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Plutonium-Aerosol Emission Rates and Human Pulmonary Deposition Calculations for Nuclear Site 201, Nevada Test Site

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PLUTONIUM-AEROSOL EMISSION RATES AND HUMAN PULMONARY DEPOSITION CALCULATIONS FOR NUCLEAR SITE 201, NEVADA TEST SITE

ABSTRACT

We conducted a study at Nuclear Site 201 (Little Fellar II), Nevada Test Site (NTS) to determine the plutonium-aerosol fluxes from the soil to quantify (1) the extent of potential human exposure by deep-lung retention of alpha-emitting particles; (2) the source term should there be any significant, long-term transport of plutonium aerosols; and (3) the resuspension factor and rate so that, for the first time at any nuclear site, one may calculate how long it will take for wind erosion to carry away a significant amount of the contaminated soil.

High-volume air samplers and cascade impactors were used to characterize the plutonium aerosols. Meteorological flux-profile methods were used to calculate dust and plutonium aerosol emission rates. A floorless wind tunnel (10-m long) was used to examine resuspension under steady-state, high wind speed.

Plutonium-aerosol concentrations were found to be normally quite low, near global background, even when the surface-soil concentrations were 665 pCi/g. Disturbing the soil pavement surface by hand raking only doubled the dust concentration but increased the plutonium-aerosol concentration by a factor of 27. The dust and plutonium-aerosol size distributions were quite different at any given time, but the plutonium median aerodynamic diameter was typically 3.4 μm and about 13% respirable. The inhalation dose was insignificant compared to background dose even in the case of exposure to 100 8-h days at the highest concentration of 1094 aCi/m³. The plutonium fluxes changed from 3 to 33 pCi/m² · d.

The resuspension factor was two orders of magnitude lower than the other comparable sites at NTS and elsewhere, and the average resuspension rate of 5.3×10^{-8} /d was also very low, so that the half-time for resuspension by wind erosion was about 36,000 y.

INTRODUCTION

The plutonium-aerosol emissions from soils contaminated during atmospheric nuclear testing are of interest for several reasons. First, it is important to quantify the extent of human exposure from inhalation of alpha-emitting particles. For this reason we have estimated the local pulmonary (deep lung) exposure rates at Little Fellar II, Nuclear Site 201 (NS201) of the Nevada Test Site (NTS). From these estimates it is possible to

deduce some general conclusions about exposures at sites elsewhere at NTS. Second, we have applied, for the first time, a method for determining the plutonium-aerosol fluxes (Curies per unit ground area per day) at NS201 so that a source term is now available for calculating a population dose should there be any significant, long-term transport of alpha-emitting aerosols off-site. Third, the older method of characterizing plutonium-aerosol concentrations by the so-called resuspension factor has been improved by the newly determined resuspension rate so that it is now possible, also for the first time, to calculate how long it will take for wind erosion to carry away a significant amount of the contaminated soil from NS201.

Our purpose was to examine the case of plutonium-aerosol fluxes from a typical nuclear site such as Little Fellar II in a series of three experiments (RUNS) from August 21 to September 22, 1980 that compared undisturbed with slightly disturbed soil conditions. The process of resuspension produces a plutonium-aerosol emission rate that is a vertical mass flux in the classic, micrometeorological definition. The methods used rely heavily on micrometeorological measurements.

The location we chose was 2400 ft (732 m) north, and 1000 ft (305 m) west of ground zero (GZ) according to the existing grid at NS201. The area had relatively smooth terrain, uniform soil contamination, and ease of access from improved roads. The NS201 has been extensively studied by the Nevada Applied Ecology Group (NAEG), thus considerable historical data and background studies were available. Gilbert gave results on NS201 soil concentrations of the alpha-emitting isotopes $^{239-240}\text{Pu}$ and ^{241}Am .¹ The plutonium to americium ratio is about 8 on high-level soil samples (plutonium > 10 nCi/g) but the median ratio is about 11.2 on low-level samples (plutonium < 7 nCi/g). A map of NS201 is available showing soil concentrations of ^{241}Am at 200-ft grid intervals.² A map grid point (N2400, W1000) shows 63.7 pCi/g ^{241}Am , from which we estimate 713 pCi/g $^{239-240}\text{Pu}$. We sampled the soil extensively (taking 50 scoops of surface soil per each spot location of a 4 x 4 grid in a 30- x 30-m area surrounding our aerosol sampling area) and obtained 665 pCi/g $^{239-240}\text{Pu}$ and 82 pCi/g ^{241}Am , which gives a plutonium to americium ratio of 8.2. This compares with the 11.2 value from historical data. The average soil concentration of the four nearest 200-ft grid points of the historical data is 42 pCi/g ^{241}Am with a range from 11 to 70 pCi/g, which indicates a relatively uniform soil contamination area.

The ground cover by native shrubs was 25% and the average height of the shrubs was 28 cm (relative standard deviation 49%) in measurements made along four 30-m transects.

* Personal communication: Earl Sorom, REECO Environmental Division, Nevada Test Site.

METHODS

Three types of air samplers were used--(1) high-volume (HV) samplers, (2) cascade impactors, and (3) personal dosimeter (PD) air samplers. The HV samplers used 8- x 10-in. Gelman (type AE) glass-fiber filters. Flow rates were nominally 100 m³/h (60 cfm) monitored at a pressure tap on the blower. The elapsed time of operation was measured by counting pulses from a crystal-controlled clock activated by a pressure sensitive switch. Three HV samplers were used, two of which were housed in standard HV sampler enclosures and the third mounted (filter facing down) 0.38 m above the ground at the center of a floorless wind tunnel. Two cascade impactors (Model 65-000, Andersen 2000 Inc.) were used for aerosol size determinations. These are a jet-plate type, mounted on blowers set to operate at a flow of 33 m³/h (20 cfm) in a standard HV sampler enclosure. Four PD air samplers were used. 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 operating at a flow rate of 0.1 m³/h (0.06 cfm). The PD used 37-mm Teflon fiber filters (Millipore Corp., type Fluoropore FA). Four PDs were attached to stakes with their inlets at heights of 0.125, 0.25, 0.5, and 1 m to measure the vertical profile of dust concentration. All filters were weighed before and after exposure in a room with regulated temperature and humidity. The HV sampler and cascade impactor filters were weighed with an accuracy of ± 1 mg. The 37-mm PD filters were weighed on a balance with a precision of ± 1 μ g; the accuracy of the weighings was approximately ± 10 μ g.

Special chemical methods after Wessman and Leventhal were employed on the filters by Environmental Analysis Laboratory, Richmond, CA (formerly LFE Corporation) to determine the concentration of ²³⁹⁻²⁴⁰Pu using isotope dilution and alpha spectrometry with a precision of 8×10^{-14} Ci for ²³⁹⁻²⁴⁰Pu.² (Any ²⁴⁰Pu would be inseparable from ²³⁹Pu by this method.) If the background concentration of ²³⁹⁻²⁴⁰Pu is taken to be 40 aCi/m³ (1 aCi = 10^{-18} Ci), which was the lowest aerosol concentration measured over undisturbed soil, and the HV air sampler operates for 4 d at 100 m³/h, the relative precision for ²³⁹⁻²⁴⁰Pu is 21%. Under actual experimental conditions the HV air sampler errors for ²³⁹⁻²⁴⁰Pu detection were about 1.3% with a range from 0.2 to 22%. Cascade impactors have one-third the flow rate of the HV air samplers. We obtained a precision for the cascade impactors comparable to the HV air samplers by running duplicate samples and increasing the exposure time.

Meteorological measurements consisted of both continuous monitoring and special diagnostic observations. Wind speed, wind direction, temperature, and temperature difference (ΔT) were continuously measured at 2- and 12-m heights. Net radiation, ground

heat flux, and relative dust-aerosol concentrations were also monitored continuously. In addition, special measurements consisted of vertical-profile measurements of wind speed below 4 m and relative dust-aerosol concentrations and wind speed in the lower 30 cm of the wind tunnel. These special vertical-profile measurements were made over several 15-min periods during selected days.

Wind speed sensors at heights of 12 and 2 m were photo-chopper types (Met-One, Model 010) and the wind direction vanes (Met-One, Model 020) at 12 and 2 m were modified for separate sine and cosine output. Temperature differences were measured between 12 and 2 m using motor-aspirated quartz thermometers (Dymec, Model 2801A), calibrated for precision in ΔT of 0.01°C but recorded with only 0.1°C resolution. Absolute temperature sensors at 12 and 2 m were motor-aspirated, linearized thermistors (YSI, Model 44203) calibrated for 0.1°C precision. Net radiation was measured by two Fritschen-type net radiometers (Weather Measure Corp., Model R421), inflated through desiccant columns with aquarium pumps. Ground heat flux was measured by two 25-cm^2 flux plates (Hy-Cal Engineering, Model BI-7120) buried 1 m apart at the 2-cm depth. Relative dust-aerosol concentrations were monitored using an integrating nephelometer (Meteorology Research Inc., Model 1560), which measures the bulk scattering coefficient proportional to mass concentration of dust in the $1\text{-}\mu\text{m}$ size range.

The diagnostic, vertical-profile measurements of wind speed were made with a portable, sensitive, cup-anemometer system (modified from C.W. Thornthwaite Associates) operated at five height levels logarithmically spaced to 4 m above the ground (stall speed 0.09 m/s , distance constant 0.83 m). Wind tunnel vertical-profile measurements of wind speed were made with an electronic air-flow meter (Kurz Instrument Co., Model 441) and relative dust profiles were made using a light-scattering particle counter (Climet Instrument Co., Model CI-208A) with the intake directed into the flow stream by a 90° sampling tube of 0.6-cm i.d.

The flux (emission rate) of dust and plutonium aerosols is computed by the method discussed by Anspaugh, which is called the meteorological flux-gradient method³:

$$F = k u_*' z \frac{\partial \chi}{\partial z} . \quad (1)$$

The F is aerosol flux in mass (or Curies) per unit ground area per unit time, k is the Karman constant, u_*' is the friction velocity, z is the height, and χ is the concentration of aerosol at height z in mass (or Curies) per unit volume of air. The average concentration variation with height can be represented by a power law where p is the power of z and the mean concentration gradient becomes

$$\frac{\partial \chi}{\partial z} = p \frac{\chi}{z} , \quad (2)$$

where p can be represented as the slope of a $\log \chi$, $\log z$ plot. Hence, Eq. (1) simplifies with the aid of Eq. (2) to

$$F = pk u_*' \chi, \quad (3)$$

a result we have previously obtained.^{3,4} In practice, Eq. (3) is simple to use. The Karman constant $k \approx 0.4$. Dust profiles are assumed to be similar so that p is a constant for a given experiment. Here u_*' is the friction velocity corrected for diabatic influence,

$$u_*' = u_*/\phi. \quad (4)$$

The Dyer-Businger relationships^{5,6} are used to determine the diabatic influence ϕ and the integral influence ψ as a function of the Richardson number, which is calculated from the monitored wind speed and ΔT . The roughness length z_0 is determined from the special wind speed u , vertical-profile measurements by solving the log-linear, boundary layer equation for friction velocity u_* and z_0 :

$$u = \frac{u_*}{k} \left(\ln \frac{z}{z_0} - \psi \right). \quad (5)$$

Thereafter, z_0 is held constant and Eq. (5) is used to determine u_* from measurements of u at 2 m (z).

An improvement on Eq. (3) is necessary. The difficulty arises when one attempts to time average it. In practice, we measure the long-term average concentration of χ , not the average of the product $u_* \chi$. In fact, χ varies as u_*^2 , usually when saltation is not significant. (In cases where saltation is significant, the dependence is evidently even more geometric.⁴ We assume empirically that

$$\chi = \bar{\chi} \left(\frac{u_*^2}{\bar{u}_*^2} \right), \quad (6)$$

where the overbar denotes time averaging such as performed during HV air sampling for a week. Combining Eq. (6) with Eqs. (3) and (4) and then time averaging, we obtain a modified flux equation,

$$F = pk \bar{\chi} (\bar{u}_*^3 / \bar{u}_*^2). \quad (7)$$

(The prime notation has been dropped for convenience.) This equation is more appropriate when the aerosol sampling is done for a long period when wind speed is

varying dramatically. For our purposes, the ratio in parenthesis in Eq. (7) will be called resuspension velocity V :

$$V = \overline{u_*^3} / \overline{u_*^2} . \quad (8)$$

We used a floorless wind tunnel to control the resuspension velocity and investigate the characteristic fluxes during a steady state. The tunnel was constructed to develop a wind speed profile similar to that observed in the field, but the speed was set at a constant 11.5 m/s (25.7 mph), and of course, the turbulence scales would be 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 28,300 m³/h (17,000 cfm).⁷ The tunnel was 10-m long with a 0.75- x 0.75-m 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 latter contained a 1-m-long floor section artificially roughened by boards 0.1 m in height that were placed across the tunnel to trip the boundary layer and create a wind profile similar to that outside the tunnel. The 8-m-long test section was floorless so that erosion and resuspension processes could occur naturally, and care was taken to place the tunnel over the soil surface without disturbance. Measurements of wind and dust concentration profiles were made at a downwind distance of 20x the tunnel half-height. 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, an HV air 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. A Bagnold-type saltation and creep sampler was used on the tunnel floor to determine the vertical distribution of large particles bouncing or rolling along the surface at the end of the test section.⁸

RESULTS AND DISCUSSION

DESCRIPTION OF METEOROLOGICAL CONDITIONS

The experimental periods (RUNS) were characterized by winds that were relatively weak every morning but increased regularly from mid-morning to a maximum of 2 to 4 m/s (4.5 to 9 mph) every afternoon. Wind directions were always from the south between 8 a.m. and 6 p.m. (local time), then would change to light northerly winds that persisted all night because of cold-air drainage from nearby hills.

The time series of wind speed, air temperature (2 m), net radiation, and ground heat flux for each RUN are shown in Fig. 1. The normally clear weather produced regular diurnal patterns in these variables, except during RUN 2 when convective summer storms produced rain squalls frequently. (The net radiation sensor failed during RUN 2.) The RUN 1 from August 21 to September 3 had a resuspension velocity of 0.544 m/s; RUN 2 from September 4 to September 11 had a resuspension velocity of 0.426 m/s. The RUN 3 from September 18 to September 22 had a resuspension velocity of 0.44 m/s. The resuspension velocity in the wind tunnel was 0.37 m/s during all RUNS.

The z_0 was found to be 11.6 cm (standard deviation 2.3 cm) and the desert shrubs (25% ground cover) form an aerodynamically rough surface. The boundary layer was displaced upward from the ground surface by 16.3 cm (standard deviation 9.2 cm) while the vegetation height was 28 cm (standard deviation 13.7 cm). Inside the wind tunnel, which was placed over the bare soil between shrubs, the surface was considerably smoother. For example, after raking the soil in the wind tunnel, the value of z_0 was 2×10^{-4} cm.

AEROSOL CONCENTRATIONS AND SIZE DISTRIBUTIONS

Three RUNS were conducted sequentially beginning with a study of a normal, or undisturbed, surface in RUN 1, August 22 to September 3. At the beginning of RUNS 2 and 3, the surface was hand raked to loosen the soil and to remove the pebbles that formed a desert pavement. The raked area extended from 10 m north of the aerosol samplers to 50 m south of the samplers (prevalent upwind direction) and extended 40 m in the east-west direction. The RUN 2 was marked by a number of summer rainstorms that intermittently wet the surface nearly every day of the RUN from September 4 to September 11. (These rainfall occurrences showed up in the irregular net radiation and air temperature traces of Fig. 1.) The RUN 3 was conducted in a dustier environment, September 18 to September 22, when the soil stayed dry after it was raked.

Dust concentrations were $17.1 \mu\text{g}/\text{m}^3$ and plutonium concentrations ($^{239-240}\text{Pu}$) were $40.1 \text{ aCi}/\text{m}^3$ at 1.2 m above the normal surface of RUN 1. Plutonium-aerosol concentrations such as this are about global background for the western U.S.⁹ Once the surface was disturbed by raking in RUN 2, the plutonium-aerosol concentration increased to about 5.5 times normal even though the soil surface was wet and the dust concentration slightly decreased (see Table 1). Finally, with the surface both dry and raked in RUN 3, the plutonium-aerosol concentration increased to 27 times normal while the dust concentration scarcely doubled. The same effect occurred in the wind tunnel (see Table 1). These results point out that the availability of plutonium aerosols for

Figure 1. The time series of wind speed $\bar{u}$, air temperature T_0 , net radiation, and ground heat flux for three RUNS at NS201, August to September 1980.

Table 1. Dust and plutonium-aerosol concentrations at a 1.2-m height at NS201, NTS.

Experiment	Open desert		Wind tunnel	
	Dust ($\mu\text{g}/\text{m}^3$)	Plutonium (aCi/m^3)	Dust ($\mu\text{g}/\text{m}^3$)	Plutonium (aCi/m^3)
<u>RUN 1</u>				
Normal surface Aug. 21 to Sept. 3	19.1	40.1	14.6	645
<u>RUN 2</u>				
Raked and wet surface Sept. 4 to Sept. 11	15.4	220	13.6	853
<u>RUN 3</u>				
Raked and dry surface Sept. 18 to Sept. 22	30.3	1094	33.6	3326

resuspension was highly dependent on the surface condition. The plutonium-aerosol concentration increased geometrically compared to the dust concentration from RUN 2 to 3. (We will suggest an explanation for this paradox later.)

The data indicated that the median, aerosol aerodynamic diameter increased from $2.7 \mu\text{m}$ in RUN 1 to $6.8 \mu\text{m}$ in RUN 2, but decreased to $4.5 \mu\text{m}$ in RUN 3 as a result of the combination of disturbance and wetting and drying. Nevertheless, it was found that the mass, median aerodynamic diameters (MMADs) and activity, median aerodynamic diameters (AMADs) were always about equal. The mass ($\mu\text{g}/\text{m}^3$) and activity (Ci/g) aerosol sizes were distributed far differently, however, with the plutonium aerosol narrowly distributed (geometric sigma 3.1) and the dust-aerosol broadly distributed (geometric sigma 8.4). The contrast of these two distributions is shown in Fig. 2, calculated for the particle size distribution data of RUN 3, which gave a plutonium AMAD of $4 \mu\text{m}$ (geometric sigma 3.1) and a dust MMAD of $4.9 \mu\text{m}$ (geometric sigma 8.4). If the two curves of Fig. 2 are multiplied, one obtains the plutonium-aerosol concentration distribution (Ci/m^3). The effect of the broadly distributed dust concentration is to produce a plutonium concentration distribution that is only slightly more narrowly distributed than the plutonium activity.

Figure 2. Particle size distributions of dust concentration and plutonium-aerosol activity for RUN 3 at NS201, September 1980.

The geometrically increasing plutonium concentrations from RUN 2 to 3 both in the open desert and in the wind tunnel were the result of increasing aerosol activity (see Table 2). The only apparent difference between RUNS 2 and 3 was the better stability of the soil surface during RUN 2 caused by the occasional showers that wetted and crusted the recently raked soil. The plutonium activity and the dust concentration each approximately doubled so that their product, the plutonium concentration, approximately quadrupled. The increase in plutonium activity may be because of a combination of processes. For example, the broken soil crust would free more particles in the 1- μ m size range and increased saltation of larger particles would sandblast the surface, ejecting large numbers of smaller particles.

The log-normal particle distribution has a median diameter of number of particles D_N that is related to D_M , which is the AMAD, as follows:

$$D_N = D_M e^{-3(\ln \sigma_g)^2}, \quad (9)$$

Table 2. The $^{239-240}\text{Pu}$ activity in aerosols (1.2-m height) and in soil at NS201, NTS.

Experiment	Open desert (pCi/g)	Wind tunnel (pCi/g)
<u>RUN 1</u>		
Normal surface Aug. 21 to Sept. 3	2.1	44.2
<u>RUN 2</u>		
Raked and wet surface Sept. 4 to Sept. 11	14.3	62.7
<u>RUN 3</u>		
Raked and dry surface Sept. 18 to Sept. 22	36.1	99
Soil (0 to 5 cm)	665	1856

where σ_g is the geometric sigma. Between RUNS 2 and 3 the AMAD decreased from 7 to $4\text{ }\mu\text{m}$ while σ_g remained constant at 3.1. Using Eq. (9), this means that 50% of the plutonium-bearing particles were less than $0.15\text{ }\mu\text{m}$ in RUN 2 but a good deal smaller, less than $0.086\text{ }\mu\text{m}$, in RUN 3.

AEROSOL EMISSION RATES AND RESUSPENSION FLUXES

The power-law profile parameter p from Eq. (2), which can be thought of as the slope of the log-concentration versus log-height curve, was determined from the vertical-profiles taken in the wind tunnel and from the PD air samplers set on stakes at four heights. By normalizing the data to the mean concentrations measured at an arbitrary height H , the data from the open desert and the wind tunnel were compared (Fig. 3). In the open desert, H was 1 m and in the wind tunnel it was 9.5 cm. A p value of -0.77 was the best fit. Assuming that the plutonium activity remained constant with height (which could not be verified with the methods used), the same p value was used to

Figure 3. Relative vertical distributions of dust concentration comparing field data of RUN 1 (solid circles) and RUN 2 (open circles) with wind tunnel data of RUN 2 (triangles). Dashed line is overall p value of -0.77.

calculate both dust and plutonium flux for all three RUNS. In earlier studies we found p values for dust between -0.25 and -0.35.^{3,4} However, in our recent study at Savannah River Plant where a change in plutonium activity was successfully measured with height, we found a p value of -0.23 for plutonium concentration but a p value of -0.09 for dust.¹⁰ On Bikini Island we found a p value of -0.55 for dust.¹¹ The p value at NS201 was the largest yet found. This steeper gradient probably reflects the particle sizes and other factors of the soil surface condition.

The dust and plutonium-aerosol fluxes (Table 3) were computed with the above data and resuspension velocities of 0.544, 0.426, and 0.44 m/s for RUNS 1 to 3, respectively, in the open desert and a resuspension velocity of 0.37 m/s in the wind tunnel.

These values were the first estimates of plutonium-aerosol fluxes available for any site other than GMX at the NTS. We have compared these ²³⁹⁻²⁴⁰Pu fluxes after normalizing to soil concentration with other measurements such as those at GMX.

Table 3. Aerosol fluxes at 1.2-m height at NS201, NTS.

Experiment	Open desert		Wind tunnel	
	Dust ($\mu\text{g}/\text{m}^2 \cdot \text{d}$)	Plutonium ($\text{pCi}/\text{m}^2 \cdot \text{d}$)	Dust ($\mu\text{g}/\text{m}^2 \cdot \text{d}$)	Plutonium ($\text{pCi}/\text{m}^2 \cdot \text{d}$)
<u>RUN 1</u>				
Normal surface Aug. 21 to Sept. 3	-276	-1.55	-144	-6.35
<u>RUN 2</u>				
Raked and wet surface Sept. 4 to Sept. 11	-175	-6.68	-13.4	-8.4
<u>RUN 3</u>				
Raked and dry surface Sept. 18 to Sept. 22	-355	-34.3	-331	-32.8

The resuspension factor S_f is defined as the ratio of the measured aerosol concentration (Ci/m^3) to the total soil contamination level expressed on a ground-area basis (Ci/m^2). Since the aerosol concentrations probably vary as u_*^2 , the S_f would vary over many orders of magnitude if we examined it on an hourly basis. For the sake of comparison with historical data, we computed the S_f integrated over each RUN; these are presented in Table 4. A bulk density of $1.5 \text{ g}/\text{cm}^3$ and a soil depth of 5 cm was used, based on historical data at NS201.

The geometric average of Table 4 S_f s for the open desert at NS201 was $4.3 \times 10^{-12}/\text{m}$, which is two orders of magnitude lower than the $2.9 \times 10^{-10}/\text{m}$ that we found at GMX³ and the $3 \times 10^{-10}/\text{m}$ that we found at the Savannah River Plant. The resuspension rate over the open desert had a geometric average from Table 4 values of $5.3 \times 10^{-8}/\text{d}$. This value is two orders of magnitude lower than the median value of $3.1 \times 10^{-6}/\text{d}$ we reported for GMX and one order of magnitude lower than the $3.8 \times 10^{-7}/\text{d}$ we reported for Savannah River Plant.

Table 4. Resuspension factors and rates and soil $^{239-240}\text{Pu}$ concentration at NS201, NTS.

Experiment	Open desert		Wind tunnel	
	Resuspension factor S_f (m^{-1})	Resuspension rate (d^{-1})	Resuspension factor S_f (m^{-1})	Resuspension rate (d^{-1})
<u>RUN 1</u>				
Normal surface Aug. 21 to Sept. 3	8×10^{-13}	1.2×10^{-8}	4.6×10^{-12}	4.5×10^{-8}
<u>RUN 2</u>				
Raked and wet surface Sept. 4 to Sept. 11	4.4×10^{-12}	5×10^{-8}	6.1×10^{-12}	6×10^{-8}
<u>RUN 3</u>				
Raked and dry surface Sept. 18 to Sept. 22	2.2×10^{-11}	2.6×10^{-7}	2.4×10^{-11}	2.3×10^{-7}
<u>Soil $^{239-240}\text{Pu}$ concentration</u>				
Per unit mass	665 pCi/g		1856 pCi/g	
Per unit area	50 $\mu\text{Ci}/\text{m}^2$		139 $\mu\text{Ci}/\text{m}^2$	

The resuspension rates measured over NS201 open desert are supported by the values obtained in the floorless wind tunnel (see Table 4). The low S_f and resuspension rate values compared to other sites mean that the soil is inherently very stable. The half-time for $^{239-240}\text{Pu}$ resuspension at NS201 using the resuspension rate of $5.3 \times 10^{-8}/\text{d}$ is about 36,000 y, which is comparable to the half-time of radioactive decay.

Some new information on the transient nature of the initial phases of the erosion process was provided by the wind tunnel. In the first phase, after the wind tunnel was started and brought immediately to a speed of 11.5 m/s (25.7 mph), the dust concentration in the center of the tunnel decreased in proportion to $t^{1/2}$. That is, the concentration decreased by a factor of ten from the first half-minute to the end of the first hour. In the second phase, the dust concentration decreased exponentially with time. The

exponential decay rate varied with soil condition. Under normal soil conditions at NS201, the decay half-time was 3.5 h, but after hand raking the surface not only was the first-hour dust concentration 20x greater than the normal condition, but the decay half-time was 7 h. In the third phase, the erosion stabilized after about 10 h.

PULMONARY EXPOSURE RATES FROM AUGUST 21 TO SEPTEMBER 22, 1980

The particle-size distribution of plutonium-aerosol concentration was obtained by multiplying the log-normal distributions of dust concentration and aerosol activity. The highest concentration, 1094 aCi/m^3 , was for the raked and dry surface of RUN 3. We obtained a median aerodynamic diameter of $3.4 \text{ }\mu\text{m}$ and geometric standard deviation of 2.7 for plutonium aerosols by multiplying the distributions of Fig. 2. These distributions will not have a high percentage of respirable particles because a large fraction lies in the intermediate size range. We determined the pulmonary (deep lung) retention by integration with the retention function recommended by the Task Group on Lung Dynamics after Morrow¹² and found the retention to be only 13%. This estimate will be used in our exposure calculation.

Another important parameter is the plutonium activity enhancement factor EF, which we define as the ratio of $^{239-240}\text{Pu}$ activity in aerosol relative to $^{239-240}\text{Pu}$ activity in soil. The EF would be a multiplicative factor in an estimate of inhalation exposure if only the soil contamination levels were known. Taking data from Table 2, we find that the EF for RUN 1 during normal conditions is 0.003 over the open desert and 0.024 in the wind tunnel. During RUN 2 the EF value increased to 0.022 over the open desert and 0.034 in the wind tunnel. During the worst conditions, raked and dry soil of RUN 3, the EF was 0.054 over the open desert and 0.053 in the wind tunnel. An EF value from 0.02 to 0.05 is an order of magnitude lower than those found in a similar experiment at the Savannah River Plant¹⁰ and two orders of magnitude lower than those found on Bikini and Eniwetok of the Pacific Test Range.¹¹ Data of Sehmel for the Hanford Site, by comparison, shows EF values greater than 1 for soil contamination levels that are about three orders of magnitude lower than those at NS201.¹³ The EF values in the wind tunnel closely agree with the open desert except for RUN 1, the normal case. We would expect them to agree because of the similar values of resuspension velocity. Their differences in RUN 1 may be because of the difficulty in placing the tunnel without some slight disturbance to the soil surface. On the other hand, the soil surface in the wind tunnel was raked in RUNS 2 and 3 just as it was in the open desert and agreement was better.

Pulmonary deposition (worst case) can be estimated from the standard breathing rate of $20 \text{ m}^3/\text{d}$ multiplied by the highest $^{239-240}\text{Pu}$ concentration observed, 1094 aCi/m^3 , and by the fraction of aerosols respirable. The result, an inhalation of less than 3000 aCi/d , is an inconsequential amount. The dose commitment would be an insignificant amount compared to worldwide background. For example, Milham computed the dose commitment for ^{239}Pu aerosol with pulmonary retention of 17%, ^{239}Pu concentration of $49,000 \text{ aCi/m}^3$, and an exposure of 100 8-h days.¹⁴ They obtained a bone dose of 50 mrem for Class Y material (the same properties assumed for NS201) during a 70-y lifetime, which compares with a nominal 7000 mrem obtained from natural background over the 70-y period. A similar exposure period at NS201 would result in a bone dose of less than 1 mrem by comparison, because the maximum plutonium-aerosol concentration observed was 50 times less.

SUMMARY AND CONCLUSIONS

Plutonium-aerosol concentrations over the open desert at NS201 were normally quite low, near worldwide background, even when soil concentrations were about 665 pCi/g . Disturbing the soil by hand raking followed by light rain resulted in plutonium concentrations increased over background by a factor of 5.5, while the dust concentrations slightly decreased. Raking followed by dry conditions, however, resulted in plutonium-aerosol concentrations that increased over background by a factor of 27 while the dust concentrations were doubled. This result is explained by the fact that the plutonium-aerosol activity also increased geometrically in this case. The MMAD and AMAD changed with the soil condition; for example, decreasing as the distributed soil became dry. The characteristic dust-aerosol distribution was broad, but the plutonium-aerosol activity was narrow, even though the median diameters were about the same. The product of these distributions was the plutonium-aerosol concentration, which in the dry, disturbed-soil case had a median aerodynamic diameter of $3.4 \text{ }\mu\text{m}$, geometric standard deviation of 2.7, and a value of 1094 aCi/m^3 . This distribution is only 13% respirable (deep lung) and constitutes an insignificant dose when compared to natural background. For example, there is less than a 1-mrem bone dose over 70 y for an exposure of 100 8-h days, compared with the nominal 7000 mrem obtained from natural background.

The S_f for NS201 was two orders of magnitude lower than other comparable sites at NTS and elsewhere with a geometric average of $4.3 \times 10^{-12}/\text{m}$. The plutonium-aerosol fluxes were also relatively low, even though the slope of the vertical dust-concentration

gradients had a larger magnitude than any yet found. The plutonium-aerosol fluxes were normally about $3 \text{ pCi/m}^2 \cdot \text{d}$, and increased to $33 \text{ pCi/m}^2 \cdot \text{d}$ after raking the soil. The geometric average resuspension rate of $5.3 \times 10^{-8}/\text{d}$, determined by normalizing the plutonium-aerosol fluxes to the soil contamination levels, was also one to two orders of magnitude lower than values reported at NTS and elsewhere. The low resuspension rates mean that the NS201 soil is inherently very stable even when disturbed by raking away the desert pavement. The half-time for resuspension is about 36,000 y according to the average rate measured.

The experiment successfully measured $^{239-240}\text{Pu}$ aerosol fluxes for both normal and slightly disturbed soil conditions in an environment where the S_f and resuspension rates were two orders of magnitude lower than expected. We were able to determine the EF, S_f , resuspension rates, and resuspension half-times for the first time at any nuclear site of the NTS.

Based on these observations, it is recommended that the vertical gradients of plutonium activity be determined and compared with dust concentration gradients in future studies. Furthermore, there is no satisfactory explanation for the unusual stability of this soil surface, which should be investigated by soil-chemical and soil-physical studies at the soil-particle level.

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