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Enhancement Factors for Resuspended Aerosol Radioactivity: Effects of Topsoil Disturbance

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ABSTRACT

The enhancement factor for airborne radionuclides resuspended by wind is defined as the ratio of the activity density (Bq g^{-1}) in the aerosol to the activity density in the underlying surface of contaminated soil. Enhancement factors are useful for assessment of worst-case exposure scenarios and transport conditions, and are one of the criteria for setting environmental standards for radioactivity in soil. This paper presents results of experimental studies where resuspension of ^{239}Pu was measured when air concentrations were equilibrated to the soil surface. Enhancement factors were observed for several types of man-made disturbances (bulldozer-blading, soil raking, vacuum-cleaning) and natural disturbances (springtime thaw, soil-drying, wildfire). For some cases, enhancement factors are compared over a range of geographical locations (Bikini Atoll, California, Nevada, and South Carolina). The particle-size distributions of aerosol activity are compared to particle-size distributions of the underlying soil.

1. INTRODUCTION

It is sometimes important to estimate the radioactivity in aerosols that have been resuspended from a contaminated soil surface. For example, it is common in environmental risk assessments to estimate an approximate or worst-case aerosol concentration, C, by multiplying an observed suspended dust particulate concentration, M (g m^{-3}), by the activity of a particular radionuclide, A (Bq g^{-1}), in the soil. This estimate may be improved with further multiplication by an enhancement factor, EF, to include disturbance effects on the soil:

$$C = EF \times M \times A \qquad (1)$$

In the case where one can be assured that the airborne concentration of radionuclides is in equilibrium with the underlying soil surface, it is possible to observe the EF directly. Strictly speaking, the EF is the ratio of the activity density of the suspended aerosols (Bq g^{-1}) to the activity density of the underlying soil, but only when the condition is met that there are no "imported" aerosols in the air sample. This problem was pointed out in data analyses by Sehmel (1983). One case when the condition is assured is the micrometeorological requirement for the uniformity of horizontal upwind fetch. That is, the extent of upwind soil-contamination level, surface roughness, and vegetation cover is uniform at a distance sufficiently far that there are no appreciable horizontal gradients and wind-erosion flux (resuspension) depends only on vertical gradients of particle concentration. Because of the depth of the surface boundary layer, which increases approximately linearly with distance from an upwind discontinuity, it is necessary to make vertical-gradient measurements within two meters above the surface to meet the horizontal upwind fetch requirements of 50 m to 100 m of uniform surface contamination. In practical terms, such uniform zones of contamination are rarely found, but they do provide unique sites of opportunity for studying resuspension and the airborne activity densities (Shinn, Homan, and Gay, 1982).

Another case when there is unlikely to be "imported" radioactivity in the airborne particle activity density is when a wind tunnel is used during measurements, and the horizontal scale is compressed over which the uniform upwind fetch requirement must be met. Bottomless, portable wind tunnels have been employed to conduct resuspension studies over natural surfaces (Garland, 1979; Shinn, Homan, and Gay, 1982). In order to use this method to get activity densities, there must be essentially no radioactivity in the background air measurements at the wind-tunnel intake, and the representative airborne-activity densities must be determined within the shallow surface boundary layer of the wind tunnel (less than the half-height of the wind tunnel at the outflow end). It has been shown (Shinn and Homan, 1987) that the vertical profiles in the wind tunnel are similar to those observed in the field over the same natural soil-surface conditions, provided a different scale factor is used for vertical similarity, for example, 9-cm depth of scale in the wind tunnel compared with 100 cm in the field for the same applied surface-shearing stress.

A third case when the effects of any "imported" airborne radioactivity might be minimal is when a regional equilibrium is obtained between the resuspended aerosols and the underlying soil surface. Such a condition is difficult to predict, and only when long-term monitoring shows that inflow and outflow concentrations are the same can it reasonably be assumed that an equilibrium activity density for a region has been observed.

In this paper are presented the results of many observations during the three cases discussed above; the activity density was determined during equilibrium conditions and, in turn, the EF was computed. Of special interest are the observations made following disturbance of the surface, so that a range of EF values was obtained. It should be pointed out that no attempt is made to adjust EF for the difference in particle size of the soil host compared to the particle size of the suspended aerosol. (In fact, we know something about these particle-size distributions which we will discuss later.) In the absence of our ability to understand and quantify the myriad factors of geology, soil structure, soil texture, soil moisture, soil cohesion, organic matter, ridge roughness, and so on, which influence the availability of soil particles to erosion by the wind, we are forced to use EF values obtained by empirical observation as a means of estimation of the possible ranges in exposure concentrations (C, in Equation 1).

2. EXPERIMENTAL METHODS

2.1 Soil Concentrations.

Soil samples were taken by the standard Nevada Applied Ecology Group method: from the surface, a 12.7-cm-diameter ring template was used to extract a core of 2.5-cm depth. Samples were oven dried, weighed, and sieved to pass a 1.7-mm opening. Soils were homogenized by mixing in a roller-mill and analyzed by either one of two methods: the Pu-daughter isotope, ^{241}Am, was determined by a Ge solid-state detector with pulse-height spectrometer to measure 60-keV gamma emissions, or both the ^{241}Am and the ^{239}Pu were determined by special alpha chemistry methods to a precision of 3 mBq. In the latter case, the ^{239}Pu values contain a negligible amount of ^{240}Pu also. The ratio of Pu/Am was obtained by alpha spectrometry in order to make field determination of approximate ^{239}Pu concentrations using *in situ* gamma spectroscopy with portable systems; these had a circular view of the surface over several meters in radius and a precision for ^{241}Am of about 20 mBq g^{-1}. The portable systems were used to map the soil contamination and to assure horizontal uniformity of the soil concentration (Shinn et al., 1989).

2.2 Aerosol Concentrations

High-volume (HV) samplers were used to obtain air samples for periods from 3 to 14 days on either glass-fiber or cellulose-fiber filters at a nominal flow rate of 100 m^3 h^{-1}. The filters were totally dissolved and analyzed to obtain both ^{239}Pu and ^{241}Am by the same special alpha chemistry as was used for the soil samples. In addition to HV, cascade impactors were used to obtain size distributions of resuspended particles and for redundancy to simultaneous HV samples. The cascade impactors were 5-stage jet-plate type with fiber filters, which practically eliminate any bounce-off problems on intermediate stages. The cascade impactors were operated at about 33 m^3 h^{-1}, and the filters were analyzed in the same way as the HV filters. HV and cascade filters were weighed on a precision balance before and after exposure after the filters were equilbrated to at a standard temperature and low humidity. The HV and cascade impactor measurements were made in standard HV enclosures at the 1.2-m height and were located at sites that met the requirements discussed in the Introduction.

3. RESULTS

3.1 Regional and Seasonal Effects

The seasonal trend of particulate concentration (M, in Equation 1), the specific activity of ^{239}Pu in air, and the air concentration of ^{239}Pu (C, in Equation 1), were monitored for one year in Area 5 at the Nevada Test Site, beginning in February of 1981. Two stations were located about 5 km apart along the upwind-downwind axis of the prevalent wind in Frenchman Basin: the upwind station was to the southwest about 200 m east of Well 5B, and the downwind station was on the northeast end of Frenchman Playa at the 62-m meteorological tower. Prevalent winds (daytime) are from the southwest every month of the year, while nighttime winds are weak, with divergent upslope flows off the Basin floor after sunrise and convergent downslope flows into the Basin beginning at sunset. The two stations were in remarkably good agreement for values of measured quantities averaged over two-week periods, which gives assurance that they were equilibrium values representative of the region. The important observation was that the values of M and activity were seasonally out of phase. The dust concentration, M, reached a peak in the mid-summer dry period as one might expect, but the ^{239}Pu activity density reached a peak in the springtime with the increasing values correlated to increasing numbers of frost-free days in springtime (days with observed minimum air temperature above freezing). The data are shown on Figure 1 and a smooth curve has been fitted by Hanning smoothing. The spring-thaw effect produced an increase in the air activity density by a factor of about 6.5, and assuming that the soil concentrations of ^{239}Pu radioactivity were constant, we would estimate that the EF, which is normally near unity, was increased to a value of 6.5 by the spring thaw.

3.2 Enhancement Factors in Non-Disturbed Cases

The EF values were determined for desert-pavement covered soils at Nevada Test Site and for stabilized, cultivated fields at Bikini Island, South Carolina, and California. The values for those sites were typically unity or slightly less. At the Nevada Test Site, experiments were conducted in the 1960's in which ^{239}Pu was deposited on the soil by non-nuclear explosions. Measurements of EF at Area 5, GMX, gave a mean value of 0.87 and at Area 11, Site D, gave a mean value of 1.04. These

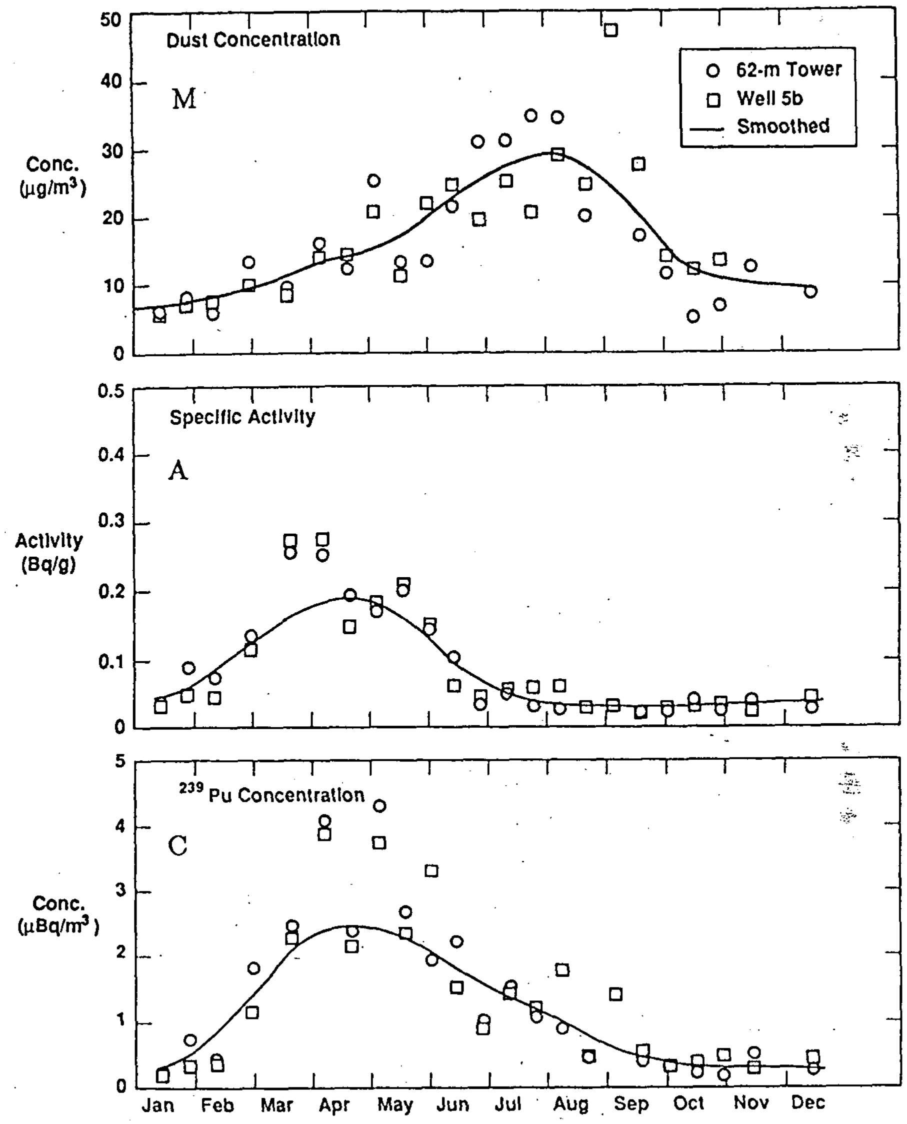

FIGURE 1. Seasonal course of M, A, and C during 1981 in Area 5 of the Nevada Test Site. (Here A is the activity in the air.)

values should be compared with EF values close to nuclear event sites at the Nevada Test Site, where the ^{239}Pu is contained in small glass beads mixed into the soil and the EF values are two to three orders of magnitude less than unity and less of an inhalation exposure threat. For example, at Area 18, Little Fellow II site, the EF was 0.024 and at Area 20, Palanquin site the EF was 0.002. See Table 1.

For cultivated fields that had been cleared of vegetation, the topsoil stirred by farm implements, and the bare soil allowed to settle for one week, the EF values were all less than unity. At Bikini Island, which had been contaminated by fallout ^{239}Pu at some distance upwind, the EF under these conditions was 0.80. At Savannah River Laboratory, South Carolina, two fields that had been contaminated by ^{239}Pu from a nearby smokestack had EF values of 0.21 and 0.49. For a small garden plot at Lawrence Livermore National Laboratory that had been contaminated by ^{239}Pu in sewage sludge, the wind tunnel was used to obtain an EF value of 0.73. The EF values for cultivated fields are compared to other cases, such as at the Nevada Test Site, in Table 1.

TABLE 1. Typical Enhancement Factors for ^{239}Pu in Aerosols Resuspended from Soils at Nevada Test Site and from Bare Fields.

	Site	EF	Contaminating Event
1.	Nevada Test Site*		
	Area 5, Site D	0.87	Safety shot.
	Area 11, Site D	1.04	Safety shot.
	Area 18, Little Fellow II	0.024	Nuclear shot.
	Area 20, Palanquin	0.002	Nuclear shot.
2.	Bare Cultivated Fields		
	Bikini	0.80	Nuclear fallout.
	South Carolina, Field 1	0.21	Processing facility.
	South Carolina, Field 2	0.49	Processing facility.
	California	0.73	Sewage sludge.

*NTS locations have desert pavement and 5-20% native plant cover.

3.3 Enhancement Factors in Disturbed Cases

Several conditions of disturbance to the topsoil yielded empirical observations of effects on EF values. In no case was the disturbance so great as to effect more than an order of magnitude increase in EF, this usually occurred after drastic stirring of the surface, e.g., bulldozing, tilling, raking-off of desert pavement. At Bikini Island, the first few days after bulldozing and clearing the vegetation from a field, the EF value was increased by a factor of 3.9. At the Nevada Test Site, Area 11, a cleanup that involved tilling and removal of 60% of the surface soil by a single pass of a vacuum cleaner resulted in an increase in EF by a factor of 3.6. At the Nevada Test Site, Area 20, the effect of a natural wildfire that removed all the grass cover was to increase the EF by a factor of 3.5. In the case of studies to evaluate the efficiency of desert pavements, the increased wind erosion caused by hand-raking to remove the pebble cover resulted in an increase in EF by a factor of 2.2. And, when the soil surface on a field in South Carolina dried out after a soaking rain, the EF remained about the same. The factors of increase of EF are compared in Table 2 as well as the factor caused by springtime thaw. It should be emphasized that the values given in Table 2 were not the EF values but the factors of increase of EF values, so that only when the undisturbed EF was unity would the disturbance EF have the values given in Table 2.

It is interesting to contemplate how difficult it is on the basis of first principles to predict what might have happened during these disturbances. Most of the physical constants of the soil must have stayed the same (bulk density, surface roughness, soil texture, and even soil moisture), but there were more contaminant particles available for wind erosion nevertheless. In these cases, empirical observations may have to suffice for environmental risk assessments.

TABLE 2. Observed Factors of Increase in Aerosol Activity of ^{239}Pu(Bq/g) due to Soil Disturbance.

Type of Disturbance	Relative Increase	Site
Soil thawing in springtime.	6.5	NTS, Area 05
Bulldozer blading.	3.9	Bikini
After 60% vacuum cleanup.	3.6	NTS, Area 11
Wildfire, removing grasses.	3.5	NTS, Area 20
Raked off desert pavement.	2.2	NTS, Area 18
Soil dried and eroded for 2 weeks.	0.8	So. Carolina

3.4 Changes in the Airborne Particle Size Distributions

During many of the experimental determinations of air-activity density, the particle-size distributions were measured for aerodynamic diameters between 0.3 and 10 µm. These data show almost universally that the ^{239}Pu activity density was approximately log-normally distributed across particle diameters and have median aerodynamic diameters in the range of 2 to 6 µm. The geometric standard deviations of airborne activity density had a range between 2 and 3.6 (nondimensional units), while quite often the suspended soil dust had a much broader distribution with geometric standard deviations in excess of 6. This points out that the resuspended ^{239}Pu was bound to a particularly narrow range of the soil particles available for wind erosion. In all of the cases observed, however, disturbance had a minor effect on the particle-size distributions. Only a slight decrease in the median aerodynamic diameter was noted, and that may not have been statistically significant. It remains to be shown that particles are freed by the disturbances.

Lee, Tamura, and Essington (1987) have found that soils at Nevada Test Site that have been contaminated by ^{239}Pu, have only 1% of the total ^{239}Pu associated with soil particles below 5 µm in diameter. Yet that was the particle size which was active in resuspension, and which was in the respirable size range. It may be only coincidence that the undisturbed EF values tend to unity, because that practically negligible fraction of the soil accounts for all of the resuspension and the amplification of that fraction must therefore be enormous.

4. CONCLUSIONS

The complexities of all the geophysical processes affecting the wind-erosion process make it necessary to rely on the empirical estimation of the activity density of resuspended radioactivity. To this end, the observed EF values serve the useful purpose of allowing an estimate of air exposures for an unusual range of natural and disturbed soil conditions. For the purposes of most environmental risk assessments, the EF values should be entirely satisfactory considering the possible alternatives of unviable theoretical prediction and of site-specific monitoring.

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DISCUSSION

J. A. GARLAND. The results presented here suggest that the EF approach can be applied with uncertainties that are probably smaller than those in other approaches. It necessitates the measurement of airborne mass concentration, and the value of the method may depend on whether the mass concentration varies very much with surface condition, so I would like to ask the author whether the disturbances to the surface resulted in much change in airborne mass concentration.

J. H. SHINN. We have observed increases in airborne mass concentrations in the respirable size range in every case when the surface of the soil was disturbed. The degree of this effect of course is unpredictable and must be determined empirically, but the increase in EF is independent of that effect. We are not suggesting that airborne concentration should be estimated by EF alone, without the accompanying measurement of airborne mass.

J. A. GARLAND. Like the resuspension factor method, this approach may be of limited value in areas of inhomogeneous contamination. Has the author any experience of the problems that arise in these circumstances?

J. H. SHINN. A major point of this paper is that you have to go to great pains to determine the airborne activity density and the EF when the conditions of horizontal homogeneity are met. Having so determined the EF, however, doesn't mean that it shouldn't apply more than locally. That is, if the meteorological conditions (surface roughness, etc) remain horizontally uniform, and the soil surface properties except for contamination level remain uniform, then the EF and also the resuspension factor could be applied with the aid of a diffusion model to estimate downwind concentrations over a large area.

R. JAENICKE. You claim that the greatest mass of soil is in particles larger than 5 micrometers. For resuspension calculations it is probably appropriate to restrict the observation to those soil particles which might be transported over large distances. Such a limit would be in the 3-5 micrometer range.

J. H. SHINN. The observation of aerosol particles is restricted to the size range less that about 10 micrometers by virtue of the types of filter-impactors, optical particle counters, and other methods we use. And of course, these are the particles which are transported long-range and have a small settling velocity compared to the turbulent transport velocities. But it is useful nevertheless to examine how the radioactivity is distributed in the soil. Having said that I must point out that the latter is seldom done because it is very difficult to determine the distribution in the soil with any detail below the particle size of 5 micrometers.

R. JAENICKE. Your wind tunnel is open at the bottom. Does the wind tunnel not produce a very special climate?

J. H. SHINN. The wind tunnel is sealed to the soil on the sides, and is intended only to produce a steady surface shearing stress on the soil. It is made portable so that it does not need to stay too long in one place, and the soil and ground cover conditions could be quite natural. There may be other limitations of the tunnel, but it is a very useful tool in a supplementary sense.

S. E. SCHWARTZ. You discussed airborne particle size distributions for radioactivity and dust that were lognormal. Are these data or schematic?

J. H. SHINN. We have lots of data on airborne particle size over the range from 0.3 to 10 micrometers from both cascade impactors and optical particle counters. At these remote sites the distributions are approximately lognormal with the medians and geometric standard deviations I presented. Strictly speaking, however, the lognormal approximation applies only to that range investigated.

O. I. VOZZHENNIKOV. It seems important to emphasize the advantages of EF to the commonly used resuspension factor.

J. H. SHINN. The EF offers more information about the surface condition than the resuspension factor in the case of an aged deposit. But it is empirical nevertheless, and we admit that the soil processes are too complex to quantify when we resort to such an approach. We use the EF when seeking to put bounds on the air concentration estimates, for example.

O. I. VOZZHENNIKOV. The soil activity, A, in your tests depends on particles on the surface which resuspend. The conditions on the surface can be different from those at a certain depth. Is this limitation of the method inherent in undisturbed soil?

J. H. SHINN. It is commonly known that there is an approximate exponential decrease of activity with depth between 2 and 10 cm in the soil. On the other hand, this function probably does not apply above 2 cm. 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. For example, rain drop impact, shrinking-swelling cycles, wind saltation, root growth, freezing-thawing, etc, are processes which mix the surface. Our studies show also that the statistical variability of surface soil samples is quite large even when normalized to the total deposition. This means that the problem is difficult to study but perhaps more studies are needed.

O. I. VOZZHENNIKOV. In this respect it would be interesting to discuss a hypothesis of a thin sublayer (less than the roughness length) in which concentration of radioactive dust is in equilibrium with pollution density of the soil surface. What is your view of the problem.

J. H. SHINN. In principle, I agree that this hypothesis has merit. But there are a few factors which make the hypothesis difficult to test except for some very ideal cases. First, all natural surfaces of interest have a zero-plane displacement of the atmospheric surface boundary layer. Even in bare soil, the ridge roughness is as large as the aerodynamic surface roughness length. Perhaps this is the layer which is in equilibrium. Second, I cannot think of a method to test the hypothesis that is experimentally nonintrusive. Our methods of particle collection are bulky and flow distorting when placed so near the ground. So I remain interested in the hypothesis, but I haven't resolved the question of how to test it.