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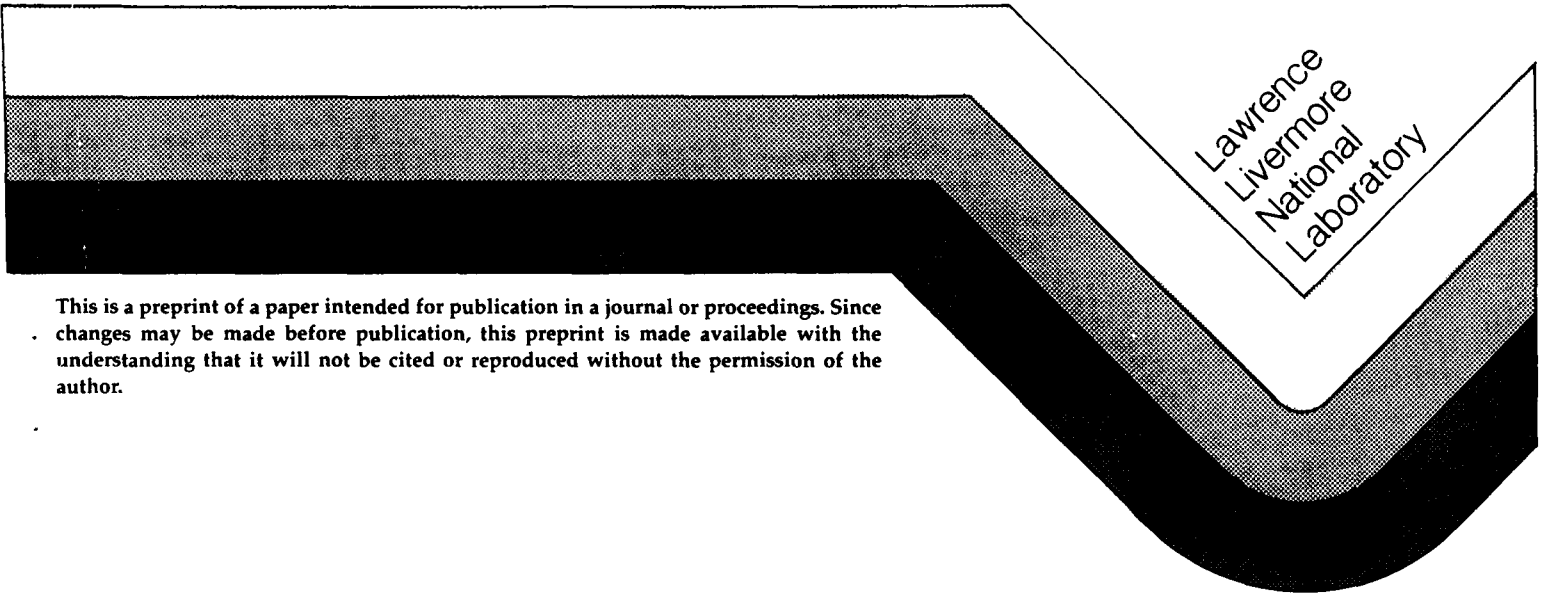
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**The Technical Basis for Air Pathway  
Assessment of Resuspended Radioactive  
Aerosols: LLNL Experiences at Seven  
Sites Around the World**

Joseph H. Shinn

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# THE TECHNICAL BASIS FOR AIR PATHWAY ASSESSMENT OF RESUSPENDED RADIOACTIVE AEROSOLS: LLNL EXPERIENCES AT SEVEN SITES AROUND THE WORLD.

Joseph H. Shinn  
Health and Ecological Assessment Division  
PO Box 808 L-453  
Lawrence Livermore National Laboratory  
Livermore, CA 94551-9900  
(510) 422-6806

## INTRODUCTION

There is a large uncertainty in quantifying the inhalation pathway and the aerosol emission rate in human health assessments of radioactive-contamination sites. The need for site-specific assessments led to formation of our team of specialists at LLNL, who have participated in numerous field campaigns around the world. Our goal was to obtain all the information necessary for determining potential human exposures and to estimate source terms for turbulent transport of the emissions during both normal and disturbed soil conditions. That is, measurements were made of the key variables to quantify the suspended aerosols at the actual contamination sites, but different scenarios for habitation, site management, and site cleanup were included.

The most notable locations of these site-investigations were the Marshall Islands (Bikini, Enewetak, and Rongelap), Nevada Test Site (GMX, Little Feller, Palanquin, and Plutonium Valley), Tonopah (Nevada--site of Roller Coaster), Savannah River Lab (South Carolina--H-Area site), Johnston Island (cleanup of rocket-impact site), Chernobyl (Ukraine--grass field and sandy beach sites near Nuclear Power Plant Unit 4), and Palomares (Spain--site of aircraft accident).

This discussion will review the variables quantified, methods developed, general results, uncertainty of estimations, and recommendations for future research that are a result of our experience in these field studies.

## DEFINITIONS OF VARIABLES

We investigated the following subjects at actual contaminated sites:

- **concentration in air:** contaminant per unit volume of air,  $C$
- **concentration in soil:** contaminant per unit mass of soil,  $A_s$
- **deposition:** contaminant per unit area of surface soil,  $D$
- **emission rate:** flux of contaminant; mass, or Bq, per unit area per unit time,  $Q$
- **resuspension rate:** fraction of surface contaminant resuspended in unit time,  $R$
- **resuspension factor:** air concentration per unit of deposition,  $S_f$
- **activity coefficient:** radioactivity per unit mass of particles,  $A$
- **enhancement factor:** activity coefficient of aerosols per unit soil activity,  $E_f$
- **particle size:** median aerodynamic diameter, MAD, and distribution function
- **turbulent diffusion:** micrometeorological transfer of contaminants to air,  $K$

From the basic definitions, some more are derived:

$$Q = K (dC/dz) \quad (1)$$

$$R = Q/D \quad (2)$$

$$S_f = C/D \quad (3)$$

$$E_f = A/S \quad (4)$$

Here in Eq.1, the quantity  $(dC/dz)$  is the vertical gradient of  $C$ , the concentration in air. Eq.1 is known as the **meteorological flux gradient** method. It has been in long term use by us (Anspaugh *et al.* 1975) and has its basis defined in the DOE Handbook of Atmospheric Diffusion (Hanna *et al.* 1982). A modification of of this method has also been used in the Soviet Union (Garger *et al.* 1990).

Some further definitions and algebraic manipulations are necessary:

$$A_s = A_0 \exp (-\alpha z) \quad (5)$$

which is the empirical observation that the soil concentration will decrease exponentially with depth, and  $\alpha$  is known as the **inverse relaxation depth**. But,  $A_0$  is only an extrapolated value of the soil concentration at the soil surface. In actuality "we believe that the soil surface is well-mixed in the first few centimeters, because the geophysical and biological factors are very active in the span of a few years" (Shinn, 1992, in discussion with conference attendees). Because of the vigorous mixing processes, let us define  $S$ , the **soil surface concentration** over the shallow soil layer of depth  $z_1$ , that is the variable most often measured, and then define  $A_0$  in terms of  $S$ :

$$A_0 = S \exp (\alpha z_1/2) \quad (6)$$

Usually  $z_1$  is taken to be 2 to 2.5 cm in depth, because of the extensive use of that depth increment in soil sampling protocols.

The enhancement factor,  $E_f$ , is then defined as the ratio of suspended aerosol activity,  $A$ , to the surface soil concentration,  $S$ , in Eq.4, and is dimensionless. The deposition,  $D$ , is determined by summation of measured  $A_s(z)$  in soil profiles, which is the same as integration of Eq.5 from the soil surface to infinity, and multiplying by  $\rho$ , the soil bulk density:

$$D = \rho Sh \quad (7)$$

Here  $h$  is the **characteristic depth of depositon**, obtained by integration:

$$h = (1/\alpha) \exp (\alpha z_1 / 2) \quad (8)$$

It should be noted that  $h$  and  $\rho$  can be easily estimated for the typical values of  $z_1$  (2 to 2.5 cm), and values of relaxation depth commonly found,  $0.2 < \alpha < 2.0 \text{ cm}^{-1}$ , except in sandy or disturbed soil. That is:

$$h = 5 \text{ cm } \pm 1.5 \text{ cm} \quad (9)$$

and usually:

$$\rho = 1.5 \text{ Mg / m}^3 \pm 0.2 \text{ Mg / m}^3 \quad (10)$$

## **METHODS**

We will briefly review our methodologies for field studies of resuspension here. The purpose, scope, responsibilities, and procedures are documented in a "Study Plan", (Shinn 1993), as required by U.S. Department of Energy Orders when human health assessments are made. The latest Study Plan, dated May 1, 1993, contains or refers to fifteen different Detailed Procedures, and is about 50 pages in length. Since these details are beyond the scope of the paper, the study plan and procedures will be only outlined here.

But the methods discussed here are the same, except for minor modifications, as those previously published by us (Anspaugh *et al.* 1975, Shinn *et al.* 1980, 1982, 1986, 1989, Shinn and Homan 1985, and Shinn 1992).

### **Air Sampling**

Four types of air sampling are required--(1) high-volume air samplers (66 to 100 m<sup>3</sup>/h) for concentration in air, (2) cascade impactors (34 m<sup>3</sup>/h) for the aerosol size distribution, (3) vertical sampling array (10 m<sup>3</sup>/h) used for concentrations in air simultaneously at four heights above ground and, (4) electronic suspended-mass monitors and optical particle counters for determination of minute by minute variations in dust concentrations and particle mass size-distributions during the experiment. Additional air sampling is optional by our ultra-high volume air samplers (250 to 1000 m<sup>3</sup>/h), and by personal air-samplers.

Aerosols are collected on either glass-fiber or cellulose fiber media, that are weighed and handled carefully to obtain high precision after the filter media are equilibrated to constant humidity and temperature. Air samplers are calibrated with critical orifice and venturi units, and flow rates are monitored in the field.

Our suspended mass monitors are a combination of electronic microbalance and light-scattering (laser and broad-band) devices. The microbalance and light-scattering mass monitors are recorded continuously by our automatic weather station. The laser and optical particle counters are fitted with isokinetic vanes, and multichannel analyzers, and are usually used infrequently.

## **Soil Sampling**

Two types of *in situ* gamma spectroscopy are used to obtain area-averaged values of concentrations in surface soil,  $S$ . The Field Instrument for Detection of Low Energy Radiation (FIDLER) is calibrated for specific isotopes and used to map their horizontal distributions; for example  $^{241}\text{Am}$  is used to map the  $^{239-240}\text{Pu}$  distribution. Also, portable *in situ* gamma spectroscopy units are used for mapping details of other isotopes by means of either high-purity germanium diodes or sodium-iodide detectors that allow detailed peak identification and quantification. Gamma spectra recorded in the field are processed by commercial software, with verification against our custom-designed software.

Soil sampling by means of soil cores in 2.5-cm increments is done primarily to obtain estimates of the soil-profile characteristics,  $A_s(z)$ , and to obtain the isotopic ratios (for example, the ratio  $^{239-240}\text{Pu} / ^{241}\text{Am}$ ) using our precision laboratory techniques for alpha, beta, and gamma spectroscopy.

## **Analytical Methods**

Laboratory gamma spectroscopy of soil and aerosol samples is done in deep-well detectors of various types, where gamma counting can be done for long enough times to quantify natural and background isotopes. For the transuranic isotopes, however, special chemical methods are required. On filter media and soil samples, the samples are dissolved first, followed by chemistry using tracers to determine recovery efficiency. In all of these techniques, our laboratories use a rigorous quality assurance program with interlaboratory comparisons, and quality control by duplicate samples, blanks, and spiked samples. For example, our alpha chemistry precision is better than 0.005 Bq/sample for  $^{239-240}\text{Pu}$  and  $^{241}\text{Am}$ .

## **Meteorological Methods**

Measurement of the turbulent diffusion characteristics and other relevant meteorological variables (for example, wind direction) is done with special weather stations set up in the field simultaneously with the air sampling. The method of Schuepp *et al.* (1990), as recommended by Leclerc and Thurtell (1990), is used to determine the representativeness of the heights of the air samplers and meteorological sensors. At the typical height of 1.13 m in a short grass field, a sampler is 90% representative of an upwind range (fetch) of 145 m, (Shinn and Gouveia 1992).

Two types of weather stations are set up; one is simply a portable set of sensitive cup anemometers and recorder that is used to measure the boundary layer parameters of zero-plane displacement height and the surface roughness length. Another, an automatic micrometeorological station, is set up to continuously record wind speed, wind direction, suspended mass loading, and special variables that allow quality control of the estimate of turbulent diffusion. The special variables are two methods of estimation of the diffusivity of heat through the energy balance technique and the eddy correlation technique, converted to diffusivity by means of accurately-measured air-temperature gradients. This result is applied to estimate the radioactive aerosol emission rates when combined with the measurement of the vertical gradient of radioactivity obtained by the vertical array air sampler. The estimate is stability

corrected by means of parameterization to the Monin-Obukov scaling factor with coefficients derived from the actual field data (Hanna *et al.* 1982).

### Special Measurements

A portable wind tunnel is used on many experiments to control the surface shearing stress on the eroding soil and to investigate the flux,  $Q$ , during a steady-state. The tunnel consists of a single duct, 10-m long and 75-cm in cross-section. The wind tunnel has no bottom, and can be sealed to the surface without disturbing the eroding soil. Diagnostic data about the effects of surface cover, surface disturbance, and rate of recovery after disturbance are obtained. Wind speeds are controllable from 2 m/s to 12 m/s.

Our ultra-high volume air samplers are used when it is necessary to obtain air concentrations over a very short time period. For example the flow rates are high enough to obtain background air concentrations of  $^{239-240}\text{Pu}$  in 24 hours.

### GENERALIZED RESULTS

There are a number of results from our experiences which we will offer as generalizations. The reader should consult the references for details of a site-specific nature.

#### Estimates of air concentration of contaminants.

We have observed the resuspension factor at many sites of aged deposits, and the results tend to confirm that the resuspension factor,  $S_f$ , approaches the value recommended by Anspaugh *et al.* (1975) of  $10^{-9} \text{ m}^{-1}$ . The uncertainty of this value tends to vary with the site, but Kercher (1993) found a large coefficient of variation between 115 trials at the GMX site in Nevada. The most common approach to estimation of the concentration in air,  $C$ , is to assume a constant value of  $S_f$ :

$$C = S_f \cdot D \quad (11)$$

Thus the uncertainty in estimation of  $C$  is directly dependent on the large uncertainty in the resuspension factor, providing uncertainty of  $D$  is small.

We suggest that a more mechanistic approach would be to estimate air concentration from the mass loading,  $M$  (usual units,  $\mu\text{g}/\text{m}^3$ ), the soil activity,  $A_s$ , and the enhancement factor,  $E_f$ :

$$C = E_f \cdot A_s \cdot M \quad (12)$$

Here  $M$  is the measured, or modeled, mass loading by total suspended particulates. This approach allows one to consider the physics of the suspended particulate process separately from the problem of how much activity is present. Where there are no measurements of the enhancement factor, use the following:

$$E_f = 0.7 \quad \text{in the case of an } \textit{undisturbed surface soil} \quad (13a)$$

$$E_f = 4 \quad \text{in the case where } \textit{soil is recently disturbed} \quad (13b)$$

$$E_f = 0.01 \quad \text{near to a } \textit{site of nuclear detonation} \quad (13c)$$

Examples of enhancement factors were listed by Shinn (1992) for bare, cultivated fields, nuclear event sites, non-nuclear explosively released "accident" sites, man-made disturbances (bull-dozer blading, soil raking, vacuum-cleaning), and natural disturbances (springtime thaw, soil-drying, wildfire). While there may be an influence of disturbance on particle size, we do not find it to be significant, and the size-distributions are usually similar before, during, and after disturbance. The differences expressed in Eqs. 13 are apparently related to the distribution of contamination among soil particle sizes, and the bonding of contaminant to soil, among other things.

We have an example (Shinn 1992) where biweekly values of  $M$  in the Frenchman Flat desert reached a peak in the dry summer months, but the airborne activity,  $A$ , reached a peak in the springtime. This out-of-phase relationship produced maximum potential exposure in the springtime, not when the dusty period prevailed. It illustrates that two separate processes should be considered.

**We do not understand the processes where the activity in the suspended aerosols should greatly increase just by physical changes such as disturbances.** Most of the physical constants of the soil must have stayed the same (bulk density, surface roughness, soil texture, and soil moisture). As a result we resort to empirical tabulation of the enhancement factors. On the other hand, **there is some hope of predicting or monitoring the mass loading,  $M$ .**

#### Estimates of resuspension parameters

As examples of variation of resuspension parameters, we present data in Tables 1 and 2 comparing four Pu-contaminated sites at the Nevada Test Site, each with low density shrub cover (Shinn *et al.* 1986). Here the friction velocity,  $u^*$ , is a measure of the difference in turbulent diffusivity between the sites. It can be seen that **"non-nuclear" contamination sites (simulated accident events, called "safety shots") with similar resuspension factors have very different resuspension rates.** Also, nuclear sites have very low enhancement factors due to the fact that much of the soil contamination is contained in miniature glass beads (formed in the nuclear blast) that are not resuspendable. By assuming that the measured resuspension rates are characteristic of all future time, we calculated the resuspension half times, just for further illustration of the importance of resuspension rates in prioritizing the remediation measures. **The differences in particle sizes, MAD, between sites, are probably not all that important as far as health assessments are concerned,** because the particle distributions are broadly dispersed-- approximately lognormal with geometric standard deviation greater than 4. This means that the integrated pulmonary deposition will not be much different, although the extrathoracic deposition will be slightly different.

#### Uncertainty Estimates

Our experience in estimation of uncertainty can be summarized briefly. Let us use the coefficient of variation, CV, (defined as the ratio of the standard deviation to the mean determined from empirical data) as a figure of merit. Note that in many sets of data the variables are distributed lognormally, so that the estimate of CV depends on a lognormal transform.

Table 1. The  $^{239+240}\text{Pu}$  aerosol characteristics at four locations of the Nevada Test Site, each with low-density shrub cover.

Location	S surface soil Pu (Bq/g)	C aerosol (Bq/m <sup>3</sup> )	E <sub>f</sub> Enhancement factor	M Mass loading (μg/m <sup>3</sup> )	MAD <sup>a</sup> (μm)
Non-Nuclear					
GMX	310	$4.6 \times 10^{-3}$	0.87	17	5.7
Pu Valley	23	$9.6 \times 10^{-4}$	1.04	41	5.5
Nuclear					
L. Feller 2	25	$8.1 \times 10^{-6}$	0.02	22	2.7
Palanquin	666	$8.9 \times 10^{-6}$	0.002	7	2.5

a MAD = Activity median aerodynamic diameter.

Table 2. The  $^{239+240}\text{Pu}$  aerosol emissions at four locations of the Nevada Test Site, each with low-density shrub cover.

Location	S <sub>f</sub> Resuspension factor (m <sup>-1</sup> )	u* (m/s)	Q Pu flux (Bq/m <sup>2</sup> s)	R Resuspension rate (s <sup>-1</sup> )	Resuspension half time (y)
Non-Nuclear					
GMX	$2.0 \times 10^{-10}$	0.20	$1.9 \times 10^{-5}$	$7.9 \times 10^{-13}$	27,000
Pu Valley	$6.1 \times 10^{-10}$	0.20	$6.2 \times 10^{-5}$	$3.8 \times 10^{-11}$	560
Nuclear					
L. Feller 2	$4.3 \times 10^{-12}$	0.23	$6.1 \times 10^{-6}$	$6.0 \times 10^{-13}$	36,000
Palanquin	$1.8 \times 10^{-13}$	0.45	$3.3 \times 10^{-7}$	$6.7 \times 10^{-15}$	3,300,000

When the resuspension factor method Eq.11, is used, the CV for deposition, D, is about the same as the CV for S, the surface soil contamination, and is usually much smaller than the CV for S<sub>f</sub>. The CV for S<sub>f</sub>, the resuspension factor, at GMX site in Nevada was more than 10 (Kercher, 1993), but in Chernobyl examples it had a value of about 2 (Garland and Pattenden, 1991). In the case of GMX, perhaps site factors had an influence on the variation, and this was an extreme case. The CV for S (and also, D) is more than 2 when soil sampling is used, and about 1.4 when *in situ* gamma spectroscopy is used. The reason for the (unanticipated) large CV for S is that when properly stratified, the soil samples have contamination values that are lognormally distributed with geometric standard deviations greater than 3 and often greater than 5 (Shinn and Gouveia 1991). That *in situ* gamma spectroscopy has surprisingly large CV is due to the dependence of calibration on inverse relaxation depth, α, and on the CV of α, combined with the CV of the isotopic ratios such as  $^{239-240}\text{Pu} / ^{241}\text{Am}$ . Thus, in our experience, the CV for predicted concentrations in air using the resuspension factor method is not less than 2 and often as great as 10.

When the enhancement factor method is used, Eq.12, the CV of  $E_f$  is typically less than 0.5 when it is measured at a site, but of course when it is estimated by Eqs.13 it may be larger. Usually, the CV for M is less than 0.5 when it is measured, but it can be as high as 10 in the case of disturbed soil (Loshchilov *et al.* 1992). The CV for S was discussed above. Combining these uncertainties, our experience is that **using the enhancement factor method, the CV of predicted air concentration is usually less than 2**, except in the case where disturbance is present. Of course, traffic and cleanup would be examples of disturbance that would be expected to occur during mitigation enterprises.

Uncertainties in estimation of the aerosol emission rate, Q, and the resuspension rate, R, are dependent on the CVs of C and D (discussed above). The turbulent diffusion coefficient has a CV less than 0.5 assuming that wind records are collected, and the vertical gradient measurements have an uncertainty proportional to C (by a coefficient which has a CV less than 0.5). Thus the CV of Q is dominated by CV of C, and the CV of R is dominated by the CV of C and D. **The predicted aerosol emission rate usually has a CV not less than 2**, disregarding the uncertainty of wind estimates, and the predicted resuspension rate usually has a CV not less than 3.

### **RECOMMENDATIONS FOR FUTURE RESEARCH**

Future research should center around two themes. First, **we recommend the documentation of and the reduction of uncertainty** in observed parameters. That is, while reduction of uncertainty may be difficult, at least the knowledge of the source and magnitude of variation may be useful in stochastic simulations and risk assessments. This recommendation can be carried out while conducting the human health assessments needed for management, and mitigation of contamination sites. Secondly, **we recommend that mechanistic studies be conducted on the processes** which bind contaminant particles to soil, and on the ability to predict the three-dimensional time-dependent distribution of aerosol mass loading during natural and man-made conditions of disturbance (such as cleanup methodologies, traffic, etc). This recommendation cannot be carried out without some fundamental research support, independent of the health assessment studies.

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