

Reprinted from
6 August 1976, Volume 193, pp. 488-490

SCIENCE

Plutonium Hazard in Respirable Dust on the Surface of Soil

Carl J. Johnson, Ronald R. Tidball and Ronald C. Severson

Plutonium Hazard in Respirable Dust on the Surface of Soil

Abstract. *Plutonium-239 in the fine particulate soil fraction of surface dust is subject to suspension by air currents and is a potential health hazard to humans who may inhale it. This respirable particulate fraction is defined as particles ≤ 5 micrometers. The respirable fraction of surface dust was separated by ultrasonic dispersion and a standard water-sedimentation procedure. Plutonium concentrations in this fraction of off-site soils located downwind from the Rocky Flats Nuclear Weapons Plant (Jefferson County, Colorado) were as much as 380 times the background concentration. It is proposed that this method of evaluation defines more precisely the potential health hazard from the respirable fraction of plutonium-contaminated soils.*

Methods of evaluating Pu inventories in soils are important because of the possibility of soil contamination near Pu processing plants and nuclear generating stations and areas where Pu has been accidentally released—for example, at Palomares, Spain, and Thule, Greenland, where Pu was released in airplane

accidents in January 1966 and January 1968, respectively. Evaluation of Pu (*1*) in the soil is of special importance in contaminated areas that are now considered for residential development. One such area is in the vicinity of the Rocky Flats Nuclear Weapons Plant (Jefferson County, Colorado), which is currently oper-

ated by Rockwell International for the Energy Research and Development Administration (ERDA). Activities at the plant include processing radioactive chemicals and making weapons from radioactive metals (*2*).

The Colorado State Health Department in 1973 proposed an interim standard for soil contaminated with Pu, setting the maximum allowable concentration at 2 disintegrations per minute per gram (dpm/g) (*3*). Land with Pu concentrations in excess of the standard would require ameliorative treatment before residential development could be approved. However, the standard fails to define "soil." Either single or composite samples of the soil at a depth of 0 to 0.5 cm from numerous locations in a development area are required. Because such samples include soil particles much too large to be resuspended or inhaled, the possible risk to health cannot be properly evaluated (*4*). Further, no provision is made to prevent the treated soil from being recontaminated by redeposition of Pu from more highly contaminated soils upwind. This redeposition mechanism potentially exists because winds in the area exceed 30 km/hour for 500 to 600 hours yearly. Wind speeds commonly reach 130 km/hour or more, with winds blowing predominantly to the east and southeast toward the Denver metropolitan area (Figs. 1 and 2).

The plant is located about 16 km northwest of Denver and about 8 km from the cities of Boulder, Westminster, and Arvada. Approximately 200,000 people live within 16 km and 600,000 people within 32 km of the plant. Residential development is now proposed within about 5 km of the plant (Fig. 1), involving as many as 3000 homes or a potential population of about 10,000 persons (*5*).

Since the plant began operation in 1953, there have been two major fires (1957 and 1969), a large release of Pu to off-site soils from a spill of metal-laden cutting oil, and an accidental release of Pu to the air in 1974. The major sources of off-site contamination are considered to be emissions from the 1957 fire and the oil leakage from corroded barrels of contaminated cutting oil that were stored

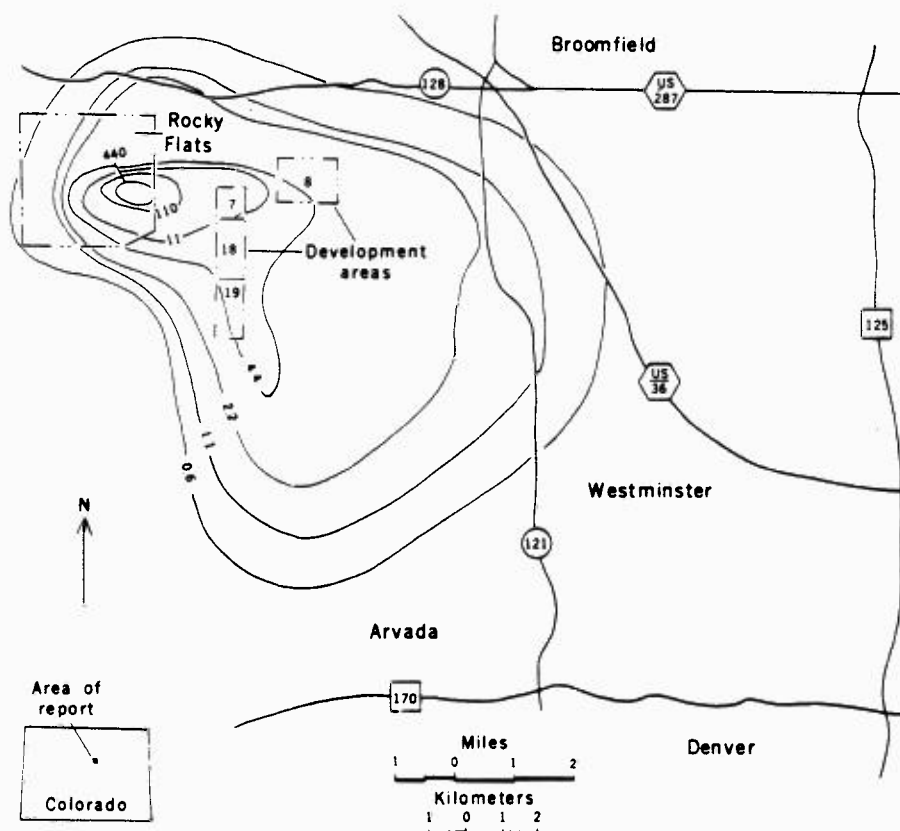


Fig. 1. Rocky Flats Nuclear Weapons Plant and proposed housing development area. Isopleths are labeled in disintegrations per minute per gram of whole soil, calculated from values in (*2*).

outside beginning in 1958 (2, 6). Although leakage of the barrels was first detected in 1964, storage in this manner continued until 1968. The oil-spill area has since been partially covered with asphalt.

A survey of Pu inventories in off-site soils was conducted by the Health and Safety Laboratory of the Atomic Energy Commission in 1970 (2). The results are used in Fig. 1. An off-site area of more than 50 km² had concentrations of Pu in excess of 10 mc/km². Soil samples were collected to a depth of 20 cm, which was considered sufficiently deep to account for the total deposition of Pu. The Pu inventory was based on the weight of the whole soil sample, including plant parts but excluding larger rocks.

Contaminated soils must be measured against a background of Pu released during atmospheric weapons testing. Of the 300 to 500 kc of Pu released worldwide, about 10 to 15 kc is estimated to be present in the soils of the United States and less than 10 percent is still suspended in the atmosphere. In Colorado the background level has been estimated to be 0.04 pc/cm² or 0.08 dpm per gram of whole soil (7).

Sampling localities, each about 4 ha in area, were selected within proposed residential development areas downwind from the plant (Fig. 1). Several sampling sites were randomly selected within each locality. In addition, one stream-sediment sample in section 18 and one sample of eolian sediment in section 19 were collected. Within a 4-m² area at each site, when the ground surface was dry, a representative quantity of loose, surficial (about 0 to 0.5 cm deep) soil material was collected with a clean brush and a clean plastic container. This area provided a sufficiently large composite sample. All samples were compared against a background level estimated from a control sample collected about 23 km south-southeast of the Rocky Flats plant.

The samples were analyzed in random order so that any systematic laboratory error would be converted to a random error. The objective in sample preparation was to disperse the soil microaggregates to expose the Pu as much as possible. Each sample was sieved through a 2-mm stainless steel screen; only the material that passed through the screen was retained for analysis. Approximately 50 g of material in the size fraction ≤ 2 mm was placed on a steam bath and treated with hydrogen peroxide to oxidize the organic material, particularly that present as grain coatings or cementing agents.

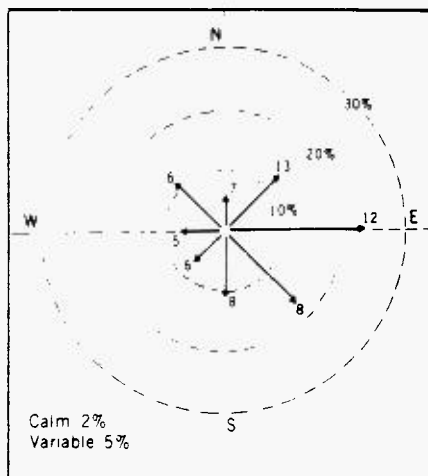


Fig. 2. Rose diagram showing average direction and velocity of wind at Rocky Flats for 1953 to 1970. Arrows point in the direction of wind movement; velocity (miles per hour) is given at the end of each arrow; concentric circles show frequency of wind direction (2).

Table 1. Analyses of Pu in respirable dust (size fraction $\leq 5 \mu\text{m}$ in soil material ≤ 2 mm) and in whole soil. In column 2 respirable dust is shown as the percentage of whole soil (≤ 2 mm). Localities are shown in Fig. 1. Values are given as disintegrations per minute per gram of material $\leq 5 \mu\text{m}$; for Health Department samples these are averages of two analyses. Relative enrichment (RE) is the ratio of measured value to background value.

| Local-ity | Health Department samples | | | | Contractor's samples (13) | |
|------------|---------------------------|------------------------|-----|-------|---------------------------|--|
| | Respi-rable dust (%) | Pu in respi-rable dust | | | | |
| | | dpm/g | RE | dpm/g | RE | |
| Section 7 | | | | | | |
| 7-1 | 36.1 | 83 | 180 | 13.5 | 169 | |
| 7-2 | 41.4 | 59 | 130 | | | |
| 7-3 | 17.9 | 120 | 270 | | | |
| 7-4 | 18.8 | 170 | 380 | 14.1 | 176 | |
| Section 18 | | | | | | |
| 18-1 | 19.8 | 36 | 80 | 0.2 | 2.5 | |
| 18-2 | 29.6 | 24 | 53 | 0.14 | 1.8 | |
| 18-3 | 27.0 | 26 | 58 | 2.96 | 37 | |
| 18-4 | 25.1 | 40 | 89 | 0.14 | 1.8 | |
| Section 19 | | | | | | |
| 19-1 | 62.2 | 1.4* | 3.1 | 0.23 | 2.9 | |
| 19-2 | 46.3 | 2.1† | 4.7 | | | |
| 19-3 | 36.5 | 1.3† | 2.9 | | | |
| Section 8 | | | | | | |
| 8-1 | 47.7 | 1.1† | 2.4 | 0.05 | 0.6 | |
| 8-2 | 51.2 | 1.0† | 2.2 | | | |
| 8-3 | 42.1 | 3.8† | 8.4 | | | |
| 8-4 | 31.5 | 2.8† | 6.2 | 0.72 | 9.0 | |
| 8-5 | 47.0 | 9.6 | 21 | | | |
| 8-6 | 48.8 | 8.1 | 18 | | | |
| 8-7 | 37.6 | 11 | 24 | | | |
| 8-8 | 33.3 | 7.7 | 17 | | | |
| 8-9 | 24.8 | 14 | 31 | 0.72 | 9.0 | |
| 8-10 | 25.7 | 19 | 42 | | | |
| 8-11 | 34.5 | 9.4 | 21 | | | |
| 8-12 | 37.9 | 6.9 | 15 | | | |
| Background | | | | | | |
| Control | 48.9 | 0.45 | 1 | 0.08 | 1 | |

*Eolian sediment from ground surface. †Sample from recently disturbed soil.

The samples were washed and filtered to remove soluble salts and dispersed with a 300-watt ultrasonic probe for 15 minutes (8).

Particles were separated according to size by a standard water-sedimentation technique (9). Sodium metaphosphate was added when necessary to avoid flocculation. The suspension containing the desired size fraction was collected and freeze-dried (10). The sedimentation technique is an arbitrary measure of the "effective" diameter of particles with irregular shapes that have settling rates equivalent to those of spheres of the same diameter and density. The threshold parameters used were based on particles of plutonium oxide having an effective maximum diameter of $5 \mu\text{m}$ and a density of 11.46 g/cm^3 . The soil particles separated include other mineral grains that have an equivalent maximum settling velocity, but that also have some combination of smaller density and larger diameter. The size fraction thus defined is hereafter called the soil material $\leq 5 \mu\text{m}$ or "respirable dust" because we assume this size fraction to be an adequate measure of the respirable material.

Plutonium concentrations are given in Table 1. The results shown are the averages of the determinations for duplicate split samples by two laboratories (1). Averaging the two determinations is justified because a *t*-test indicates no significant difference between determinations at $P = .05$. Analytical procedures used for Pu were those described by Talvitie (11, 12).

Our estimate of background is 0.45 dpm per gram of soil material $\leq 5 \mu\text{m}$. Samples from areas immediately downwind from the plant show evidence of Pu contamination considerably above background in the respirable dust. The sampling area in section 7, which is about 2.4 km immediately downwind from the plant area, has the largest amounts of Pu: 59 to 170 dpm/g. Sites 7-3 and 7-4 are on the flat crest of a low ridge that trends east from the plant, and sites 7-1 and 7-2 are on the north-facing slope of the same ridge but about 12 m lower in elevation. Section 8, which is on the principal wind vector but about 5 km downwind, has 1 to 19 dpm/g. Although sample sites in section 18 are nearly the same distance from the plant as those in section 7, we measured less Pu there, perhaps because section 18 lies away from the principal wind vector. Section 19, which is farther from the plant and from the principal wind vector, has the lowest values. Eolian sediment (sample 19-1) that was derived from a freshly plowed field upwind

had only 1.4 dpm/g. The field is located in a zone of expected contamination about 3.3 km southeast of the plant area. Plowing tends to distribute the Pu throughout the plowed layer and reduce its probability of entrainment for the present, but it also creates a more erosive surface until plant cover is reestablished. Stream sediment collected in section 18 contained 9.4 dpm/g. This sediment could be susceptible to wind erosion during periods when the stream was at low flow or dried up.

The distribution of Pu in respirable dust may be compared with previous determinations by other methods in the same areas (13). However, the different denominators used to express concentrations make it inappropriate to draw direct comparisons between Pu in respirable dust and in whole soil. The present maximum allowable level in Colorado, 2 dpm per gram of whole soil, represents an enrichment of 25 times the background level for whole soil (0.08 dpm/g). Corresponding enrichment factors based on respirable dust are given in Table 1: for example, sample 7-1 contains 180 times more Pu in the respirable dust fraction than is contained in our background sample.

The surficial soil materials at different locations contain different percentages of material $\leq 5 \mu\text{m}$ (Table 1). The Pu concentration per gram represents a potential dose rate. This concentration is to be distinguished from a total inventory of respirable Pu. The greatest long-term hazard, expressed as the largest total inventory, occurs at sites where both the concentration per gram and the percentage of material $\leq 5 \mu\text{m}$ are high.

Several sampling sites appear to have been disturbed recently by land-development activity. In particular, sites 8-1 through 8-4 lie in a zone where elevated levels of Pu could be expected. These samples have Pu enrichment factors of no more than about eight times background, compared with 15 to 40 times background only 0.6 km closer to the source (samples 8-5 through 8-12). Disturbance of the ground surface may account for this diminished concentration, or the local topography may have produced a fallout "shadow."

Estimates of health hazards from Pu have been made on the basis of air-monitoring data (14), measurements of total soil inventories of Pu (2, 6), and measurements of Pu concentration on the surface of the soil (13). Air-monitoring data are of importance in estimating human exposure through inhalation, and have been used with soil-contamination data to esti-

mate resuspension factors (15). These factors are influenced by the moisture content of soil, wind speed, elevation above ground, type of ground cover, and presence of paved surfaces. Mechanical disturbances such as those produced by plowing, vehicular traffic, construction work, or street sweeping can resuspend surface dust. However, air-monitoring data and resuspension factors do not account for the total Pu hazard in residential areas (16). Some examples of other types of potential exposure to Pu in the respirable dust on the surface of the soil are listed below.

1) Children playing on the ground or adults working outdoors can pick up mud and dirt on their shoes and clothing and thus introduce dust into their homes. Washing and drying of contaminated clothes can release significant amounts of dust through the exhaust of the dryer. Such a mechanism of exposure has been demonstrated by the finding of characteristic neoplasms in the wives of asbestos workers (17, 18). Dust can also enter a house through windows and ventilating systems and be resuspended by household vacuuming and other cleaning operations (19). Pets track in fine particulates, which may eventually become airborne. These conditions can occur even if a developed area has grass cover.

2) Children playing outdoors may eat food with soiled hands and in other ways ingest or inhale unusual amounts of dust. Heavy-metal intake by this method was found to be important in children with lead poisoning who lived near a smelter at El Paso, Texas (20).

3) Heavily used playgrounds tend to be dusty, and strenuous playground activity can result in suspension and inhalation of dust by children.

4) Local resuspension of dust may occur in the preparation and maintenance of domestic gardens. Plutonium that has been plowed under before residential development may again be exposed by digging for gardens.

The present regulatory code in Colorado requires that soils with radiation contamination that surpass the interim standard of 2 dpm per gram of whole soil must receive special treatment to reduce the hazard to acceptable levels (7). Mixing the soil by plowing is presently accepted as one technique for treatment of contaminated land. We believe that this is an insufficient treatment because (i) plowing tends to displace the Pu particles from the ground surface to some position at depth, but the Pu is still in the soil; (ii) a treated area can be recontaminated either from untreated land upwind

or from subsequent accidental releases of radioactive material; and (iii) subsequent disturbance of the soil by construction activity or cultivation by home gardeners may expose Pu particles at the surface again.

Useful data for evaluating the health hazards of Pu contamination of soil may be obtained by determining the amount of Pu in the respirable-dust fraction (material $\leq 5 \mu\text{m}$) on the surface of the soil. It would be more realistic to base interim standards on the respirable-dust fraction because the very small particles in this fraction have the greatest potential for suspension and inhalation.

CARL J. JOHNSON
Jefferson County Health Department,
Lakewood, Colorado 80226

RONALD R. TIDBALL
RONALD C. SEVERSON
U.S. Geological Survey, Federal Center,
Lakewood, Colorado 80225

References and Notes

1. Samples were analyzed for ^{238}Pu , ^{239}Pu , and ^{240}Pu in laboratories of both the Colorado State Health Department and the Energy Research and Development Administration. Both ^{238}Pu and ^{240}Pu are reported as ^{239}Pu ; ^{239}Pu is not reported, but values ranged from 0.04 to 12.4 dpm/g.
2. P. W. Krey and E. P. Hardy, *USAECL Publ. HASL-235* (1970).
3. R. L. Cleere, unpublished memorandum.
4. C. J. Johnson, unpublished report.
5. —, in *Proceeding of Public Hearings on Plutonium and Other Trans-Uranium Elements*, vol. 2, *Proceedings of Hearings in Denver, Colorado, January 10, 1975* (No. ORP/CSD-75-1, Environmental Protection Agency, Washington, D.C., 1975), pp. 208-224.
6. S. E. Poet and E. A. Martell, *Health Phys.* 23, 537 (1972).
7. Final Report of the Lamm-Wirth Task Force on Rocky Flats to the Colorado State Health Department, Denver, 1 October 1975. The following relationship is given in this report: $(\text{mc}/\text{km}^2) \times 0.2 = \text{dpm}/\text{g}$.
8. A. P. Edwards and J. M. Bremner, *J. Soil Sci.* 18, 47 (1967).
9. P. R. Day, in *Methods of Soil Analysis*, E. A. Black, Ed. (American Society of Agronomy, Madison, Wis., 1965), part 1, p. 545.
10. R. L. Malcolm, *U.S. Geol. Surv. Prof. Pap.* 600-C (1968), pp. C211-C216.
11. N. A. Talvitt, *Anal. Chem.* 43, 1827 (1971).
12. —, *ibid.* 44, 280 (1972).
13. L.F.E. Environmental Analyses Laboratories, Richmond, Calif., unpublished reports of soil analyses for Pu (1973).
14. M. A. Thompson and D. D. Hornbacher, *Dow Chem. USA Rocky Flats Div. Publ. RFP-ENV-74* (1974).
15. J. W. Healy, *Los Alamos Sci. Lab. Publ. LA 5483-MS, UC-41* (1974).
16. Despite the fact that air-monitoring data do not corroborate the redistribution of Pu, resuspension of contaminated dust from the ground surface has occurred, because we do find Pu widely scattered downwind.
17. A. Fahy, *Natl. Tuberc. Respir. Dis. Assoc. Bull.* (December 1972), p. 12.
18. P. Champion, *Am. Rev. Respir. Dis.* 103, 293 (1968).
19. N. M. Lefcoe and I. I. Inoulet, *Arch. Environ. Health* 30, 565 (1975).
20. B. F. Rosenblum, J. M. Scholtz, J. J. Chisolm, *U.S. Morb. Mortal. Wkly. Rep.* 22, 405 (1972).
21. We are indebted to G. Lucas of the Colorado School of Mines, who provided assistance in the sample design; M. Hanrahan of the Colorado State Health Department, who assisted in the survey procedure; and to D. L. Bokowski, ERDA, and C. Rising, Colorado State Health Department Laboratories, who performed the analysis for plutonium.

17 February 1976; revised 27 April 1976