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PLUTONIUM-AEROSOL EMISSION RATES AND POTENTIAL INHALATION EXPOSURE DURING CLEANUP AND TREATMENT TESTS AT AREA 11, NEVADA TEST SITE

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ABSTRACT

A Cleanup and Treatment (CAT) test was conducted in 1981 at Area 11, Nevada Test Site. Its purpose was to evaluate the effectiveness of using a large truck-mounted vacuum cleaner (similar to those used to clean paved streets) for cleaning radiological contamination from the surface of desert soils. We found that four passes with the vehicle removed 97% of the alpha contamination and reduced resuspension by 99.3 to 99.7%. Potential exposure to cleanup workers was slight when compared to natural background exposure.

INTRODUCTION

A Cleanup and Treatment (CAT) test was performed in Area 11 (Plutonium Valley) of the Nevada Test Site during the summer of 1981. The test area had been contaminated with $^{239-240}Pu$ and ^{241}Am by a series of experiments that dispersed plutonium oxide about 25 years earlier. The CAT test was sponsored by the U.S. Department of Energy and carried out by Reynolds Electrical and Engineering Co., Inc. (REECo). The purpose of the CAT test was to investigate the utility of a large truck-mounted vacuum unit (VAC-ALL) for soil collection as an alternative to conventional earthmoving techniques.[1]

During the CAT test we performed experiments to:

1. Determine the plutonium-aerosol fluxes (emission rates) before and after cleanup,
2. Estimate the extent of potential inhalation exposure for observers and equipment operators had they been unable to utilize respiratory protection,
3. Support the REECo Environmental Sciences Department with additional aerosol monitoring, and
4. Document meteorological conditions encountered at the site during the CAT test.

The CAT test was carried out on two strips of land, nominally 100 m by 10 m each, located on northwest-southeast axes, and separated by 30 m.[1]

Each strip was divided into ten plots (10 m X 10 m) and surveyed for soil contamination. Before cleanup, the southwest strip had an average of 553 pCi/g of $^{239-240}Pu$ and 97 pCi/g of ^{241}Am. The northeast strip had an average of 265 pCi/g of $^{239-240}Pu$ and 47 pCi/g of ^{241}Am. The variability between plots within a strip was small; relative standard deviations of ^{241}Am, determined by in situ gamma spectroscopy, were about 14%. The first cleanup pass with the VAC-ALL removed approximately 60% of the contamination, mostly in the top 5 cm. Resurveys located the remaining hot spots of contaminated soil; three additional passes with the VAC-ALL resulted in removal of about 97% of the alpha contamination.

Before, during, and after the cleanup, we determined the plutonium aerosol concentrations and particle size characteristics. We also made the measurements necessary to calculate the emission rates of plutonium as a result of wind erosion (resuspension) and to determine the pulmonary deposition (deep lung) to a hypothesized unprotected worker.

METHODS

The methods used were basically the same as those employed in recent resuspension studies both at the Nevada Test Site[2] and the Savannah River Project.[3] Three types of air samplers were used:

High-volume (HV) samplers. The HV samplers used glass-fiber filters, and flow rates were nominally 100 m^3/h (60 cfm) monitored at a pressure tap on the blower. Three HV samplers were used; two were housed in standard HV sampler enclosures, and the third was mounted (filter facing down) 0.38 m above the ground at the center of a floorless wind tunnel.

Cascade impactors. Two cascade impactors (Model 65-000, Anderson 2000 Inc.) were used for aerosol size determinations. These were jet-plate type instruments mounted on blowers set to operate at a flow rate of 33 m^3/h (20 cfm) in a standard HV sampler enclosure.

Personal dosimeter (PD) air samplers. Five PD air samplers were used, one as an area monitor and four in workplaces. The PD (Model S, Monitaire, Mine Safety Appliance Co.) consisted of filter holders mounted on standard nylon cyclones and driven by battery-operated portable pumps, which produced a flow rate of 0.1 m^3/h (0.06 cfm). The PD used 37-mm Teflon fiber filters.

One HV and one cascade impactor were each placed 2 m downwind of the edge of the southwest cleanup strip. One HV and one cascade impactor were also placed far upwind as the background monitors. The four PDs were assigned to vehicle drivers and to vacuum-hose operators on the VAC-ALL.

Environmental Analysis Laboratory, Richmond, California (formerly LFE Corporation) used isotope dilution and alpha spectrometry[4] to determine the concentration of $^{239-240}Pu$. With a precision of 0.08 pCi for $^{239-240}Pu$, the relative errors of the HV air sampler were less than 4%.

Meteorological measurements consisted of both continuous monitoring and special diagnostic observations. Windspeed, wind direction, air temperature, air temperature difference (ΔT) between 3.20 m and 0.25 m, and soil temperature were continuously measured and summarized for 8-min periods. In addition, vertical-profile measurements of wind speed below 4 m were made to determine aerodynamic properties. Vertical-profile measurements of speed and dust conditions were also made in the lower 30 cm of the wind tunnel. The speed in the wind tunnel was measured with an electronic air-flow meter, and the relative dust concentration profiles in the wind tunnel were measured with a light-scattering particle counter.

The flux (emission rate) of dust and plutonium aerosol was computed by our formerly described method[2,3]:

$$F = pk\,\bar{x}\,(\overline{u_*^3}/\overline{u_*^2}), \qquad (1)$$

where x is the dust or plutonium concentration measured at a convenient height above the ground, u_* is the diabatically adjusted friction velocity for a given 8-min period, k is the Karman constant, and p is the slope of the log x, log height plot. The overbar denotes time averaging over a period of several days.

A floorless wind tunnel made it possible to control the wind speed associated with resuspension and determine the characteristic fluxes during a steady state. The tunnel was constructed to develop a wind-speed profile similar to that observed in the field; the speed was set at a constant 11 m/s (25 mph), and of course the turbulence scales were much smaller in the tunnel than in the open field. This open-circuit wind tunnel was constructed of a single duct, open at either end, with a coaxial fan at the outlet end, discharging 22,000 m^3/h (13,000 cfm). The tunnel was 10 m in length and

0.75 m X 0.75 m in cross section. The first 2 m of the inlet end consisted of an entrance cone, a honeycomb section, and a boundary-layer conditioning section. The lower 20 cm of the tunnel satisfactorily reproduced the constant-stress zone of the atmospheric boundary layer. At the center of the tunnel near the end of the test section, a HV sampler was placed in an inverted position to obtain plutonium and dust aerosol samples. The discharge of the HV sampler was directed outside the tunnel through a plastic exhaust pipe.

RESULTS AND DISCUSSION

DESCRIPTION OF METEOROLOGICAL CONDITIONS

Meteorological records were summarized over two one-month periods during the cleanup phase (Phase II) of the CAT test: June 10 to July 9, 1981, and July 10 to August 12, 1981 (see Figures 1a and 1b). Both periods were typical of the hot, dry summer season. The wind directions were usually from the south during the day. (Our pretest planning assumption that they would be from the southwest[1] was incorrect.) Wind speeds during the day increased regularly from about 1 m/s at 8 a.m. to a maximum of about 4.5 m/s (10 mph) near noon, and remained steady each afternoon until 6 p.m. The winds at night were usually from the southwest at 1 m/s. Average wind speed was 2 m/s at this site.

Maximum daytime air temperatures were typically 30 to 35°C at the 3.2-m height and 37 to 39°C at the 0.25-m height. During midday the soil temperatures at 2-cm depth were typically 14 to 17°C warmer than the 3.2-m air temperature. Hence, 50°C (122°F) midday soil temperatures were common.

AEROSOL CONCENTRATIONS

The upwind background dust and plutonium concentrations were determined from four consecutive HV determinations. The values obtained about 2-km "upwind" to the northwest were 42 (±12) $\mu g/m^3$ for dust and 267 (±103) aCi/m^3 of $^{239,240}Pu$. (Values in parentheses are standard deviations.) This compares with a background of 19 $\mu g/m^3$ for dust and 40 aCi/m^3 of $^{239,240}Pu$ observed at NS201 in Area 18, Nevada Test Site, during the previous summer.[2]

We conducted experiments for plutonium aerosol analyses in the contaminated area at four distinct times:

- o Prior to cleanup.
- o During cleanup; a 3-wk period of the CAT test Phase II. However, the aerosol samples would not represent a specific dust-plume created by traffic or the VAC-ALL.
- o Immediately after cleanup.
- o After the soil had stabilized for two weeks. (A water spray used during cleanup played a large role in stabilizing the surface.)

The aerosol measurements were made at the 1.2-m height and 2 m downwind of the plots, rather than directly over the plots. The downwind location was necessary for unhindered CAT test operations. The measurements can be assumed to be close enough to the plots to represent the erosion and resuspension from the plots and not from upwind or downwind sources.

The data show (Table 1) that, before cleanup in the undisturbed conditions, the dust concentration was at background levels but the plutonium concentration was nearly 100 times background. During cleanup the plutonium concentration increased to nearly 400 times background, but gradually decreased, so that with the surface stabilized after 60% cleanup the plutonium aerosol levels were about 50 times background. This latter value observed was about half the value observed before the CAT test.

Table 1. Dust and plutonium aerosol concentrations at the 1.2-m height, 2 m downwind of cleanup plot.

Period	Dust ($\mu g/m^3$)	Plutonium (aCi/m^3)	Enhancement factor
Before CAT (normal)	41	25900	1.03
During cleanup	88	103,000	2.00
After 60% cleanup[a]	39	40,500	3.71
After stabilized[a]	41	13,800	1.20
Background[b]	42	267	--

[a] Following one vehicle pass of the VAC-ALL, nominally 60% efficient.

[b] Background determined about 2 km upwind.

In contrast to plutonium aerosol concentrations during cleanup, the dust concentration increased less dramatically (to twice background) and rapidly returned to background. The enhancement factor is defined as the ratio of aerosol plutonium activity (pCi/g) to soil plutonium activity.[2,3] This factor is normally less than one, but at this site it was essentially equal to one prior to cleanup (Table 1). It did increase during CAT but not by much more than the enhancement factors found at other disturbed sites. For example, the highest enhancement factor observed (3.71) compares with a value of 3.1 observed during tilling of a bare field on the island of Bikini.[5] Also, the enhancement factor increased by a ratio of 2.2 in the wind tunnel at Nuclear Site 201[2] when the surface was disturbed by raking away the desert pavement, which compares with a ratio of 2.5 observed in the wind tunnel after raking the stabilized surface following the CAT test.

Before cleanup and with the surface as undisturbed as possible, the wind tunnel produced considerably more resuspension than observed outside the tunnel. Before the CAT test, with the wind tunnel placed carefully over the undisturbed surface, the plutonium aerosol concentrations were 8.8 times greater than outside (850 times greater than upwind background) and the dust concentrations were 4.5 times greater than outside and upwind background. This result can be explained by the increased windspeed in the wind tunnel (11 m/s compared to 2.2 m/s outside). Apparently the soil is wind erodible in its undisturbed condition. After cleanup when the soil was stabilized, however, the plutonium aerosol concentration in the wind tunnel was reduced more than the reduced soil contamination would predict; this observation confirms that the soil is more erodible in its undisturbed state.

AEROSOL SIZE DISTRIBUTION AND LUNG RETENTION FACTOR

The aerosol size distribution showed that both the mass and activity median aerodynamic diameters were about 5.5 µm. The plutonium aerosol concentration size distribution (aCi/m^3) was approximately log-normal with a geometric standard deviation (GSD) of 8.5; this is a broadly polydispersed aerosol distribution.

We determined the respirable-size fraction by multiplying the plutonium aerosol size distribution by the pulmonary (deep lung) retention function recommended by the ICRP Task Group on Lung Dynamics[6] and integrating from

0.05 µm to 50 µm. It was found that the lung deposition so calculated was about 14% of the total aerosol concentration.

The median aerodynamic diameter of the specific plutonium activity per unit mass (pCi/g), however, is 1.2 µm and the GSD is 3.6; this distribution is nearly monodispersed. Thus the polydispersed concentration distribution (aCi/m^3) is a result of integrating the latter monodispersed activity distribution (pCi/g) into a broadly polydispersed dust aerosol mass concentration ($\mu g/m^3$). This dispersing effect has been observed elsewhere at both the Nevada Test Site[2] and the Savannah River Project.[3]

PLUTONIUM AEROSOL FLUXES AND RESUSPENSION RATES

The parameters of the flux, p, u_*, and $\bar{x}$, Equation (1), were determined as follows: p was found from three separate determinations in the wind tunnel, u_* was found from observations of hourly averaged wind speed (diabatically corrected), and $\bar{x}$ was found from observation of the mean plutonium aerosol concentration, Table 1.

We assumed that the value of p in the wind tunnel was representative of values outside the tunnel, as verified in other studies.[2,3] The mean value of p was -0.273 with a standard deviation of 0.077. (Negative sign indicates decreasing concentration with height.) There was no significant difference in p values found before cleanup versus those found after cleanup. The mean value of p found was comparable to that found at Savannah River Project,[3] but lower by about a factor of three than the value of p found at Area 18, Nevada Test Site,[2] where the plutonium resuspension rate was unusually low.

The value of u_* was computed from a value of roughness length (z_0) measured to be 3.06 cm ± 0.45 (zero plane displacement, 9.4 cm) over the undisturbed shrubs, which were 35 to 40 cm tall with about 15% ground cover. However, the cleanup had a secondary effect on resuspension by reducing the value of u_*. The value of z_0 was reduced by shrub removal to a value in the range 10^{-2} to 10^{-4} cm, variable because of the microterrain and residual root fragments embedded in the surface. This reduction in roughness would reduce the resuspension rate by a factor of 5 to 10 just by reducing u_*.

The mean value of the ratio $\overline{u_*^3}/\overline{u_*^2}$ was 0.4 m/s while the value of u_* was 0.207 m/s prior to cleanup. The diabatic corrections to u_* were performed with the aid of temperature gradient measurements (T_*, averaged over each hour, and by using conventional micrometeorological techniques for this approach.[2]

The plutonium aerosol emission rate for the natural, undisturbed surface was found to be 145 pCi/(m^2 day). Because the $^{239-240}Pu$ soil contamination (90% in the top 5 cm) near this site was 4.26×10^7 pCi/m^2, this flux results in a resuspension rate of 3.39×10^{-6}/day (3.92×10^{-11}/s) and a resuspension factor of 6.1×10^{-10}/m. If this rate was representative of the long-term normal resuspension, and if the resuspension rate were assumed to be constant with time, the half-time for resuspending the $^{239-240}Pu$ would be about 560 years. This rate, however, was 60 times greater than the rate in Area 18, Nevada Test Site,[2] and 9 times greater than the rate at Savannah River Project.[3] The high rate at Area 11 can be attributed to erodibility and saltation effects.

It should be noted that the resuspension factor is defined as the plutonium aerosol concentration (pCi/m^3) divided by the soil contamination on an area basis (pCi/m^2), and the resuspension rate (fraction per day) is defined as the resuspension flux (plutonium aerosol flux, pCi/(m^2 day)) divided by the soil contamination on an area basis (pCi/m^2). Thus it is possible to reduce both the soil concentration and the plutonium aerosol concentration by like fractions, and the result would be no change in either the resuspension factor or the resuspension rate. On the other hand, two other parameters (p, u_*) could change both the flux and the rate. We found that p remained constant before and after cleanup.

The effect of cleanup was to reduce the resuspension rate, but not only by removal of soil. That is, the fractional reduction in aerosol concentration closely followed the fractional reduction in soil concentration so that their ratio would not be affected, while a 5- to 10-fold reduction in the resuspension rate was caused by the drop in u_* from a decrease in surface roughness. On the other hand, the combined effect of decreasing the plutonium aerosol concentration and the roughness was to reduce the plutonium aerosol flux from 145 pCi/(m^2 day) to 8-15 pCi/(m^2 day) when the soil was stabilized following just one VAC-ALL Pass (60% efficient). Because subsequent VAC-ALL vehicle passes did not affect the surface roughness, but would continue to decrease the plutonium-aerosol concentrations, we calculated that three additional passes with the VAC-ALL would result in a final plutonium-aerosol flux of 0.5-1.0 pCi/(m^2 day). Therefore, while the CAT test was 97% efficient in reducing soil plutonium, it was 99.3% to 99.7% efficient in reducing resuspension.

POTENTIAL INHALATION EXPOSURES

Personal dosimeters (PD) placed in the work environment showed the potential inhalation exposure for equipment operators, had they been unable to utilize respiratory protection. Two occupations were particularly well monitored: drivers inside the semi-closed cabs of the VAC-ALL and the radiological survey (IMP) vehicle, and operators outside the VAC-ALL vehicle manipulating the vacuum cleaning nozzle.

These data show that the drivers had a potentially greater exposure than the operators outside (see Table 2). This is because the cabs served as settling chambers for dust without the benefit of continuous ventilation and dilution. There were also differences between the exposure for two vacuum operators, probably because of differences in the efficiency of the dust control with a water spray. For comparison, a PD was placed on a downwind HV at the same height as the HV intake port as an area-monitor. The drivers had a potential exposure about 4 times greater than the outside area-monitor.

Table 2. Estimated plutonium aerosol levels in the work environment during Cleanup and Treatment test.[a]

Occupation	$^{239-240}Pu$ (aCi/m^3)	Ratio to area-monitor
Driver No. 1	292,000	3.6
Driver No. 2	303,000	4.3
VAC Operator No. 1	47,500	0.46
VAC Operator No. 2	167,000	2.1

[a] Estimates made by obtaining mass loading on personal dosimeters (PD), correcting for the efficiency of the PD relative to HV, multiplying by the average activity of both cleanup strips (409 pCi/g), and multiplying by an average enhancement factor of 1.5.

while the vacuum operators had a potential exposure not significantly different than the area-monitor, taking into account their variability (ratio 0.4 to 2.1, geometric average 0.92).

The average potential exposure of a driver was about 300,000 aCi/m^3 and that of an operator about 100,000 aCi/m^3, but these aerosols were only 14% respirable.

A driver exposed to ^{239}Pu aerosol without respiratory protection for 60 8-h days at 300,000 aCi/m^3, assuming a work-day breathing rate of 10 m^3/d and 14% pulmonary retention, would obtain a bone dose of 60 mrem (Class Y material) during a 50-y lifetime. A vacuum head operator exposed to ^{239}Pu aerosol without respiratory protection at 100,000 aCi/m^3, but otherwise the same as above, would obtain a bone dose of 20 mrem during a 50-y lifetime. In making these potential exposure calculations, we assumed the personnel had been unable to use any respiratory protection, and yet the doses were small compared to 7000 mrem nominally obtained from natural background over 70 years.

SUMMARY AND CONCLUSIONS

Resuspension studies performed during the Cleanup and Treatment Test in Area 11, Nevada Test Site, showed that in the undisturbed state prior to cleanup the plutonium aerosol fluxes were about 145 $pCi/(m^2$ day) on a soil with contamination of 4.26×10^7 pCi/m^2 in the top 5 cm. The resuspension factor was $6.1 \times 10^{-10}/m$, larger than others measured at the Nevada Test Site.[2,7] The resuspension rate, 3.39×10^{-6}/day, indicates that if this rate is constant with time, the half-time for resuspending the $^{239-240}Pu$ would be about 560 years.

The effect of a 97% soil cleanup was to reduce the resuspension about 99.3% to 99.7%. This added efficiency was because reducing the surface roughness with removal of the vegetation resulted in decreased turbulence. The final plutonium aerosol emission rate therefore was about 0.5 to 1.0 $pCi/(m^2$ day). Water stabilizing the soil after cleanup at this site made it less erodible than in its original state.

Personnel dosimeters in the work environment and determinations of the size distribution showed that the aerosol was about 14% respirable and that the greatest potential exposure was to truck drivers. In the semi-closed vehicle cabs the levels were typically 4 times greater than an outside area-monitor. Operators using the vacuum head outside had less potential exposure, typically the same as the outside area-monitor but with more variability. The potential exposure of a truck driver for 60 8-h days at the plutonium aerosol concentration of 300,000 aCi/m^3 amounts to a bone dose of 60 mrem during a 50-y lifetime, which is small compared to 7000 mrem obtained from natural background.

The plutonium aerosol concentrations were measured just downwind of the southwest cleanup plot in Area 11 (precleanup soil level of 553 pCi/g of $^{239-240}$Pu). The plutonium aerosol concentration prior to cleanup was about 100 times the upwind background. After 60% cleanup and with the soil stabilized the concentration was about 50 times background. The maximum aerosol concentration observed was 103,000 aCi/m^3 during cleanup; upwind background was 267 aCi/m^3.

Meteorological data show that the winds were typically from the south and increased from a speed of 1 m/s at 8 a.m. to a maximum of 4.5 m/s at noon, remaining steady each afternoon. Air temperature maximums were 37 to 39°C near the ground at midafternoon, and maximum soil temperatures were 51 to 56°C at midafternoon.

In conclusion, the resuspension data show that the cleanup was more efficient than expected in terms of reduced plutonium aerosol emissions. The suspended plutonium aerosols were not a significant hazard to workers. At the same time, it was found that the chosen cleanup site had the highest resuspension rate among those sites studied both at the Nevada Test Site and elsewhere.

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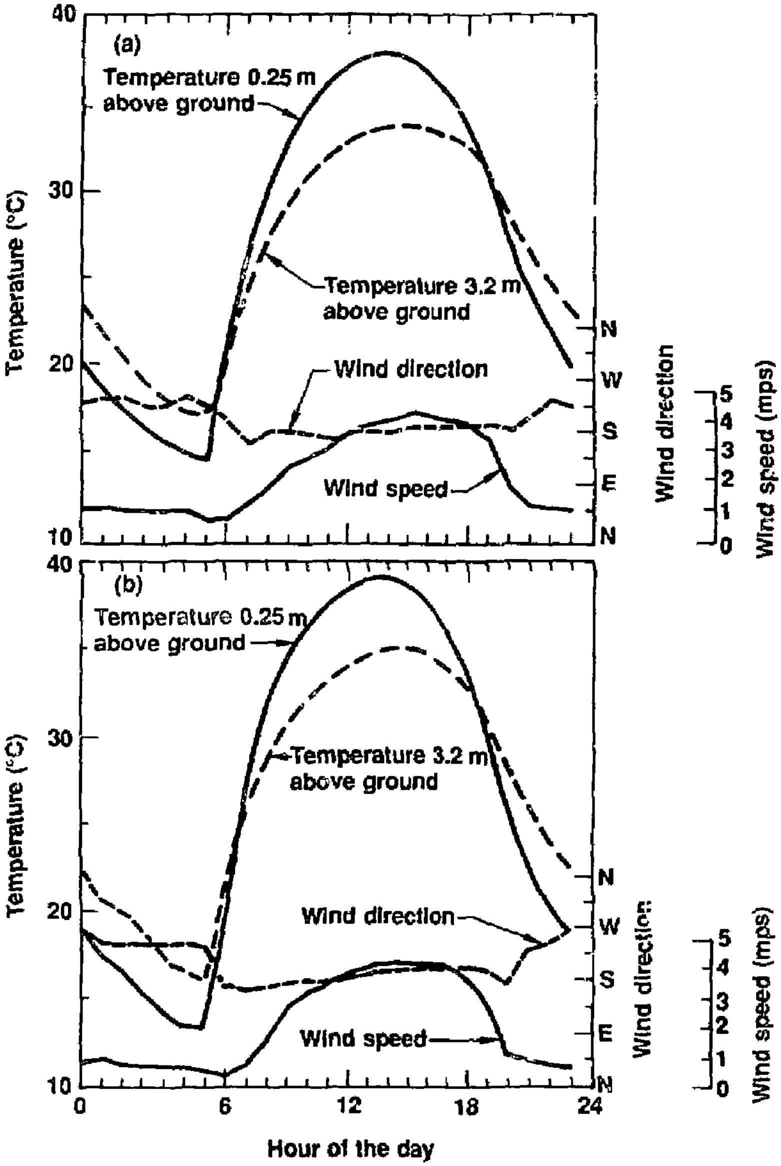

Figure 1. Meteorological records are summarized over two one-month periods (a, June 10-July 9, 1981, and b, July 10-August 12, 1981).

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