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EVALUATION OF THE HAZARD TO RESIDENTS OF AREAS CONTAMINATED WITH PLUTONIUM

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INTRODUCTION

Plutonium oxide particles deposited on the ground surface by accidental spills or atmospheric fallout (Table 1) are subject to resuspension by wind or other means. Particles in the size range of 5 μm and smaller are considered to be of respirable size because when inhaled they may be retained within the lung(1). Most of the plutonium oxide particles released offsite by nuclear installations are in this size range(2). This paper describes a method of measuring the concentration of plutonium in the potentially respirable surface dust and discusses the potential health effects of exposure to such dust.

SAMPLE COLLECTION AND PROCESSING

In our study area (downwind from the Rocky Flats plant) sites for sampling were selected following guidelines proposed by the Atomic Energy Commission(3). A composite sample of the loose, surficial (about 0-0.5 cm deep) soil material was collected with a clean brush and a clean plastic container within an area of 4 m^2 when the ground surface was dry(4). Plutonium oxide particles likely exist in association with other soil particles as micro-aggregates and, therefore, behave as such. Microaggregates of soil are dynamic and are affected by freezing and thawing, wetting and drying, the kind and amount of natural cementing agents present, and by other forces that tend to disrupt or reconstitute them(5). These plutonium-oxide-microaggregate associations cannot be measured except by methods that tend to alter their natural state. In contrast, our procedure seeks to maximize the dispersion of the microaggregates to expose the plutonium oxide particles and to evaluate the maximum potential hazard. Attention to the dispersion of microaggregates will tend to alleviate the problem of the effect of microaggregation on the precision of the data and will provide data that are comparable from season to season or from site to site.

About 50 g of soil material that passed a 2-mm screen was treated by standard methods with hydrogen peroxide to remove organic material. The sample was then washed and filtered to remove soluble salts, and was mechanically dispersed with a 300 watt ultrasonic probe for 15 minutes. Sodium metaphosphate was added when necessary to facilitate dispersion of the particles.

Particles of respirable size were separated by size and density with a standard water-sedimentation technique(6). The threshold settling velocity was computed from Stoke's equation using an effective diameter of 5 μm and a density of 11.36 g/cm^3 (plutonium oxide). The size fraction thus collected includes other mineral particles with equivalent settling velocities that have some combination of smaller density and larger diameter. The collected fraction was freeze-dried and analyzed for plutonium by radiochemical procedures(7,8). Contamination of offsite soils in the study area (an area proposed for residential development) was found to be as great as 380 times the background value. We believe that the concentration of plutonium in the respirable fraction of surface dust, as defined here, is an index that can be more readily related to the potential health hazard than other indices utilized for this purpose.

PARTICLE SIZE AND ACTIVITY

Relationships between particle size, volume, mass and radioactivity are shown in Table 2. A particle of plutonium oxide one μm in diameter weighs about six millionths of a microgram (μg). This particle is very small but has an activity of 0.3 picocuries (pCi) or about one disintegration every 1.5 minutes. A particle 5 μm in size has a mass of

.0007µg. This particle is also of respirable size and will have an activity of about 41 pCi or some 90 disintegrations per minute (dpm). One hundred of the 5 µm particles together would weigh about 0.07 µg. Since plutonium has a biological activity 15 or more times as great as radium, this many particles of plutonium oxide may be similar in effect to 0.5 µg of radium, the smallest bone dose found associated with osteosarcoma in radium dial painters[9]. Although plutonium may be present in contaminated areas offsite in extremely small quantities, this may be sufficient to produce bone cancer and other types of tumors.

ESTIMATES OF DOSAGE AND EFFECT

The recommended dose limit of plutonium 239 (occupational exposure) to lung is at present 15 rem/year or 16,000 pCi. This maximum permissable annual dose (MPAD) is equivalent to about 400 five µm particles, or a total mass of about 0.3 µg of plutonium oxide. A single particle of plutonium oxide 40 µm in size has more than this much activity and is within the range of atmospheric dust (0.3 to 100 µm). A particle of this size might not be retained in the respiratory tract, but could lodge in an abrasion or other wound.

Meyers gives evidence to support a recommendation that a much more realistic limit than the present 15 rem MPAD for lung burden is the "maximum permissable pulmonary lymph node burden" which is placed at 230 pCi [10]. The maximum lung level that could produce this is 67 pCi. Meyers points out that this is less than 0.5% of the currently accepted MPAD (occupational) of 15 rems for the lung. The inhalation and retention of two 5 µm particles annually would exceed this amount. Morgan also demonstrates that the present limit for exposure to plutonium may be too high by a factor of 240 or more, in relation to potential effects on bone[11]. Again, two of the 5 µm particles would exceed this more conservative dose limit.

The relative risk of inhaling and retaining plutonium oxide particles in reference to the amount of plutonium in the respirable dust is illustrated in Table 3. A single 4 µm particle may produce about 50 dpm, equivalent to about 20 millirems. One such particle in 25 grams of respirable dust will produce an average activity of 2 dpm/gram of dust. If a person has inhaled and retained 12.5 grams of dust, he has a 50% chance of inhaling that 4 µm particle of plutonium oxide. However, the particle may be in the first bit of dust inhaled or in the last bit. The probability is one in 100, expressed in the table as p=.01, that if he inhales only 1/4 gram, that he may inhale the 5 µm particle. Retention of two of these particles, or an equivalent larger number of smaller particles, would exceed dose limits proposed by Morgan and Meyers. It may be that in the very first whiff of dust (25 milligrams) that he may inhale the particle. The chances are about one in one thousand that this could occur. In a total population of 100,000 or more who could be housed in a contaminated area such as that near the Rocky Flats plant, these odds seem significant.

The effects on populations of exposure to low levels of radiation have been studied by the Advisory Committee on the Biological Effects of Radiation of the National Academy of Sciences and the National Research Council[12]. The committee discusses the Federal Radiation Council's maximum permissable level of 5 rem over 30 years (170 mrem/year). This dosage, 170 mrem, is equivalent to about four of the five µm plutonium oxide particles, or to about 20 grams of respirable dust with 20 dpm/g of plutonium. The committee thinks that this level would likely result in an increase of cancer deaths by about 2%. In addition, they calculate that the effect of 170 mrem/year would cause in the first generation an increased incidence of 0.05% of serious, dominant, or x-linked diseases and defects per year. This is one birth in 2,000. However, after several generations, these numbers would be five times larger, that is, there would be one birth in 400 with defects and serious dominant, or x-linked diseases per year. In

addition the committee states that when the congenital abnormalities and constitutional diseases which are partly genetic are added to this, the total incidence would be one case per thousand births in the first generation, and about one case in 133 births for succeeding generations. The committee also believes that between 5% and 50% of ill health is proportional to the mutation rate. This much radiation per generation could eventually lead to an increase of 5% in the ill health of the population.
Estimates of excess rates of cancer may also be calculated in the following manner, again referring to an area with 20 dpm/g of plutonium in respirable dust. This example is a calculation of the increased rate of bone cancer in such a contaminated area.

$$\frac{10^6 \text{ person-rems}^{(13)}}{10 \text{ excess bone cancer cases}} \cdot \frac{1000 \text{ pCi(inhaled)}^{(14)}}{3.2 \text{ rem (bone)}} \cdot \frac{2.2 \text{ dpm}}{1 \text{ pCi}} \cdot \frac{1 \text{ g}}{20 \text{ dpm}} = \frac{3.4 \text{ g dust}}{1 \text{ excess bone cancer}/10^6 \text{ persons}}$$

Offsite lands in our study area (downwind from the Rocky Flats plant) have levels far in excess of this amount, and a development project five kilometers east of the plant site has levels that approach 20 dpm/g in respirable dust. We see that about 3.4 grams of such dust (32 pCi), inhaled and retained, may result in one excess bone cancer in a population of one million people over a life span. Other effects from this dosage of radiation may be calculated in a similar manner. Such effects would be multiplied for each additional 3-4 grams of respirable dust inhaled*. Over a period of months or years a person may inhale and retain 10 to 12 grams or more of resuspended dust (100 pCi) and may receive about 0.1 rem to the lung, 4-5 rem to the trachio-bronchial lymph nodés, and 0.12 rem to the liver. The gonadal and fetal dose is difficult to evaluate, but plutonium has been found in gonadal tissue and in the fetus. This amount of exposure to one million people could result in about 160 excess deaths due to leukemia and a total increase in all neoplasms of perhaps 1 to 3 per cent over a period of 70 years[12]. This may include from 6 to 1000 excess cases of bone cancer (this large range is due to two points of view)[11,12]. The incidence of all genetic diseases may increase by 1.5% and ill health related to chromosome mutation by 10%, for all succeeding generations[12]. Other factors, such as the ingrowth of Americium, use of chelating agents in fertilizers, etc. may change the nature of the risk and actually increase the incidence of effects. Because plutonium is stored in the body to a considerable extent, a person living several years in such an area may inhale and retain enough plutonium to produce these effects even though he may leave the area.
Effects are more likely to occur in the fetus, the child, and in persons with increased susceptibility to neoplasms. There is evidence that smaller doses of radiation are more harmful per rad than would be expected. Stewart and Neal and others have shown a greater risk of leukemia per rad at low intrauterine exposures, 0.25-0.5 rad, than at higher doses to the fetus[15]. One rem (equivalent to 25 5μm particles) may cause an 80% increase in mortality from childhood cancers after fetal exposure, and a 0.9% increase in recognized "spontaneous" abortions[13].

CONCLUSION

Federal guidelines are being promulgated by the U.S. Environmental Protection Agency to guide the use, treatment, and rehabilitation of land contaminated with plutonium. At the present time there are areas in the United States which have levels of contamination of 19 dpm/g in respirable dust which are presently being used or developed for residential purposes[16]. The importance of the establishment of a maximum allowable level of contamination based on the concentration of plutonium in the respirable dust must be emphasized. This level may be appropriately set as low as 2 dpm/g, since there is sufficient evidence to indicate probable health effects at the 20 dpm/g level and possibly at levels below this figure.

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* As much as 67 mg of dust per cubic meter (mg/m^3) has been measured downwind from farm equipment. An average value of 0.135 mg/m^3 was observed in Denver in 1970-73. Average annual respiratory volume is about 8000 m^3 for an adult.

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Table 1
Concentrations of Plutonium in Soil

Location	Disintegrations per minute/gram
Worldwide (fallout)	0.0 to 0.4
Central New Mexico (Trinity)*	0.3 to 22.2
Nevada Test Site*	0.2 to 22.2
Los Alamos Scientific Laboratory*	0.01 to 111
Rocky Flats*	0.4 to 211
Bikini Atoll	2.9 to 422
Palomares	0 to 3996
Eniwetok Atoll	76 to 7104

*Offsite
Adapted from Ref. 17

Table 2
Plutonium oxide particles: Volume in cubic micrometers (μm^3) mass in micrograms (μg) and radioactivity of plutonium 239 in picocuries (pCi) and disintegrations per minute (dpm) by particle size in μm, for sphere-shaped particles.

Particle size in μm	Volume μm^3	Mass μg	Activity pCi	Activity dpm
40	33,510	0.33	21,000	46,000
5	61	0.7×10^{-3}	41	90
4	34	3.8×10^{-4}	21	46
2	4.2	4.9×10^{-5}	2.6	5.8
1	0.5	6.1×10^{-6}	0.3	0.7

Table 3
Relative risk of inhaling and retaining plutonium particles, by level of radioactivity of respirable dust and quantity of contaminated dust inhaled.

Activity of plutonium in respirable dust	Assuming one particle, grams dust inhaled and retained to retain 50 dpm* p=.5	p=.01	p=.001
2 dpm/g	12.5	0.25	0.025
5 dpm/g	5	0.10	0.01
10 dpm/g	2.5	0.05	0.005
20 dpm/g	1.2	0.02	0.002

* 50 dpm may be produced by one particle about 4 μm in diameter, and is equivalent to about 20 millirems.

Table 4
Dose-equivalent in rem per 1000 picocuries of plutonium 239 inhaled.

Organ	Plutonium 239
Lung*	0.9
Lymph Node*	43.7
Bone	3.2
Liver	1.3
Kidney	0.2
Gonads	0.05

* Assumes that particles are retained for a longer period of time in the lung. (Adapted from Ref. 14)

Table 5
Estimated rates of plutonium-induced cancer and hereditary injury per million person-rems

Prediction based on data from:	Lung	Bone	Hereditary Injury
Man*	16-110	2-17	50**
Dog	70	70	
Rat	60(700***)	10	
Mouse		20	

* BEIR Report (also 1-7 cases of liver cancer per million person-rems)
** 50 cases per million person-rems for immediate family, 300 cases for all generations and a maximum of 600 for defects of complex origin. Expected increase in hereditary injury is about 1% per rem for the major and different indices of genetic effect.
*** 238 Pu (Adapted from ref. 14)