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Jospeh H. Shinn, PhD

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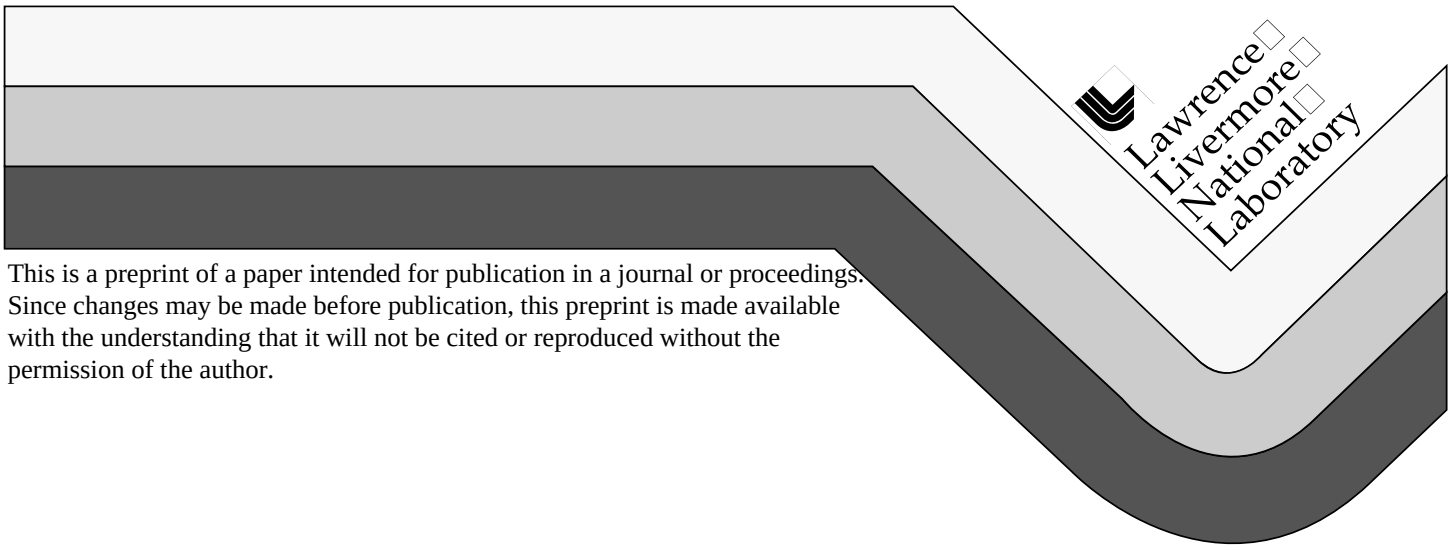

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Joseph H. Shinn, PhD
Health and Ecological Assessment Division
Lawrence Livermore National Laboratory
925-422-6806 or shinn1@LLNL.gov

INTRODUCTION

This paper addresses the issue of inhalation exposure immediately afterward and for a long time following a nuclear accident. For the cases where either a nuclear weapon burns or explodes prior to nuclear fission, or at locations close to a nuclear reactor accident containing fission products, a major concern is the inhalation of aerosolized plutonium (Pu) particles producing alpha-radiation. We have conducted field studies of Pu-contaminated real and simulated accident sites at Bikini, Johnston Atoll, Tonopah (Nevada), Palomares (Spain), Chernobyl, and Maralinga (Australia).

DISCUSSION

It has long been recognized that the most significant pathway for human exposure to Pu is the inhalation of aerosolized-Pu attached to soil, and methods have been developed to estimate human exposure by this pathway[1]. From the perspective of health risk assessment there are two physical processes that should be predicted: the first-order process that produces the "respirable" Pu-concentration in air, and the second-order process that causes a Pu flux into the air (rate of Pu-aerosol emissions) for subsequent redistribution. The term "respirable" is defined here as particles having less than 10 μm aerodynamic diameter. Resuspended Pu particles normally have a peak in the size-distribution at 2 to 5 μm, and are approximately log-normally distributed with a geometric standard deviation from 3 to 5. The Pu is found as plutonium oxide aggregated in soil particles.

Observations of normalized radionuclide concentrations in air following an accident show a remarkable agreeement, since at all sites the concentrations decrease by 5 orders of magnitude in the first 20 to 30 days. This should guide decisions on reentry into an accident site. Our hypothesis is that (1) this occurs because fallout particles initially adhere to any available surface, but are transferred to sites of greater and greater adhesion with time and (2) the process is much more rapid than migration into the soil. But eventually, fallout radionuclides find their way to the soil surface. Our time dependent empirical model is too conservative and overpredicts Pu-concentrations in air[1]. There are several time-dependent models in current literature, and none seem to be completely accurate even though they are physically-based. More work needs to be done in this area.

A simple model for predicting the Pu concentration in air is the *resuspension factor* approach. In this concept, the Pu concentration in air is integrated over a period of at least several days to eliminate the variations

due to wind and weather conditions, and these Pu concentrations are normalized by dividing the observed concentrations in air (C, Bq/m^3) by the local soil-Pu inventory from deposition (D, Bq/m^2). This method has proven useful around the world because once S_f is estimated, then the concentration can be predicted from the deposition, D, that is, $C = S_f D$. The resuspension factor values, S_f, tend to a long-term limit between $10^{-10}\,m^{-1}$ and $10^{-9}\,m^{-1}$.

Enhancement Factor and Effects of Disturbance

This "steady-state" can be interrupted, however, by disturbances such as construction, traffic, etc. Another model, the *mass loading* approach, tries to deal with this problem by predicting the Pu-aerosol activity (A, Bq/g) and the total suspended particulate mass loading (M, g/m^3). The activity, A, would be predicted from an enhancement factor, E_f, and the average surface soil activity to a depth of 0.05 m, S_o (Bq/g), that is, $A = E_f S_o$. In this model, the concentration can then be predicted by the product combination, $C = E_f S_o M$. Both S_o and M are easily measured. But both E_f and M can be expected to increase during disturbances, and in undisturbed soil, both have seasonal variations. That E_f would increase with disturbance indicates that the Pu bindings with soil aggregates are somewhat fragile. In some cases M is predictable from dust emission factor models for various types of construction and agricultural activity. In studies performed over a wide number of sites, Shinn[2] found that values of E_f were usually less than unity, typically 0.7, for the non-fissioning types of accidents and at large distances from fission events (Bikini, Palomares, Tonopah, Maralinga). For disturbances such as traffic, bulldozer blading, wildfire, and freezing-thawing cycles, Shinn reported that E_f values temporarily increased to between 2.5 and 6.5. In the case of a nuclear fission event at ground level, much of the soil Pu within a kilometer of the "ground zero" is found in small glass beads that are too large to be resuspended. So for nuclear fission accidents, these E_f values were found to be about 0.01 and the resuspension factors, S_f , decrease to a lower long-term limit between $10^{-13}\,m^{-1}$ and $10^{-11}\,m^{-1}$.

Particle Emission Rates

The second-order problem, predicting the Pu-aerosol emission rates, is determined by solving the flux equation $F = K\,(dC/dz)$, where K is conventionally measured as the turbulent diffusivity for sensible heat, and the vertical gradient dC/dz is measured from vertically-spaced air samplers. We simplified this even further by the approximation $dC/dz = p\,C/z$ where p is the power-law parameter determined as the constant slope from the log C versus log z measurements[1].

The parameter p is a measure of the surface conditions and for suspended particulate mass loading has typical values of –0.2 with a range between –0.05 and –0.6. The negative sign indicates that suspended mass is decreasing with height in the air above the soil. The values of p vary through the season and depend upon the degree of surface cover. The turbulent diffusivity K can be easily measured and varies directly with wind speed and height above

ground. To determine the resuspension rate, R, the flux F (Bq/m^2 sec) is divided by the local soil Pu-inventory from deposition (D, Bq/m^2), that is R = F/D. This gives the fraction of the contamination being resuspended per second. Typical values[3] for R are $10^{-11}\,s^{-1}$ to $10^{-12}\,s^{-1}$. But in some cases soils have a higher R, i.e. more erodible, sandy soil or disturbed soils that have R greater than $3 \times 10^{-11}\,s^{-1}$; then local redistribution of Pu is a problem[4].

Considerations in Environmental Cleanup

Environmental cleanup decisions for Pu should be based on the potential risk to human health. Since plutonium oxide does not easily pass into the systems circulation from the GI tract because of its low solubility, is not transferred into food chains, and does not produce a significant external dose (barely detectable gamma radiation), the decisions will be largely based on inhalation exposure estimates. It is important to average soil measurements over an area, because at typical inhalation heights, the trajectories of particles come from an upwind range characterized[5] as 90% within a distance of 150 m.

A first consideration is the removal of fragments and radioactive debris. This requires locating and removing fragments that are visible, and a vacuum cleaner method worked well with a 60% removal efficiency for each pass[4]. Locating Pu fragments must be done with a special instrument (high purity germanium crystal) optimized for detecting a weak gamma emission from a daughter radionuclide, ^{241}Am. At Johnston Atoll, a mining technique of sifting soil on a moving belt was used successfully to remove the fragments but did not reduce the activity in the inhalation size range[6]. At Maralinga, residual Pu fragments were mapped after the contaminated surface soil was scraped off, and the fragments were either removed by hand or by vacuum cleaning.

Consideration of potential land use, and cultural practices for habitation has led to different cleanup criteria[7] as appropriate. Experiences in cleanup at Enewetak Atoll, Maralinga, and Tonopah led to slightly different cleanup criteria and averaging areas for integrating sampled soil Pu; Table 1. Cleanup criteria for Pu in these cases were between 1.5 and 15 Bq/g (40 to 400 pCi/g). For comparison purposes, the calculated Pu-concentrations in air using the upper limit steady state resuspension factor of $10^{-9}\,m^{-1}$ could be estimated from the contamination depth of 0.05 m and a soil bulk density of $1500\ kg\ m^{-3}$ to be between 112.5 and 1125 $\mu Bq/m^3$.

Table 1. Risk-Based Plutonium Cleanup Criteria for Cleaned Sites.

SITE	POTENTIAL LAND USE	CLEANUP CRITERIA $^{239,240}Pu$
Enewetak Atoll	residential island	1.5 Bq/g over 0.25 Ha
	agricultural island	3.0 Bq/g over 0.5 Ha
	food gathering	6.0 Bq/g over 0.5 Ha
Maralinga, Australia	hunting and gathering	9-15 Bq/g ($0.7\text{-}2.2$ Bq/g ^{241}Am)
Tonopah, Nevada	cattle grazing	7.4 Bq/g over 1 Ha

CONCLUSIONS

Post-accident inhalation exposure is an important determinant entering decisions about re-entry and possible risk management schemes, because of the alpha contamination from Pu. Our experience in field studies at Pu-contaminated sites provides some insights about risk estimation and risk management. Since Pu-concentrations decrease by 5 orders of magnitude in the first 20-30 days, it would be advisable to postpone re-entry until after that time. Furthermore, because of the importance of inhalation exposure and the possible local effects on the enhancement factor (ratio of aerosol activity, Bq/g, to surface soil activity, Bq/g) it would be advisable to monitor the air concentration and the total suspended particulate mass loading and to predict future resuspension factors from these observations rather than to estimate them from empirical means. We expect nevertheless that when steady state is reached, the resuspension factor will have a long-term limit between 10^{-10} m^{-1} and 10^{-9} m^{-1}. Typical risk-based cleanup criteria will be between 1.5 and 15 Bq/g in soil and this will result in Pu-concentrations in air less than the range 112.5 to 1125 μBq/m^3. The soils will have a resuspension rate of 10^{-11} s^{-1} to 10^{-12} s^{-1} unless they are highly erodible and then redistribution of Pu is a problem if the rate exceeds 3×10^{-11} s^{-1}.

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