambda = 10^{-3} \text{ s}^{-1}$, no shear, raintime $t = 0-1800$ s (from stabilization), isopleths for 1500, 150, 15, 1.5 rad_∞ .

Fig. 8. Isoleths of infinite dose from mean time of arrival for problems A and B.

Problem C: $\Lambda = 10^{-3} \text{ s}^{-1}$, $dU/dz = 0.0015 \text{ s}^{-1}$, $dV/dz = 0$, raintime $t = 0-1800$ s, isopleths for 1500, 150, 15, 1.5 rad_∞ .

Problem D: $\Lambda = 10^{-3} \text{ s}^{-1}$, $dU/dz = 0.0015 \text{ s}^{-1}$, $dV/dz = 0.001 \text{ s}^{-1}$, raintime $t = 0-1800$ s, isopleths for 1500, 150, 15, 1.5 rad_∞ .

Fig. 9. Isoleths of infinite dose from mean time of arrival for problems C and D.

Problem B is for a removal rate of 10^{-3} per sec, no vertical wind shear and rain occurring from time of stabilization to 1800 seconds; Problem C is for a removal rate of 10^{-3} per sec, a nominal speed shear, no directional wind shear and a duration of rain from time of stabilization to 1800 seconds; Problem D is for removal rate of 10^{-3} per sec, a nominal vertical speed shear, a nominal directional wind shear in the vertical, and a duration of rain from time of stabilization to 1800 seconds. From examining this sequence of problems it is seen that the effect of noninstantaneous wet deposition on the radiation pattern at early times is in fact a most significant effect in determining the area covered by the 150 rad infinite dose contour. The resulting area in Problem B is approximately a factor of 2 larger than that in Problem A even though only 80% of the reservoir of the nuclear debris is on the ground in Problem B. The effects of shear on early wet deposition patterns appears to be much less than noninstantaneous wet deposition. Table 5 summarizes the other problems run in this parameter variation study.

TABLE 5. Survey problems and results for 1-kt rainout.

Problem No.	Speed shear $\frac{dU}{dz}$ (s^{-1})	Directional shear $\frac{dV}{dz}$ (s^{-1})	Λ (s^{-1})	Rain period ^b (s) start/stop	Peak dose ^c (rad)	Fraction of ^b initial activity remaining after rainout	Approximate area ^e inside 150-rad contour (km^2)
A	—	—	∞^a	900/900	11,000	0	12
B	—	—	10^{-3}	0, 1800	7,700	0.2	20
C	0.0015	—	10^{-3}	0, 1800	7,700	0.2	21
D	0.0015	0.001	10^{-3}	0, 1800	7,700	0.2	21
E	—	—	∞^a	4500, 4500	850	0	78
F	—	—	10^{-3}	3600, 5400	480	0.01	66
G	0.0015	—	10^{-3}	3600, 5400	450	0.01	63
H	0.0015	0.001	10^{-3}	3600, 5400	350	0.01	63
I	0.0015	0.001	10^{-4}	0, 1800	830	0.7	13
J	0.0015	0.001	10^{-4}	3600, 5400	50	0.4	0

^aInstantaneous deposition.^bFrom stabilization time $H = 600$ s.^cInfinite dose from mean time of arrival.^dInitial activity defined as $4.4 \times 10^{+8}$ Ci at $H = 1$ h.^eEstimated from contour plots.

3. MODELING OF THE TIME DEPENDENT INTERACTION OF THE AEROSOL WITH NATURAL CONVECTIVE CLOUDS.

As debris is transported downwind into a region where precipitation is occurring, the spatial distribution of the debris is greatly altered by the field of motion of the rain cloud. Precipitation rates greater than a few mm/hr generally occur as a result of local convection and are accompanied by vertical motion that is comparable in magnitude to the horizontal motion. Therefore, the spatial distribution of debris in a cloud must take account of the local advection field. Two factors which control the amount of debris deposited by precipitation are the rate of the microphysical scavenging mechanism and the dynamic history of the cloud. These two processes continuously interact with and influence each other so that it is necessary to consider both simultaneously to evaluate the deposition of debris.

Since the most significant early time deposition occurs because of an interaction with a convective cloud, one- and two-dimensional numerical models of convective clouds are being developed that incorporate a representation of scavenging and deposition of debris. For rainout of debris particles in the size range of $0.1 \mu\text{m}$ the primary method of scavenging is postulated to be collection of suspended debris in cloud droplets during condensation nucleation and the subsequent inertial capture of these polluted droplets by falling raindrops. Other possible collection mechanisms are neglected, including inertial capture of debris particles by falling raindrops and attachment of debris to droplets or drops because of phoretic, diffusive, or electrical forces. Resuspension of debris because of evaporation of droplets and drops is modeled.

Results using the one-dimensional model are summarized by Figure 10 which shows the time history of the vertical integrals of debris (1) in nonfalling cloud droplets, (2) in falling raindrops, and (3) suspended in cloud air but not inside or attached to droplets or drops from a typical convective cloud with a base at 1.4 km and a top of 4.2 km. Also included is the history of accumulation of debris on the ground; all values are given in units relative to the initial maximum concentration of debris in air before interaction with the cloud. The mixing ratios of debris in cloud air, cloud droplets, and raindrops are shown in Figure 11, 12 and 13 respectively. When the cloud forms at about 5 minutes the debris particles which serve as condensation nuclei are transferred from cloud air to cloud droplets. As the cloud grows from 5-30 minutes, debris in cloud air and in droplets is transported upward. When raindrops form out of droplets at 20 minutes, debris is transferred from droplets to drops. As the drops fall and collect droplets from 30-75 minutes they also scavenge the debris in the collected droplets. When the raindrops reach the ground the debris in the drops is deposited. From 55 to 75 minutes debris is resuspended below cloud base because of the evaporation of smaller drops. For this particular example about 2/3 of the initial vertical integral of the debris concentration is rained out.

Two important parameters describing the debris have substantial influences on the model predictions of rainout amount, the initial height of the debris before interaction and the efficiency with which debris particles serve as condensation nuclei.

Fig. 10. Time-history of vertical integral and debris accumulation for debris initially centered at 1740 m. This is the nominal height of a 0.42-TJ (100-ton) free airburst.

Fig. 11. Mixing ratio of debris in cloud air. Contour lines give the mixing ratio of debris in relative amount of debris per unit mass of dry air. The debris contributing to this phase is debris in the vertical column of the cloud, but not inside droplets or drops. Contour levels are 0.8, 0.6, 0.4, 0.2, 0.1, 0.05, 0.025, 0.01, 0.005, 0.002, 0.001, 0.0005.

Fig. 12. Mixing ratio of debris in cloud droplets. Contour lines give the mixing ratio in relative amount of debris per unit mass of dry air for debris contained in non-falling cloud droplets. Contour levels are 0.4, 0.3, 0.2, 0.15, 0.1, 0.05, 0.025, 0.01, 0.005, 0.002, 0.001, 0.0005.

Fig. 13. Mixing ratio of debris in raindrops. Contour lines give the mixing ratio in relative amount of debris per unit mass of dry air for debris contained in falling raindrops. Contour levels are 0.4, 0.3, 0.2, 0.15, 0.1, 0.05, 0.025, 0.01, 0.005, 0.002, 0.001, 0.0005.

A summary of a parameter study showing how deposition of debris depends on the height of the peak of an initial Gaussian vertical distribution is given in Table 6. As the initial height of the debris is moved upward from cloud base, the deposition of debris decreases to very small fractions of the initial vertical integral. When the debris is initially centered near cloud base the deposition amount is comparable in magnitude to the initial vertical integral. When the debris is located well above cloud base, debris enters the cloud primarily through the sides, but since lateral entrainment is a relatively slow process, the amount of debris inside the cloud is small. Furthermore, since entrained debris enters the cloud above the base, it does not pass through the condensation nucleation activation zone and, by the assumptions of the scavenging model, cannot be collected by droplets or drops. The combination of these two factors lead to the much smaller deposition amounts for high level debris.

Additional parameter studies show that the amount of deposition is approximately proportional to the fraction of debris that is activated as condensation nuclei as the debris particles pass through the activation zone near cloud base. When all debris passing through the activation zone is scavenged by droplets, the rainout amount is 1.32 times the initial vertical integral, i.e. larger than the amount of debris initially in a vertical column of the atmosphere. For thicker clouds and low level initial distributions of debris this rainout amount is several times as large as the initial vertical integral; this fact is a direct result of vertical transport and efficient scavenging.

TABLE 6. Parameter study of height of pollutant

Height (m)	Standard Deviation (m)	Maximum V.I. in Cloud Droplets (@ 30 min)	Maximum V.I. in Raindrops (@ 57 min)	Maximum Suspended in all phases (@ 57 min)	Pollutant Deposited on Ground
1340	560	0.62	1.36	2.75	0.88
1740	560	0.46	1.02	2.12	0.66
2140	560	0.19	0.41	1.01	0.27
2440	570	0.05	0.10	*	0.07
2840	580	0.003	0.007	*	0.006

* Initial value is the maximum.

4. SELECTED HIGHLIGHTS OF ASSESSMENT ACTIVITIES.

One of the earliest assessment questions of a practical nature which we addressed pertains to the duration of time that a nuclear aerosol cloud from a low yield free airburst would have the capability of delivering an infinite whole body dose of 150 rad, if it were instantaneously deposited. To answer this practical question we numerically simulated the transport and diffusion of such source clouds estimating the vertical integral of radioactivity on the cloud centerline trajectory as a function of time. Our results indicate that for a postulated 1 kt free airburst cloud subjected to nominal wind speeds the area inside the 150 rad contour is about 8 square kilometers at one-half hour, about 85 square kilometers at 2-1/2 hours and 0 at 3.7 hours due to the combination of atmospheric diffusion of radioactive decay. For the postulated 10 kt fission device the corresponding data are as follows: at 1/2 hour the 150 rad contour has an area of approximately 60 square kilometers, at 8 hours the maximum area is approximately 800 square kilometers and the area of 150 rad dose contour drops to zero at approximately 9.6 hours. These estimated time intervals are a guide to the potential radiation hazard from wet deposition during this period. They are not intended to give detailed estimates of what might take place in specific scavenging scenarios. As indicated in the previous chapter, noninstantaneous deposition and shear can alter these areas upwards or downwards by a factor of about 2.

A second practical assessment question which needs to be addressed is to estimate the probability of a rainout event at representative middle-latitude climates wherein we define the rainout event as an inter-

action of a debris cloud with a natural scavenging system that results in a finite area enclosed within the 150 rad contour. It should be quite clear that if we had a hypothetical receptor area at a place where it always rained, the probability of a rainout event would be 100%. At real-world middle latitude locations the probability of a rainout event is (roughly) the sum of the probability of rain at that location plus the probability of a nuclear aerosol cloud being born in clear air and then having an opportunity to interact with the scavenging system during the time at which the cloud has the potential for delivering 150 rad. In order to construct a regional statistical model of the rainout probability we proceed as follows. Three different interaction scenarios between the debris cloud and the natural cloud system are postulated: (a) interaction I defines the creation of the nuclear debris cloud within a large scale range shield with the potential for rapid deposition on the ground (by rainout) of the debris aerosol; (b) interaction II involves the debris cloud created in clear air and subjected to relative advection with respect to the large scale synoptic rain pattern, the debris cloud in this scenario overtakes the synoptic scale rain pattern and provides a rainout potential; (c) interaction III is defined as a debris cloud created in clear air that is subsequently transported downwind adjacent to convective cells, during which transport a portion of the debris cloud is entrained by a convective cell and is deposited on the ground as a local wet deposition hot spot. Figure 14 illustrates a typical middle latitude synoptic disturbance in which interactions I, II and III are shown in the respective parts of the disturbance in which they might be important.

Fig. 14. Schematic diagram of a typical synoptic system (summer season) with a 72-hr period. The contributions that the three sectors could make to interaction with nuclear debris are summarized as follows: Sector I, $5/72 \approx 7\%$ of the 72-hr period; Sector II, $2/72 \approx 3\%$; Sector III, $7/72$ ($1/3$) $\approx 3\%$.

In order to arrive at our estimates of probability, we have assumed that the periodicity of storms of this general structure is approximately 72 hours. To arrive at the probability of a rainout event we must estimate the fraction of the 72 hour period during which the postulated interactions are viable. From knowledge or estimates of the mean speed of storms over an arbitrary point, we estimate (see Table 7) that rainout interaction I can occur about 5 hours out of the 72 hour period; that interaction II can occur for approximately 2 hours of the 72 hour period; and that interaction III can occur for approximately 7 hours out of the 72 hour period with a probability of approximately $1/3$ that a scavenging interaction occurs. Thus the sum of these three contributions to the probability of a rainout event is estimated to be about 13% for a 1 kt nuclear aerosol cloud. The frequency weighted area enclosed by the 150 rad dose contour is estimated to be 12 square kilometers. In citing this probability statement we recognize that regions that have meteorological conditions somewhat different from those postulated, regions in which mountainous terrain affect the intensity and frequency of precipitation, etc. would have probabilities of a rainout event that could be quite different from those indicated here. However, our estimate is given as preliminary guidance such that rainout assessments may take the form of a probabilistic area statement for particular assumed energy releases. This probability statement can in turn be converted to estimate man-rem. If we assume a nominal homogeneous demographic distribution, we estimate that about 400 people would receive a million man-rem in the rainout event just described.

TABLE 7. Summary of the contributions to the probability of a rainout event.

Interaction	Time fraction	Contribution (%)
I	5/72	7
II	2/72	3
III	7/72 · (1/3) ^a	3
Total		13

^aOnly one-third of the clouds are scavenged.

Inclusions in An Annotated Bibliography on Rainout.

UCRL-51164	Potential Exposures from Low-Yield Free Air Bursts 12/26/71	J. B. Knox, T. V. Crawford, W. K. Crandall
UCRL-51328 Rev. 1	An Investigation of Scavenging of Radioactivity from Nuclear Debris Clouds: Research in Progress 1/17/73 (Rev. 10/16/73)	W. K. Crandall, C. R. Molenkamp, A. L. Williams, M. M. Fulk, R. Lange, J. B. Knox
UCRL-51530	Rainout Studies at Lawrence Livermore Laboratory 2/11/74	J. B. Knox, A. L. Williams
UCRL-51627	A One-Dimensional Numerical Model of Precipitation Scavenging with Application of Rainout of Radioactive Debris 7/17/74	C. R. Molenkamp
UCRL-51614	Further Numerical Model Studies of the Washout of Hygroscopic Particles in the Atmosphere 7/2/74	G. J. Stensland
UCRL-75731	The Adaptation of a Three-Dimensional Atmospheric Transport-Diffusion Model to Rainout Assessments 7/30/74	R. Lange, J. B. Knox

5. CONCLUDING REMARKS

In general, the results and the physical insights we have thus far gained from our investigations of rainout would indicate that the potential radiation hazard associated with rainout of nuclear aerosols is of great significance and for those reasons the problems alluded to in this paper are deserving of further in-depth research and assessment activity. The rainout effects associated with the detonation of free airburst nuclear devices appear to be capable of creating significant radiation hazards in areas far removed, a few hours travel time for 1 kt, from the close-in area.

It should be reported that there is a spectrum of opinion regarding the importance of rainout effects within our country at this time. In making the rainout assessments of Section 4, we have made the input selections shown in Table 8 for the assessment of the rainout hazard from a 1 kt fission free airburst. The principle reason for this spectrum of opinion really pertains to the selection of these inputs. It is important therefore to indicate these differing judgments about the inputs. First the particle size distribution of the nuclear aerosol generated from a free airburst of low yield and its specific activity distribution as a function of particle size constitutes perhaps a most important difference of opinion. If the aerosol created is such that the particles are much less than a tenth of a micron in size, and if the specific activity distribution were such that the bulk of the radioactivity were on the smaller particles (as some would assume) the rainout hazard is significantly reduced. Second, the specification of the relevant scavenging mechanism is a subject of debate; in our assessments we have selected in-cloud

TABLE 8. LLL position on rainout characteristics.

Particle size distribution	90% mass $>0.1 \mu\text{m}$
Specific activity	Radioactivity distributed with mass
Relevant removal mechanism	Rainout, in-cloud scavenging
Removal rate	$\sim 10^{-3} \text{ sec}^{-1}$
Frequency of a rainout event	$\sim 13\%$ based on representative summer regional model.
Assessment implied by above statements	Potential for 150 rad $\sim 13\%$; subjective confidence ~ 2 ; summer season.

scavenging as the most probable mechanism for low yield nuclear devices or aerosols created by a comparable energy release. If, however, there is very little energy in the release and the aerosols are created close to the ground washout might well be the primary removal mechanism. In general, there is an order of magnitude difference in the removal rates depending on whether rainout or washout is selected. Third, the time scale of important interactions between the aerosol or debris cloud and the natural rain cloud need better definition as well as the details of the interaction. There is also uncertainty about the fate of aerosols entrained in convective clouds along the sides or at the top. Fourth, development of a probabilistic statement for a rainout event has several different approaches in the U.S. such that the results differ. However, the first three above reasons for uncertainties in rainout assessments would be the primary causes of differences in the rainout probability statements.

Because of the importance of nuclear safety and the possible significance of rainout, the scientific community would welcome any insights or knowledge bearing on these uncertainties. We would welcome contributions from all participants in resolving these questions.

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