2000 Dust Sampling

Correlating Plutonium Activity in Fugitive Dust to Plutonium Concentration in Surface Soils at Rocky Flats, Colorado

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ABSTRACT

A primary source of airborne plutonium exposure to public receptors near the U.S. Department of Energy's Rocky Flats Environmental Technology Site is the wind resuspension of plutonium-contaminated soil particles. Attempts to model dispersion of plutonium-laden particulate matter by wind have been limited by uncertainty about the correlation between the measured soil plutonium concentration and the plutonium

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concentration observed in particles eroded from the soil reservoir by wind. This study examined the relationship between plutonium activity in resuspended dust in the less than 10 micrometer aerodynamic equivalent diameter size range and total suspended particulate size range (PM10 and TSP, respectively) as a function of soil particle size, plutonium activity, and surface condition.

A portable wind tunnel was used to simulate wind erosion of slightly contaminated soil surfaces and to sample the resulting resuspended particulate matter. Vegetation covering the study area had been burned by a lightning-caused wildfire six weeks prior to the study, which increased undisturbed soil erosion potential by removing much protective vegetation and thatch from the study area. Two wind tunnel trials were performed on soil surfaces that had been uniformly disturbed by raking; two additional trials were performed on undisturbed soil surfaces. Wind tunnel particulate samples were collected on a backup filter (PM10) and in a cyclone preseparator that retained particles larger than PM10. Cyclone samples were sieved to remove particles larger than ~45 micrometer diameter, leaving particles between 10 and ~45 micrometers (approximating TSP minus the PM10). Particulate samples were analyzed for radiochemical composition and the results were compared to radiochemical analyses of co-located surface soil samples collected to a depth of less than two centimeters. Soil samples were sieved into three size fractions to estimate soil plutonium activity distribution by particle size fraction.

This study determined that plutonium specific activity in TSP resuspended from disturbed and undisturbed soil surfaces was equal to the plutonium specific activity in the shallow soil reservoir; PM10 from disturbed soil behaved similarly. In contrast, the specific activity of PM10 resuspended from undisturbed soil was significantly lower than the specific activity of the underlying soil. These results suggest that soil samples are not necessarily representative of the source material actually available for wind erosion from undisturbed soil surfaces, though they may be used to accurately estimate the specific activity in dust blown from disturbed soil surfaces.

INTRODUCTION

Risk estimation for the inhalation of plutonium-contaminated airborne soil particles is limited by current knowledge of contaminant resuspension and transport mechanisms through the environment. Attempts to model resuspension and dispersion of plutonium-laden particulate matter by wind have been limited by uncertainty about the relationship between plutonium concentrations in source areas and downwind plutonium activity resulting from airborne resuspension of dust from those source areas.[1] This study quantifies the relationship between the average plutonium concentration in the top 2 centimeters (cm) of soil and the plutonium concentrations in airborne particulate matter samples collected using a portable wind tunnel to simulate wind erosion.

History

The U.S. Department of Energy's Rocky Flats Environmental Technology Site (Site), formerly the Rocky Flats Plant, has several areas of plutonium-contaminated soil as a

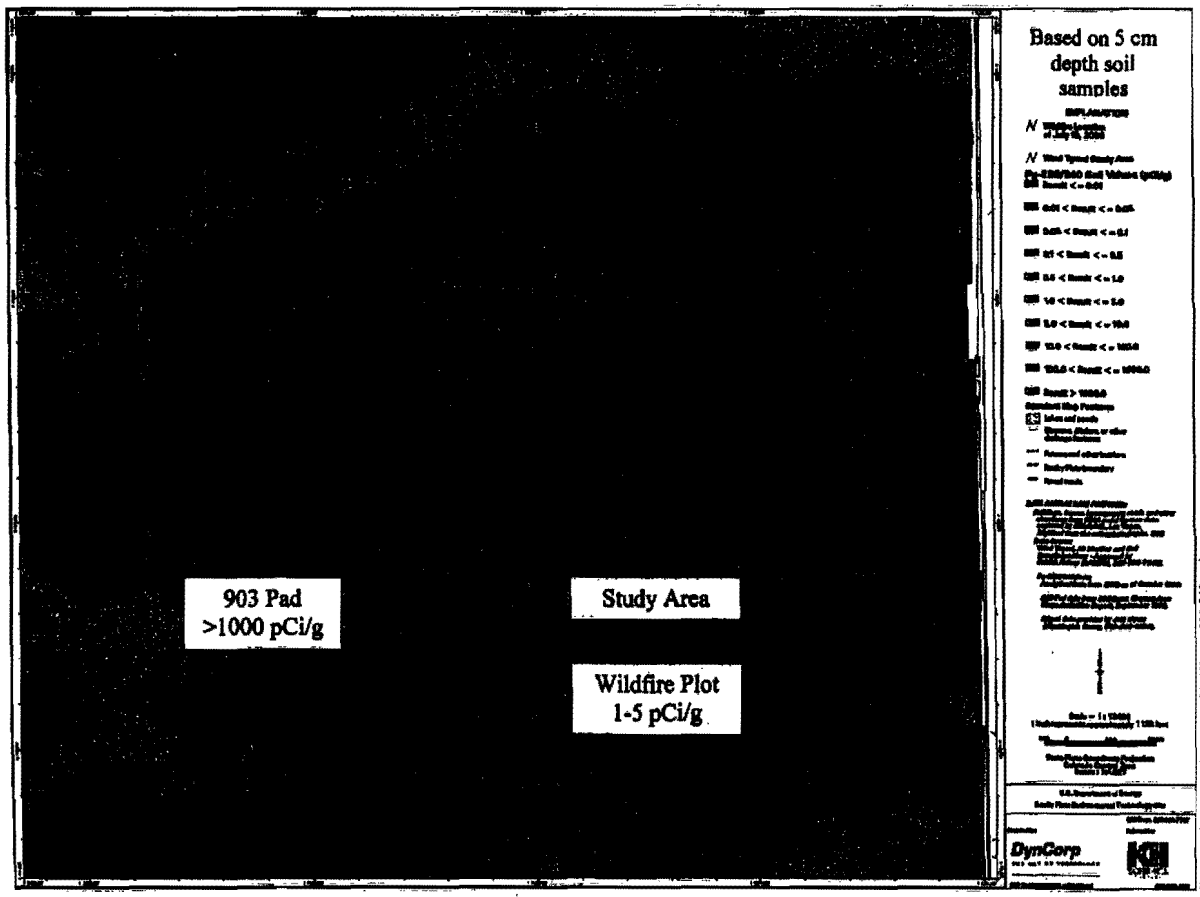
result of spills and releases during the Site's nuclear weapons production era. One of these areas, the 903 Pad, was contaminated when waste oils containing plutonium leaked out of corroded storage drums into the surrounding soils; this area source is a primary contributor to potential airborne plutonium exposure to public receptors near the Site.[2] Figure 1 illustrates the migration of plutonium particles in the predominant downwind direction from 903 Pad toward the study area. The isopleths shown are based on soil samples of 5 cm depth (the Site standard soil sampling procedure requires homogenized samples of 5 cm depth[3]). Strong westerly winds have redistributed plutonium contamination to the east of the 903 Pad. No spills or other significant releases are known to have occurred in the study area to the east of this source area – all plutonium activity in the study area soil is believed to have been redistributed from the 903 Pad through natural environmental processes, primarily wind resuspension and deposition.

In July 2000, a wildfire caused by lightning burned approximately 10 acres of grassland in the Site's buffer zone (a large, undeveloped area surrounding the Site's industrial area) east of the 903 Pad. The burned area was denuded of vegetation and thatch, which increased undisturbed soil erosion potential by removing much of the protective cover. Previous wind tunnel experiments at the Site indicated that the dense natural cover of the buffer zone vegetation and thatch reduced initial wind erosion potential by up to nine-fold compared to soil exposed after a prescribed fire.[4] The soil exposed by the wildfire, which was contaminated over time through air deposition of plutonium-contaminated particles originating from the 903 Pad, provided an opportunity to determine experimentally the relationship between soil plutonium activity and plutonium activity in the dust resuspended from that soil.

Related Studies

Studies by Ranville, et al.,[5] have suggested that soil plutonium activity is partitioned among particle size classes in proportion to the particle masses, not in proportion to particle surface area. As will be shown in this paper, soil samples taken in this study confirmed Ranville's observations. These results are also consistent with Langer's characterization of plutonium activity correlating to increasing airborne particle size downwind of the 903 Pad.[6]

Figure 1: Buffer Zone Plutonium Isopleth



METHODS

Testing of wind-generated particle emissions was initiated on August 22, 2000. A wind tunnel provided by Midwest Research Institute was used to perform the tests. The wind tunnel is similar in design to the wind tunnel described in the *Air/Superfund National Technical Guidance Study Series, Volume II, Estimates of Baseline Air Emissions at Superfund Sites,*[7] but has a smaller working section (15 cm x 240 cm open floor) and cross-section (15 cm by 15 cm). Prior to conducting wind tunnel trials, soil samples of 1 to 2 cm depth were collected from four discrete locations around the wind tunnel test plots.

Soil Sampling

Four locations surrounding the wind tunnel test plots in the wildfire area were selected for surface soil sampling. These locations were representative of the source material available to wind erosion, based on soil type, slope angle, and type and density of vegetative cover. The texture of soils throughout the study area was essentially homogenous.

Soil samples were incrementally collected using whisk brooms and shallow pans. Surface soil was sampled to a depth of less than 2 cm. In each sampling area, approximately eight incremental samples were hand sieved into three size fractions until sufficient quantity of each size fraction had been obtained to fill a 125 milliliter (ml) sample jar. The purpose of the sample size segregation was to determine whether plutonium (Pu-239/240) specific activity (radioactivity per unit mass) in the fine fraction, which would be preferentially resuspended by the wind tunnel, was significantly different than plutonium specific activity in the bulk soil. The Site soil sampling protocol[3] requiring samples of 5 cm depth was not used for this study because particulates were not expected to become resuspended by wind from such depth. The <2 cm depth soil samples collected during this study corresponded with the 1-2 cm depth disturbance created by raking in two of the four wind tunnel trials.

Each incremental surface soil sample was hand sieved, using a nest of two sieves and a bottom pan. The coarse soil particles were collected on the top sieve, a standard sieve No. 30 (600 micrometer [μm] openings). An intermediate soil fraction passed through sieve No. 30 but was captured on a standard sieve No. 200 (75 μm openings). Finally, the fine fraction passed the standard sieve No. 200 and was captured in the pan. Before the incremental surface soil samples were sieved, the larger pebbles (few) and larger pieces of organic material (dead and burnt grass, occasional deer droppings) were manually retrieved and discarded.

Sieving was performed by manually rotating and tapping the covered nest of sieves at the sampling location. Forty rotations were performed by hand for each sample, and the sieves were tapped by hand after each ten rotations. After hand sieving, each size fraction was transferred to a labeled 125 ml sample bottle. This method is very similar to the hand-sieving procedure found in AP-42, EPA's *Compilation of Air Pollutant*

Emission Factors.[8] A gravimetric analysis of each size fraction was performed in the field at each sample location to determine the mass ratio of the soil size fractions.

Wind Tunnel Trials

In operating the wind tunnel, the open-floored test section was placed directly over the surface to be tested. Air was drawn through the tunnel at controlled velocities, increasing at 2 meter per second (m/s) (5 mile per hour [mph]) increments, to a maximum velocity of about 27 mph at the tunnel centerline. This corresponded to a wind speed of approximately 100 mph at 10 meters (m) height.[9]

A pitot was used to measure the centerline wind speed in the open-floored test section. The volumetric flow rate through the wind tunnel was determined from a published relationship between the maximum centerline velocity in a circular duct and the average velocity, as a function of Reynolds' number.[10] Because the ratio of the centerline wind speed in the sampling extension to the centerline wind speed in the working section was nearly independent of flow rate, the ratio could be used to determine isokinetic sampling conditions for any flow rate in the tunnel.

The exit air stream from the test section was passed through a circular duct fitted with a sampling probe near the downstream end. The particulate sampling train, which was operated at 68 cubic meters per hour (m^3/hr) (40 actual cubic feet per minute [acfm]), consists of the tapered sampling probe pointed into the airstream, cyclone pre-collector, glass fiber backup filter, and high-volume motor. Sampled total suspended particulate (TSP) emissions were separated into two particle size fractions by the calibrated[11] cyclone: particles smaller than 10 μm aerodynamic equivalent diameter (PM10) were collected on the backup filter below the cyclone, while the cyclone captured particles larger than PM10.

For test surfaces without a well-defined threshold velocity, as was the case for this study, sampling was initiated as air began to flow through the wind tunnel. After the prescribed sampling period, which ended at the highest wind speed plateau, the flow was shut off and the particulate samples were recovered. The cyclone catch was sieved using a standard sieve No. 325 (45 μm openings) to remove vegetative material and detritus. The sieved portion of the cyclone catch, when recombined (mathematically) with the PM10 from the backup filter, represented TSP.

A high-volume ambient air sampler was operated at 68 m^3/hr (40 acfm) near the inlet of the wind tunnel to provide for measurement of the contribution of the ambient background particulate matter. The filter was vertically oriented, parallel to the tunnel inlet face. By sampling under light ambient wind conditions, background interference from upwind erosion sources can usually be minimized. Unfortunately, during two of the four wind tunnel tests it appeared that recirculation of wind tunnel exhaust or dust resuspended during surface preparation of test plots may have entered the wind tunnel inlet and co-located background sampler. Background corrections of sample masses and activities accounted for any such bias.

Dust samples from the field tests were returned to an environmentally controlled laboratory for gravimetric analysis. Glass fiber filters were conditioned at constant temperature (23 degrees Celsius [°C] ±1°C) and relative humidity (45% ± 5%) for 24 hours prior to weighing (the same conditioning procedure as used before tare weighing). The particulate catch from the cyclone pre-collector was weighed in the tared poly bag.

Isotopic Analyses

After weighing, all soil and particulate samples were ashed and acid digested. Plutonium (Pu-239/240) was separated from other radioisotopes by anion exchange chromatography, then counted by alpha spectroscopy.[12,13] The Pu-239/240 specific activity in soil and cyclone dust was reported in pCi/g; Pu-239/240 activity on filters was reported in disintegrations per minute per filter (dpm/filter) and converted to pCi/g using the following equation:

$$\text{Specific Activity, pCi/g} = \frac{(\text{filter activity, dpm})(0.45\ \text{pCi/dpm})}{(\text{filter weight, g} - \text{filter tare, g})} \qquad (1)$$

RESULTS

As shown in Table 1, over 90% of the surface soil in the wildfire area was in the coarse and intermediate particle size ranges. The coarse soil fraction contained the least Pu-239/240 specific activity ($\bar{x}$ =1.27 pCi/g, σ =0.24). The two smaller size fractions exhibited similar specific activities of 2.09 pCi/g for the intermediate fraction (σ =0.35), and 1.77 pCi/g for the fine fraction (σ =0.31), with slightly more activity among intermediate size particles. Though variability existed among the specific activities of the three soil size fractions, the mean specific activity in the fine (resuspendable) fraction was essentially equal to the mean specific activity of the bulk soil. This suggests that, for this soil reservoir, specific activity of resuspendable fractions (fines) is equivalent to bulk soil specific activity. This is useful because most soil data for the Site do not include particle size information.

Four wind tunnel trials, identified as CB-20, CB-21, CB-22, and CB-23, were conducted in the study area. Runs CB-20 and CB-21 were conducted on a soil surface that was artificially disturbed by raking to a depth of 2 cm. At the time, it was not known if sufficient mass could be generated from an undisturbed wildfire surface, and the raking ensured the release of adequate soil emissions for characterization of plutonium specific activity. After preliminary analysis of mass collected during runs CB-20 and CB-21, runs CB-22 and CB-23 were conducted on an undisturbed soil surface. These tests best represent the natural soil erosion process for the study area. Table 2 summarizes the results.

Table 1. Soil Sample Results

	<75μm particles				75-600μm particles				>600μm particles			
Sample Point	1	2	3	4	1	2	3	4	1	2	3	4
Mass fraction of soil (%)	8.2	7.2	10.1	7.4	42.0	39.6	43.3	43.0	49.7	53.2	46.5	49.4
Mean mass fraction (%) / SD[a]	8.2 / 1.3				42.0 / 1.7				49.7 / 2.7			
Specific activity (pCi/g)	2.09	0.94	2.37	1.66	2.23	1.54	2.20	2.40	1.03	0.85	1.78	1.43
Total activity (pCi)	275.9	112.8	348.5	229.1	238.6	154.0	237.6	249.6	80.3	36.6	90.8	74.4
Mean specific activity (pCi/g) / SD[a]	1.77 / 0.31				2.09 / 0.35				1.27 / 0.24			
	<75μm particles				<600μm particles				All soil particles			
Mean Specific Activity, pCi/g / SD[a]	1.77 / 0.31				2.03 / 0.24				1.66 / 0.17			

[a] Relative standard deviation, square root of sum of squares method
pCi/g = picocuries per gram

Table 2. Wind Tunnel Sample Results

Background-Corrected Sample Characteristic[a]	Raked		Undisturbed	
	CB-20	CB-21	CB-22	CB-23
TSP mass (g)	0.381	0.608	0.108	0.011
PM10 mass (g)	0.196	0.461	0.070	0.023
TSP activity (pCi)	0.608	1.153	0.129	0.023
PM10 activity (pCi)	0.203	0.859	0.078	0.013
TSP mean specific activity (pCi/g[b]) / SD[c]	1.75 / 0.46		1.64 / 0.80	
PM10 mean specific activity (pCi/g[b]) / SD[c]	1.45 / 0.26		0.85 / 0.05	

[a] Data is blank and background corrected
[b] Calculated using Equation (1)
[c] Relative standard deviation, square root of sum of squares method
pCi/g = picocuries per gram

Ambient background concentrations were measured during all four tests, as summarized in Table 3. Because runs CB-21 and CB-22 were performed on the same day, only one background sample was required. Wind tunnel effluent samples were corrected for the corresponding particulate mass and activity measured in the background samples.

Table 3. Ambient Background Sample Results

Background Sample Characteristic[a]	During CB-20	During CB-21	During CB-22	During CB-23
TSP concentration (mg/m^3)	0.677	0.053	0.053	0.240
PM10 concentration[b] (mg/m^3)	0.264	0.020	0.020	0.093
TSP activity (pCi/m^3)	1.33E-03	5.50E-06	5.50E-06	1.82E-04
PM10 activity[b] (pCi/m^3)	5.20E-04	2.14E-06	2.14E-06	7.11E-05

[a] Data are blank corrected

[b] 39% of TSP mass and activity is assumed to occur as PM10, based on Site-specific data

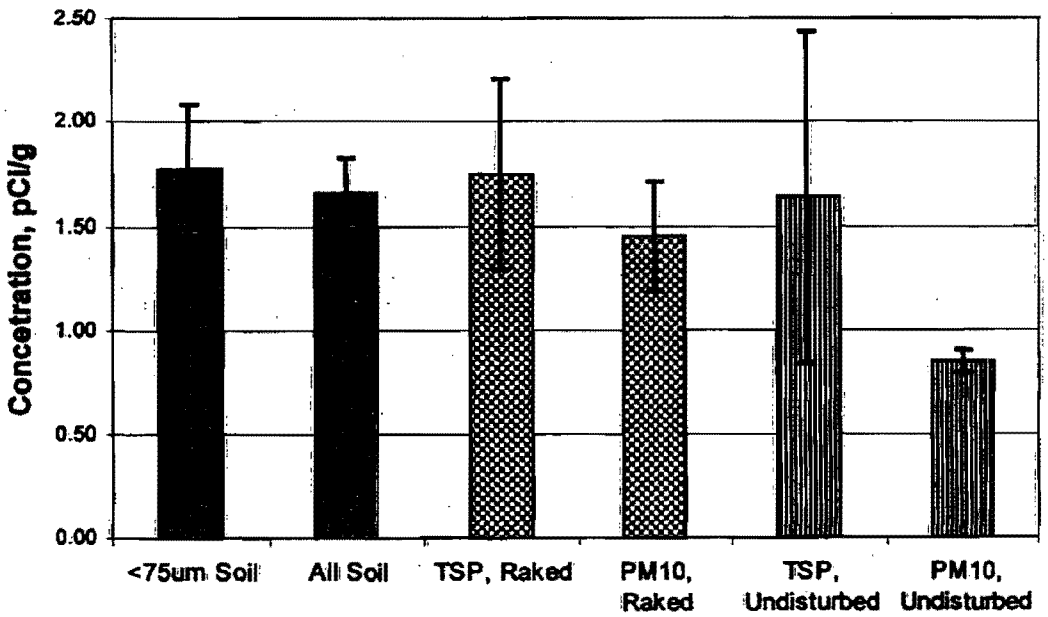
mg/m^3 = *milligrams per cubic meter*

pCi/m^3 = picocuries per cubic meter

The background particulate concentration and activity concentration measured during runs CB-21 and CB-22 are believed to represent the typical background levels for the test location. During background sampling for run CB-20, some contamination of the background sample filter may have occurred when dust raised during the preparation of the test plots by raking migrated into the wind tunnel air intake and into the background sampler, causing a significantly elevated background level. During background sampling for run CB-23, light and variable winds may have recirculated wind tunnel effluent, causing a slightly elevated background level.

Wind tunnel samples were compared to soil samples to quantify the correlation between the plutonium specific activity in resuspended dust and the parent soil, as shown in Figure 2.

Figure 2: Specific Activity of Wind Tunnel Dust Versus Soil



CONCLUSIONS

Figure 2 illustrates the similarities in specific activity between soil, TSP, and PM10 observed in this study. Specific activity in TSP resuspended from disturbed (raked) soil ($\bar{x}$ =1.75 pCi/g, σ =0.46) was not statistically different from the specific activity in both the fine soil and total soil reservoirs (1.77 and 1.66 pCi/g, respectively).[14] Specific activity in the PM10 resuspended from disturbed soil appears lower than TSP or soil specific activities, but was also statistically indistinguishable from either data set.[14] The undisturbed soil yielded PM10 particles with significantly lower specific activity than the soil reservoir even though undisturbed TSP specific activity was consistent with soil.[14]

The simplest explanation for the reduced specific activity observed in PM10 eroded from undisturbed surfaces is that the erodible layer of the undisturbed surface contains less Pu-239/240 than the erodible layer in disturbed soil. The explanation for the reduced Pu-239/240 presence is less clear. One possible explanation is that the uppermost thin layer of surface soil may be less contaminated with Pu-239/240 than a slightly lower soil profile at 1 to 2 cm below the surface, as a result of surface deposition of "cleaner" particles over top of more contaminated soil particles or through the downward migration of Pu-239/240 due to weathering. This explanation would be consistent with a redistribution model for the 903 Pad that moved most of the Pu-239/240 in a historical "slug" of migration, perhaps associated with past remediation efforts. It would also explain why mixed contaminated soil demonstrates more plutonium activity than undisturbed soil that has only the dilute surface layer available for erosion.

Alternatively, a periodic redistribution of Pu-239/240 may occur, in which the upslope winds of summer limit Pu-239/240 migration and cover the study area surface crust with "clean" dust from off-Site. During winter months, when upslope winds are infrequent, less dilution may occur. Since all test runs had essentially equivalent sample volumes, and background corrections were performed, the greater erosion potential of the disturbed soil reservoir nullified the diluting effects of uncontaminated fine particles sitting on the soil crust.

Based on these results, it appears that models and risk assessments for remediation projects and other events that disturb the soil surface could reliably assume that resuspended dust specific activity will match soil reservoir specific activity, assuming the soil specific activity is well defined for the depth of soil subject to wind erosion. This study suggests that the specific activity of fine soil particles may be accurately represented by the specific activity of bulk soil, but additional research would be needed to verify this hypothesis. Predicting resuspension of Pu-239/240 from undisturbed surfaces is less straightforward, due to the reduced specific activity observed in resuspended PM10 compared to soil. Sources of the apparent dilution effect could be explored in future experiments.

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