Negligible hazards from recently measured Rocky Flats hot particles

D. M. Wood, November 2020

Note: Blue links are clickable and will take you to the corresponding section of the document. Green numbered links are clickable and will take you to the corresponding reference in the bibliography. Green links are direct Web links.

As of late November 2020, a 2-page non-technical flyer is available here.

Contents

Key points

- *Most* plutonium worker radiation exposure historically has been in the form of inhaled 'hot particles' [Centers for Disease Control and Prevention]
- International and U.S. radiation regulatory agencies have had quantitative risk estimates for inhalation of alpha-emitting hot

particles since the 1970s. These are well understood for all relevant radioisotopes for Rocky Flats, partly thanks to Rocky Flats workers closely followed for decades after plant fires.

- We examine a simple scenario and its implications: *How many $^{239}PuO_2$ hot particles of a specified size would need to be inhaled to raise lifetime cancer risk by 1%?* Using the most recent recommended regulatory data and Ketterer and Szechenyi's poster data on how often hot particles appear in the Rocky Flats soil they measured, we estimate the amount of soil in kg that would need to be inhaled for this radiation dose.
- Using EPA recommended values for soil inhaled per day by adults, we determine *how long* it would take to inhale or swallow this much soil in the form of dust.
- We briefly consider the excess cancer risk for workers on the Jefferson Parkway Public Highway project.

Based on our results, we conclude that *under no plausible circumstances does the inhalation of Pu hot particles around Rocky Flats pose a health hazard to workers, recreational users, or neighbors.*

Introduction

The data from soil testing during the summer of 2019 along the right-of-way for the planned 'Jefferson Parkway' was more-or-less as expected based on previous work. There was one giant anomaly—a sample which showed an activity of 264 pCi per gram of soil. This was rapidly traced to a single, large 'hot particle' of PuO_2 with an equivalent diameter of 8.8 microns (10^{-6} meter) [1]. Thus to characterize this sample in terms of pCi per gram of soil (used for *uniform* contamination) was/is misleading.

It is important to note that ^{239}Pu and ^{240}Pu in Rocky Flats soil are very weak emitters of gamma radiation, the most penetrating. They emit only alpha particles, with a range in air of about 1.5 inches. As a result the principal exposure to radiation from plutonium is by inhalation of contaminated soil.

In July 2020, the Colorado Department of Public Health and the Environment (CDPHE) released the report [2] "Review of potential radiation doses during construction of the Jefferson Parkway", which summarized these findings and addressed the issue of soil safety. However, they did so in a 'brute force' way–they re-ran the large, somewhat opaque DOE radiation safety code RESRAD in a scenario in which *all of the soil* at Rocky Flats was contaminated to a level of 264 pCi per gram of soil to a depth of 2 meters. They found in this 'worst case scenario' that the resulting exposure was *still* within the negotiated safety limits for Rocky Flats.

Concerns about special toxicity of small particles of alpha emitters ('hot particles') were allayed by the mid-1970s [3]. Because of *ongoing misinformation* about the health impact of hot particles, however, it is important to address head on the issue of inhalation of hot particles. A much more refined treatment is possible with this direct approach. We focus on what the measurements (discussed below) of Dr. Michael Ketterer imply about health risks due to 'hot particles'. This builds on analysis carried out in detail in the document Hot particles on the web site `rockyflatsneighbors.org`, but is meant to be complete in its own right.

Ketterer's measurements were carried out on samples collected in 2019 on the eastern edges of the Rocky Flats National Wildlife Refuge, and in 2000-2002 in Westminster Open Space. As of November 2020 none of Dr. Ketterer's work has been published in peer-reviewed journals, so our analysis of his data is based on figures in posters distributed by the Rocky Flats Peace & Justice Center [4] and the Colorado chapter of Physicians for Social Responsibility [5].

We show readers the process by which cancer risk due to inhalation of hot particles is assessed; the point is that this is straightforward. We show
(i) how the measured activities of soil samples and hot particles may be used to extract baseline soil levels of ^{239}Pu and approximate sizes of $^{239}PuO_2$ hot particles. (This process is already partly described by Dr. Ketterer [4], [6]).
(ii) how the frameworks of the International Committee on Radiological Protection (ICRP) and of the U.S. governmental agencies (Department of Energy, the EPA, the CDC, and the Department of Health and Human Services) may be used to estimate lifetime cancer risks due to inhalation of plutonium hot particles. These frameworks are based on data for workers who inhaled plutonium as the result of accidents at processing plants, including Rocky Flats.

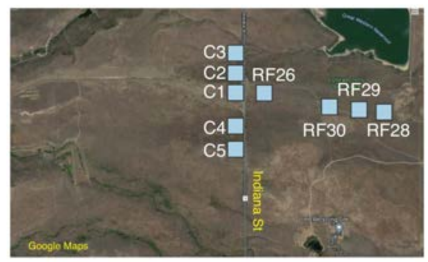

Figure 1: Approximate locations of Ketterer sampling areas, redrawn from [5].

Recent data of Ketterer and Szechenyi [K&S]

The initial K&S data [4] [6] covers four sample sets, drawn from a sequence of composite soil samples (composites 1-3) moving northward near the eastern boundary of the refuge (in the Jefferson Parkway right of way), and a fourth (RF-28) taken from soil roughly due east of composite 1, south of the east side of the Great Western Reservoir. *Addendum:* An additional poster [5] is available which reports on nine datasets, including the four original ones. The one labeled RF-28 above has changed significantly: there are now a total of 40 samples (up from 35), 5 hot particles (up from 4), a baseline radiation level of 0.377 pCi/g (up from 0.294), and an average hot particle diameter of 0.82μ (up from 0.71). The authors show an approximate particle size histogram, based on these and other samples not yet reported. The distribution peaks at about 0.6-0.8μ and there are no particles above about 2.2μ in diameter. See Appendix A.

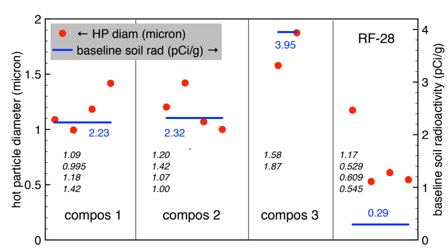

Figure 2: Graphical representation of four initial KS datasets, using their partitioning of samples into 'baseline' (average: blue line) or hot particle (red dots). Average baseline ^{239}Pu soil levels in pCi/g, shown in blue text. Hot particle diameters in μ (microns) shown in small italics: only two figures are probably significant.

According to Ketterer [6], individual samples were 200 mg in mass; this permits identification later of the average number of hot particles per kg of soil. For each sample the mass of ^{239}Pu was determined experimentally and expressed as the ratio of picograms of the isotope per gram of soil in the sample. K&S only partially report hot particle diameters.

We note that 1 picogram of ^{239}Pu per gram of soil (pg/g) is 0.0620703 pCi/g, a unit more commonly used for Rocky Flats.

For our analysis the figures from the posters were carefully digitized. For each, the mean of the 'baseline' samples was established and subtracted from the putative 'hot particle' values. This excess mass was attributed to a single, spherical hot particle. This mass, multiplied by the specific activity for ^{239}Pu, yields the excess activity in pCi. Using the numerical expressions discussed below we can find the diameter in microns (μ). In each case where K&S report a range of diameters, our values agree. Baseline levels of ^{239}Pu and average particle diameters from their their datasets are shown in Appendix A below. We also redraw there a current histogram of their observed particle diameters. Finally, assuming that the measurements reported by K&S are *typical* (not cherry-picked for hot particles), we can establish for each sampling region the mean number of hot particles per kg of soil. Properties of their data sets are summarized in Table 1 in the margin.

We note that composites 1 and 2 (and samples RF-29 and RF-30) have very similar properties, not surprising since they come from adjacent areas. The largest hot particles (and by far the highest baseline activity level) come from composite 3.

We expect relative fluctuations in, say, the baseline soil activity, of $1/\sqrt{N}$ where N is the sample number for the data set. This is roughly 15-18%.

set	HP	samps	pCi/g	$\bar{d}(\mu)$	HP/kg
C1	4	32	2.23	1.2	625
C2	4	39	2.32	1.2	510
C3	2	43	3.95	1.7	230
C4†	2	34	1.37	1.1	300
C5†	2	39	1.08	1.7	260
RF-26†	6	43	1.07	0.92	700
RF-28*	5	40	0.38	0.82	625
RF-29†	9	43	0.30	0.67	1000
RF-30†	4	35	0.30	0.72	570

†*New data since first poster*
**Values have changed from first poster*

Table 1: Summary of K&S datasets [5] available October 2020. 'HP' and 'samps' indicate the number of hot particles and the total number of samples in the dataset. Baseline soil radioactivity ('activ') is in pCi per gram of soil, average hot particle diameter $\bar{d}$ in microns.

Relating measured activity to particle size

We sketch the process of relating the diameter of a single hot particle to its 'activity', the number of alpha particles emitted per second. The number of Pu nuclei per unit volume in PuO_2 may be found from the its observed crystal structure. This may be used to calculate the number N of Pu nuclei in a sphere (ball) of specified diameter. If we multiply this by the activity per ^{239}Pu nucleus s^* (= 9.1164 $\times 10^{-13}$ decays per nucleus per second) we have the number of decays per second of a 'hot particle' of specified diameter.

For concreteness we will take ^{239}Pu as the isotope of interest at Rocky Flats. We find for the (radio)activity A

$$A = Ns^* = .01214\ d_\mu^3 \text{ Bq} = 0.3282\ d_\mu^3 \text{ pCi} \tag{1}$$

where d_μ is the particle diameter in microns; 1 Bq = 1 decay per second. The (measureable) activity is the input for predicting excess cancer risk due to radiation exposure. As can be seen from the scale of A for one particle, significant radiation doses will require inhalation of many hot particles.

Because PuO_2 is more dense than lead, large hot particles partly absorb their own alpha particles. We can include this effect by a 'transmission factor', ranging between 0 and 100%, which weights the energy emitted by alpha particles by the fraction of alpha particles not absorbed by the hot particle itself, shown below.

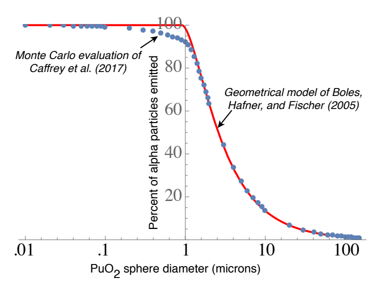

Figure 3: Fraction of alpha particles transmitted by a spherical $^{239}PuO_2$ particle of diameter d_μ in microns. Blue dots are numerical values [7]; red curve is for a geometric *model* [8].

The 1% excess risk scenario

We therefore introduce a specific scenario in which we ask, *How many 'hot particles' of a particular diameter do we need to inhale to raise our* *lifetime cancer risk by 1%?* We will return to this shortly.

From inhaled radioactivity to cancer risk

Radiation dose is defined as energy deposited per unit mass of tissue, in units of 'grays' (joules per kilogram). Tissue damage, on the other hand, must acknowledge the specific properties of the radiation, generally done by weighting the radiation dose in Gy (grays) by the 'relative biological effectiveness' of the radiation type, taken generally as 20 for alpha particles. The resulting quantity, within the framework [9] of the International Committee on Radiological Protection (ICRP), is known as the *effective dose*, measured in sieverts (Sv), the international unit of effective radiation dose.

Naturally under some circumstances (for example, exposure to background radiation in a fixed location) the exposure is incremental and 'external' to the body. The situation we consider here is a presumed brief or one-time *ingestion* of radioactive material (an *internal* or 'committed' dose), which remains in the body permanently or is slowly excreted over a long period. While the document Hot particles on `rockyflatsneighbors.org` also discusses direct *dose*-based estimates of cancer based on the epidemiological 'excess relative risk', in this document we focus on the relation of cancer risk to hot particle *activity* (as computed above) using the frameworks adopted by the International Committee on Radiological Protection (ICRP) and a slightly different description used by U.S. governmental agencies (Department of Energy, EPA, CDC, and the Department of Health and Human Services). Within this framework we write

Inhaled, insoluble hot particles are a 'committed dose'—the particles are assumed to remain fixed in the lungs where they are only very slowly eliminated as the particles very slowly dissolve.

$$D(n) = c_d(n)A \tag{2}$$

where $D(n)$ is the dose (in sieverts, Sv), A is the activity (decays per second, in becquerel, Bq), and $c_d(n)$ is the 'dose coefficient', which depends on the number of years n elapsed since the exposure to the radiation. It thus has units of Sv/Bq [10].

The LNT assumption is adequate for large doses (typically taken to be doses above about 0.1 Sv) but probably overestimates risk below that, especially at low dose rates. Nonetheless it is widely used for regulatory purposes since it is convenient and is believed to overestimate risk at low to moderate doses.

The 'linear no-threshold' (LNT) description assumes that the cancer risk from a radiation dose is linearly proportional to that dose. This tacitly underlies all current risk calculations. Eq. (1) above for the activity A of a single hot particle is based directly on the physical properties of ^{239}Pu and PuO_2. In cancer epidemiology a 'lifetime' is generally assumed to be 50 years of exposure to a carcinogen such as radioactivity. Evidently biological assumptions (the mode of exposure, biokinetics of Pu removal, body mass, exposure time, *etc.*) will affect the total dose over time.

Strictly speaking, ignoring alpha self-shielding, setting $T(d_\mu)$=1, requires the hot particle diameter to be less than about 1μ to be valid.

The ICRP approach

Within the ICRP framework ([9]; for an overview, see [11]) all biological effects are 'soaked up' into the parameter $c_d(n)$ in Eq. (2), the dose coefficient. They include the mode of exposure (for the case of hot particles, inhalation), the gradual clearance of the radioactive substances from the body, as described by a 'biological half life', excretion, *etc.*. The size of c_d encodes the *observed* biological impact (that is, cancer diagnosis or cancer death) and the dependence on elapsed time n in years reflects measured gradual elimination of the radioisotope by excretion. The simplest functional form describing the behavior is shown in the Figure; there are many more details in Appendix B.

Tabulated dose coefficients are based on animal and human data, which incidentally show that the particle *size* itself is not as important as the net dose of radiation inhaled, which depends on particle size through its volume or surface area; see Fig 3. A great deal of additional information (including data collected from Rocky Flats workers from 1952-1989 followed for long periods after their employment) may be found in the 2010 document *Toxicological profile for plutonium* [12]

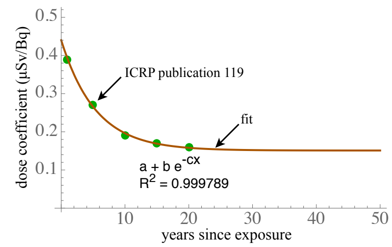

Figure 4: ICRP ^{239}Pu inhalation dose coefficient tabulated in ICRP Publication 119 [10] (blue points and dashed extrapolation). The decay over 20 years reflects the current value of the biological half-life and the baseline the 'permanent' contribution to dose.

PuO_2 is quite insoluble in water and, once in the lungs probably remains there for life.

International and U.S. risk estimates virtually coincide

Within the ICRP framework, there are three steps in determining cancer risk from exposure to internal radiation. The lifetime (50-year) risk of cancer ('morbidity', not 'mortality' [which is the risk of death]) is found as follows:

1. Find the 'dose coefficient' $c_d(50)$ for the particular mode of exposure and the radioisotope.

2. Determine the activity (decays per second) of the ingested radioisotope.

3. Use a source of excess relative risk for cancer morbidity and from the dose find the excess risk. The ICRP has its own tables of risk per Sv of radiation effective dose.

We find the dose coefficient from Annex G of Publication 119 [10] of the ICRP. The value is $1.6{\times}10^{-5}$ Sv/Bq. Table 1 of ICRP Publication 103 [9] gives the recommended risk per effective dose as 5.5% per Sv. Thus for a risk of 0.01 (1%) (as specified in our scenario above) a dose of 0.18182 Sv would be required. Using the dose coefficient and the activity per hot particle of a specified size, we can determine the required number of particles to be inhaled. As an example, we select hot particles of 3μ diameter. For $^{239}PuO_2$ the activity from

Eq. (1) is 0.328 Bq, so we would require 34,700 particles for the dose of 0.18182 Sv. We have thus confirmed that *many* particles will need to be inhaled for even the 1% excess risk scenario.

U.S. regulatory agencies *combine* what is effectively the dose coefficient with the absolute lifetime risk of cancer per unit radiation dose to find a 'risk coefficient' which depends on the radioisotope, the mode of exposure (ingestion, inhalation, *etc.*), and the solubility of the relevant inhaled material. It thus has units of absolute risk per Bq. Federal Guidance Report No. 13 (Table 2.1, p. 77) [13] gives a risk coefficient for the inhalation of ^{239}Pu of 8.96×10^{-7} per Bq or 3.32×10^{-8} per pCi for (quite insoluble, designated 'S' for the slow rate of absorption of $^{239}PuO_2$ inhaled in particulate form).

For a risk of 1% (0.01) we find that an inhaled activity of 3.01×10^5 pCi are needed, corresponding to [using Eq. (1)] 34,000 hot particles of 3μ diameter. The ICRP and U.S. estimates (34,000 vs. 34,600) are very similar. This does not imply the each framework is independently precise, but that the U.S. and the ICRP have been sharing data for decades, albeit parameterized in a slightly different way.

Thus a single 3μ hot particle contributes about 5.25×10^{-6} Sv over a 50-year lifetime. This should be compared with an average *annual* exposure to background radiation in the U.S. of about 3.1×10^{-3} Sv.

authority	dose coeff	risk/Sv	risk coeff
DOE/EPA	-	-	8.96×10^{-7}
ICRP	1.6×10^{-5}	.055	8.80×10^{-7}

Table 2: Comparison of U.S. and international lifetime cancer risk parameterizations for inhalation of $^{239}PuO_2$ particles in respirable size range, nominally 1μ diameter. Risk coefficients are per Bq of activity.

Contributions of Rocky Flats workers

The experiences of Rocky Flats plant workers have been invaluable in establishing the risk assessments described above for exposure to and inhalation of plutonium. According to the CDC, almost all exposures to plutonium came from inhaled or ingested hot particles. Ketterer [4] has claimed **"Decades of USDOE and CDPHE studies to date have failed to recognize and characterize Rocky Flats originating PuO_2 particles and have not assessed their risks to human health."** This statement reflects complete unfamiliarity with the epidemiological and scientific literature on hot particles, and with international and U.S. regulatory information. In fact, earlier data on the sizes and distribution of hot particles specific to Rocky Flats is surveyed in detail here and here.

By 1978 concerns about hot particles had been largely laid to rest. An excellent review in 1978 [3] notes

> *Rocky Flats Group.* This group also consists of 25 persons, who have inhaled $^{239}PuO_2$ aerosols as a result of a fire in a ^{239}Pu manufacturing plant in Rocky Flats on October 15, 1965; a mass related average of the aerodynamic particle diameter was 0.3μm. In all of these persons the ^{239}Pu activity in the lung was in excess of the permissible limit; it corresponded on the average to 10^4-10^5 hot particles with an activity of more than 0.07 pCi per particle [Ed: this corresponds to a particle diameter of about 0.6μ] (Mann et al., 1967). According to the Tamplin-Cochran hypothesis, such numbers of hot particles would result in an expected number of 5-50 lung tumors per person (Bair et al., 1974). In

Worked example: particle number

How many hot particles of diameter $d_\mu = 8.8$ microns (the size of the single hot particle found during Jefferson Parkway sampling) would you need to inhale to raise your lifetime (50-yr) cancer risk by 1%? We use ICRP data (Table 2).

1. *Find needed activity A:* Set the risk per Bq equal to 0.01 (risk): $.01 = 8.80 \times 10^{-7} \times A(Bq) \Rightarrow A = 1.136 \times 10^4$.
2. *Find activity of 1 particle:* From Eq. (1) $A_{part} = 0.01214 d_\mu^3 = 8.273$ Bq/particle.
3. *Solve for N_{part}:* Divide needed A by activity per particle: $N_{part} = 1370$.

This is larger than the number 400 I gave in September 2019 because none of the biological effects which reduce risk (and thus increase the particle number) were included in the older calculation.

> this group, too, no lung cancers have been found to date, i.e., 10 years after the accident. According to the experiences with uranium miners an increased frequency of lung cancers would have to manifest itself as early as 5 years following exposure.

The exhaustive 2010 document *Toxicological profile for plutonium* [12] notes

> *U.S. Nuclear Facilities (Hanford, Los Alamos, Rocky Flats).* Lung cancer mortality in plutonium workers employed at the Rocky Flats nuclear weapons plant has been examined in a case-control study (Brown et al. 2004). Lung cancer cases (n=180) were employed at the Rocky Flats facility for at least 6 months during the period 1952–1989, when plutonium pits were fabricated at the facility. The control group (n=720) consisted of Rocky Flats workers who were matched with cases for age, birth, year, and gender. Internal lung radiation doses in the cohort derived primarily from exposures to ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, and ^{238}U; however, 98% of the internal effective dose equivalents in cases (96% in controls) were estimated to have come from Pu and ^{241}Am (inbred from ^{241}Pu). Estimated effective dose equivalents for internal α radiation (cases plus controls) ranged from 0 (54%) to >940 mSv (5%). In the full cohort, the odds ratio for lung cancer mortality was significant for the internal lung dose strata 400–644 mSv, but was not significantly elevated at higher doses; there was no significant trend with dose (2.71, 95% CI: 1.20–6.09); the odds ratios were <1 for most dose categories for persons employed for <15 or >25 years. When the analysis was restricted to workers employed at the facility for 15–25 years, a significant trend was evident for increasing odds ratio in association with increasing internal lung effective dose equivalents; however, there was no evidence of a positive trend for those employed for <10 or ≥25 years.

An odds ratio less than 1 means that the exposed population had a *lower* risk than the unexposed. In epidemiology this is termed a 'protective factor'.

The quoted dose range in Sv should be compared with the lifetime dose found above for a single 3μ hot particle: 5.25×10^{-6}Sv.

A survey of National Institute for Occupation Safety and Health (NIOSH) studies of Rocky Flats workers includes publications through 2005. As noted in the August 2019 article, *USTUR Special Session Roundtable——US Transuranium and Uranium Registries (USTUR): A Five-decade Follow-up of Plutonium and Uranium Workers* [14]

> At Rocky Flats, a bioassay program was established to follow workers after they terminated employment. The resulting data continue to help researchers to improve the biokinetic models that are used to estimate intakes and radiation doses. After 50 y, the US Transuranium and Uranium Registries continues to contribute to our understanding of actinides in humans, which is a testament to the vision of its founders, the generosity of its tissue donors, and the many dedicated scientists who have worked together to achieve a common goal.

It is insulting to Rocky Flats workers who participated in these studies or donated their bodies to the USTUR tissue bank, and to researchers who carried out careful studies and followed workers' health for decades, to claim that anyone has "failed to recognize and characterize Rocky Flats originating PuO_2 particles and have not assessed their risks to human health".

Implications

It is very important to remember that since the radiation dose from a single hot particle is so tiny, the particle size alone conveys no information about the radiation hazard of inhalation—the actual number of particles breathed in is crucial. We found above the number of hot particles of diameter d_μ in microns needed to acquire an alpha radiation dose sufficient to raise the lifetime cancer risk by 1%. In compact form (with more than appropriate significant figures) using the ICRP values,

$$N^{1\%}_{part} = 9.3605 \times 10^5 / d^3_\mu \tag{3}$$

This clearly makes the point that the 'natural scale' of the required particle number is around 1 million particles for 1μ diameter (respirable range).

We now have a calculational framework which would be readily recognizable to and accepted by U.S. and international regulatory agencies. We have chosen to estimate the number of $^{239}PuO_2$ hot particles required to raise lifetime cancer risk by 1%. Our estimates above can be used together with the data of Ketterer and Szechenyi to answer other important questions. For instance, how many kg of soil from each region would need to be inhaled for this level of risk? If we have an estimate of the *rate* at which soil is inhaled by an adult, how long would it take for a typical human to inhale this much dirt?

Mass of inhaled dirt for 1% excess cancer risk

We note that the radioactivity inhaled for a fixed volume of dirt is proportional to $N_p\ \overline{d}^3_\mu$ where N_p is the number of particles in the volume and $\overline{d}_\mu$ is the average hot particle diameter. This factor will usually be dominated by $\overline{d}_\mu$ since it depends on its cube. We return to the K&S data of Table 1 and note that among all 9 regions composite C3 has the largest average hot particle diameter and thus the smallest total inhaled mass to achieve 1% excess cancer risk. Composite C3 (average particle diameter, 1.73μ) has about 233 hot particles per kg of dirt. To achieve the '1% dose' we must thus inhale about [Eq. (3)] 182,000 particles, requiring $182{,}000/233 \simeq 781$ kg of dirt. With an average density for Rocky Flats soil of about 1.5 g/cm^3, this corresponds to a ball of dirt about 39 inches in diameter.

It should be remembered that at least 95% of the alpha-particle emitting radioisotopes in Rocky Flats soil are naturally occurring.

The radiation exposure modeling program RESRAD used by the DOE uses this value.

Time needed to inhale dirt

The U.S. Environmental Protection Agency has long maintained standards for inhalation of potentially dangerous substances. This includes contaminated soil breathed in as fine dust. A recent report [15] reviews recent work, the careful analysis of statistics from many sources, and updates suggested daily values for soil and dust ingestion for children and adults. Their Table 5-1 gives a recommended value of 10 mg/day for soil ingestion (inhalation plus swallowing) for the general population. For the 95th percentile (very heavy breathing) the value is 50 mg/day.

The phrase 'upper percentile' is quite non-standard. It appears to mean "95th percentile rounded to one significant figure" [16]. According to the EPA [17], "Upper Percentile, $x_{0.95}$: Based upon an established background data set, a 95th percentile represents that statistic such that 95% of the sampled data will be less than or equal to $\leq x_{0.95}$. It is expected that an observation coming from the background population (or comparable to the background population) will be $\leq x_{0.95}$ with probability 0.95." This is *still* not very clear unless the 'UP' coincides with the 95th percentile.

We show in Table 3 results for the nine K&S data sets together with a tenth scenario informed by the others. This will be termed the 'exaggerated risk' (ER) case, with an average hot particle than the highest reported by K&S). In addition, in the ER case diameter of 3μ

set	part #	kg	diam	years
C1	580,000	930	42	260,000
C2	580,000	1,100	44	310,000
C3	180,000	780	39	210,000
C4	750,000	2,600	58	700,000
C5	200,000	800	40	220,000
RF-26	1,200,000	1,700	51	470,000
RF-28	1,700,000	2,700	59	730,000
RF-29	3,200,000	3,000	62	830,000
RF-30	2,600,000	4,500	70	1,200,000
ER	35,000	29	13	1,600

Table 3: For K&S data sets, extracted data for the '1% scenario'. Columns are number of hot particles required, kg of dirt which must be inhaled, diameter in inches of dirt ball with this mass assuming 1.5 g/cm^3, and the number of years of inhalation needed at 10 mg (ingested+inhaled) per day. The 'exaggerated risk' (ER) scenario assumes 1200 3μ diameter hot particles per kg of soil, and an inhalation rate of 50 mg/day (5× average).

(larger than the largest reported by Ketterer in any documents), and with 1200 hot particles per kg of soil (larger the rate at which dirt is inhaled or ingested is 50 mg per day, the EPA recommended value for the 'upper percentile', meaning that 95% of the population takes in less than this. Obviously as estimated directly from the data of Ketterer and Szechenyi several hundred thousand years (over which the actualy activity of ^{239}Pu would have declined to .2% of its initial radioactivity) would be required. This in turn means that over a 50-year lifespan for the C3 example, only $50/210,000 \simeq 2.4 \times 10^{-4}$ of the '1% excess risk' does would actually be achieved, reducing the risk to about 2.4×10^{-6}. Statistically, for 420,000 similarly exposed people one would develop an excess cancer.

The estimates above have been selected to be reasonable but with a bias toward overestimation of risk. They show that risks would have to be tens of thousands of times larger in order to be even detectable, much less to constitute any sort of health risk. It is worth noting that changes in lifestyle (adjustment of diet, vaccinations, tobacco use,

Worked example: inhalation mass and time

Exaggerated risk scenario: 3μ particles, 1200 per kg, 50 mg per day breathed

1. *Find number of hot particles:* Eq. (3) yields 34,700 particles
2. *Use number per kg to find total dirt weight:* 34700/1200 = 28.9 kg
3. *Find dirt volume:* Dirt density 1.5 g/cm^3 = 1500 kg/m^3, volume = 28.9 kg/(1500 kg/m^3) = 0.01927 m^3.
4. *Find dirt ball diameter D:* Equate ball volume $4\pi/3\,(D/2)^3$ to volume of dirt in cubic meters ⇒ D = 0.3326 m = 13.1 in.
5. *Find years T needed for inhalation:* 50 mg/day= 50 kg/$\left(10^6\text{days}\right)$, so T = 28.9/50 $\times 10^6$days × (1 year/365.25 days). ⇒ T = 1582 years

The *average* inhalation rate for adults is about 5 times lower, which would raise the needed time to 7900 years.

V 1.1

control of obesity, alcohol *etc.* may change lifetime cancer risks by 20-30%.

Worker exposure during 5-year Jefferson Parkway project

We briefly consider the excess cancer risk for workers on the Jefferson Parkway Public Highway project. Some of the motivation for soil testing was to assure the safety of such workers. We assume the project lasts about 5 years, that workers are present only on workdays (over-estimated as 280 days per year), and that they breathe in dust from soil at a rate higher than 95% of the population, 50 mg/day. For a more realistic estimate we select the K&S 'composite 3' data set, once again as the area with the largest hot particles; see Table 1. We also consider an 'exaggerated risk' (ER) case with a much higher density of hot particles per kg of dirt (1200). The 50-year lifetime

scenario	HP/kg	N_{hp}	$\overline{d_\mu}$	Bq inh	risk
C3	233	16.3	1.73	1.03	9.0×10^{-7}
ER	1200	84	3	27.5	2.4×10^{-5}

Table 4: Worker excess lifetime cancer risk for 5 years of work (280 workdays/yr) with dirt/dust inhalation rate of 50 mg/day (5 times average). We use ICRP risk coefficient [Table 2] for $^{239}PuO_2$ of 8.80×10^{-7} per inhaled Bq.

risk for the exaggerated risk scenario (unlikely since most workers in a high-profile project such as the Jefferson Parkway would be wearing masks and dust abatement procedures will probably be in effect during construction) means that on average out of every 42,000 workers one would expect 1 additional cancer.

Worked example: 5-year exposure risks

What is your excess lifetime risk of cancer if you breathe in hot particles from dirt for only 5 years?
Use the C3 sampling area as example: 233 hot particles/kg of dirt, average diameter 1.73 μ. Assume 10 mg per day of inhaled dust from dirt.

1. *Find amount of dirt inhaled:*
   5 yrs × 365.25 days/yr × 10 mg/day = 18.25 g = 0.01826 kg
2. *Find average number of hot particles in this much dirt:*
   .01826 kg × 233 particles/kg = 4.2
3. *Find activity of inhaled hot particles:* Use Eq. (1). 4.2 particles × .0629 Bq/particle = 0.264 Bq
4. *Find risk from this activity:* Use Table 2 ICRP value 8.80×10^{-7} per Bq: 2.32×10^{-7}.

(The ICRP value is for 50 years after the exposure, but we ignore this.) *Interpretation:* of every 43,000,000 people exposed similarly, one would develop a cancer (above the rate expected in the absence of plutonium hot particle exposure).

Takeaway messages

- Elementary calculations relate PuO_2 hot particle diameters to their (radio)activity.
- International (ICRP) or U.S. (DOE/EPA) tabulations based on the medical histories of nuclear plant workers who inhaled ^{239}Pu hot particles and animal data provide direct estimates of lifetime risks of excess cancers as the inhaled activity increases.
- Measurements reported by Ketterer and Szechenyi may be used to estimate the frequency and size of hot particles in soil along the (most heavily contaminated) eastern boundary of the Rocky Flats National Wildlife Refuge and the Jefferson Parkway right of way. These permit calculation of the number of hot particles and equivalent soil weight for a specified level of excess cancer risk.
- The EPA provides suggested standard values for daily amounts of inhaled/ingested dust from soil. These can be used to estimate the rate at which the radiation dose is acquired.

- For a scenario of 1% lifetime excess cancer risk, realistic estimates suggest that several hundred thousand years of inhalation would be required. For an 'exaggerated risk' scenario with much larger, more numerous particles than occurred for any region measured by Ketterer, this figure drops as low as 1,600 years.
- For more typical data, over a 50-year lifetime an adult is expected to inhale around 43 hot particles for a lifetime risk of about 2.4×10^{-6}. In statistical terms, this means that out of 420,000 similarly exposed adults, one will develop cancer.
- The Jefferson Public Parkway Highway Authority project is envisioned to take 5 years to complete. Realistic estimates indicate a cancer risk of about 9×10^{-7}, (about 1 in 1.1 million).
- The claims of K&S about health risks are completely without merit.

As with all calculations based on the linear no-threshold (LNT) description of radiation dose and cancer risk, the actual risk is probably well below what is estimated in this document. It should also be remembered that at least 95% of alpha-emitting radioisotopes in Rocky Flats soil are *natural* in origin.

Dedication

This document could not have been prepared without easy access to work done by scientists, technicians, and bureaucrats at a host of government agencies in the U.S. (aka the 'Deep State') and abroad. These include the Department of Energy, the Center for Disease Control and Prevention, the Environmental Protection Agency, and agencies like the World Health Organization and the International Committee on Radiological Protection. On these repositories of deep expertise and experience the health of the American people depend.

Those concerned about plutonium and health owe a debt of gratitude to workers at the former Rocky Flats plant (and other U.S. and European processing plants). Their misfortunes (accidents at the plants) and careful monitoring of their health for almost 65 years by epidemiologists and health physicists have made possible detailed understanding of plutonium biochemistry and health impacts. As of 2018, 12 Rocky Flats workers donated their bodies (and more than 120 others made partial body donations) for the study of plutonium health effects–more than at any other of the 16 DOE sites participating in the USTUR project.

Appendix A: Graphical summary of Ketterer and Szechenyi data

As of late October 2020 the data of K&S has not been published in peer-reviewed journals so we re-plot in a compact way the data from their most recent poster [5]. Each sample is assumed to have a mass of 200 mg. We accept their partitioning into 'baseline' samples and 'hot particle' samples with distinctly higher Pu masses. We use the standard deviation of baseline sample values, and of the measured hot particle diameters, as bars to indicate measurement uncertainties. We also note that while the number of hot particles in their nine samples sums to 38, the numbers shown in their histogram sum to 46, so evidently some of their data has not been presented.

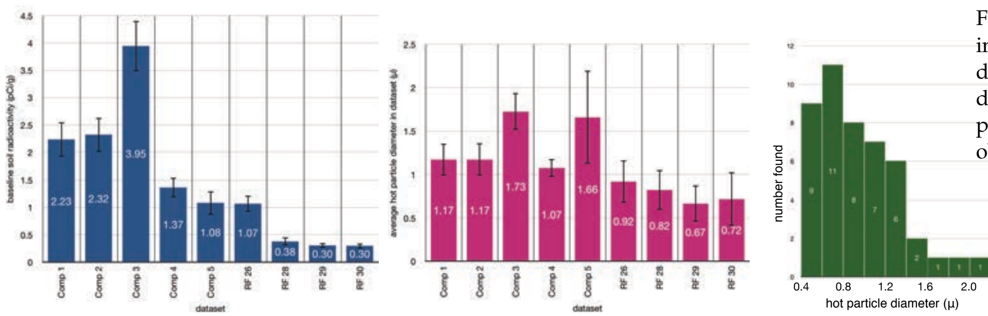

Figure 5: Baseline soil levels of ^{239}Pu in pCi per gram of soil for each of 9 datasets, left panel. Average hot particle diameter for each dataset, middle panel. Histogram of diameters of their observed hot particles, right panel.

Appendix B: Time-dependent dose coefficients

I have seen very little in the literature about what appears in this Appendix, hence its inclusion.

As noted in the text, the time-dependence of the ICRP dose coefficient reflects the biokinetics of solubilization and then excretion of (in our case) ^{239}Pu. The simplest functional form reflecting a decay to a steady-state value is $a + b\, e^{-cx}$. To my surprise (see top panel of Fig. 6), this is a very good representation of the ICRP table data (evaluated at 1, 5, 10, 15, and 20 years after exposure). The fit quality is so good that it is probable that table values were interpolated from such a continuous curve.

It is useful to think of the *activity* (decays per second) as what is absorbed, and the *dose* as the resulting effect. The general expression for the time dependence of the dose $D(t)$ as a result of a time-dependent activity A(t) delivered is

$$D(t) = \int_{t_0}^{t} dt'\, \mathring{A}(t')\, c(t - t') \quad (4)$$

$$c(\tau) = a + b\, e^{-\tau} \quad (5)$$

The activity rate begins at a reference time t_0; at the present time t, the integrand reflects the differential dose from $\mathring{A}(t')$ at all earlier times, weighted by the dose coefficient evaluated at the time lapsed between the activity and the present. In words: the dose D right now depends on the history of the delivered activity (via its time derivative $\dot{A}(t)$ weighted by the dose coefficient evaluated at the time elapsed between an earlier time t' and right now (time t). We use a common physics notation of an overdot indicating a time derivative, and would call the expression for $D(t)$ the 'convolution' of the activity delivery rate and the dose coefficient.

Convolutions are extremely common in physics and applied math.

Here we consider three important cases

1. *Exposure all at one time*
   This is a very common scenario for accidents. Under these conditions the activity absorbed is a 'step function' and so

$$\dot{A}(t) = A_0 \, \delta \left(t - t_1 \right) \tag{6}$$

   where δ indicates the 'Dirac delta function' and t_1 is when the dose is delivered. Then, using properties of the δ function, $D(t) = A_0 \, c \left(t - t_1 \right)$, where A_0 is the total (constant) activity absorbed. This simply recovers the usual expression.

2. *Exposure in a time window*
   This is also common. An example for *external radiation* might be a trans-Pacific 15 hour flight at 40,000 feet, during which a passenger is exposed to much higher levels of cosmic rays than at sea level. For internal exposure in which a radioactive substance is ingested, an example is breathing in contaminated soil at a fixed rate (say, in mg of soil per day) during a 5-year project. For this situation in the case where the exposure *rate* is constant

$$\dot{A}(t) = \dot{A}_0 \left[\theta \left(t - t_1 \right) - \theta \left(t - t_2 \right) \right] \tag{7}$$

   where $\dot{A}_0$ is the constant exposure rate (say, in pCi added per hour or Bq added per day). In words: the dose *rate* is a constant, but only between the starting time t_1 and the ending time t_2.

$$D(t) = \dot{A}_0 \left[a \left(t_2 - t_1 \right) + \frac{b e^{-ct}}{c} \left(e^{c t_2} - e^{c t_1} \right) \right]. \tag{8}$$

   As expected, for times long after the exposure this becomes a constant extra dose and even *during* the exposure the dose rises approximately linearly with time.

3. *Ongoing exposure:* As an example, at age 20 you move to a region where radon in houses is common and ask what your extra radiation dose would be when you are 70 years old. Here $t_1 = 20$ and the 'ending time' t_2 is the current time t ($t_2 = 70$)—the exposure is ongoing.

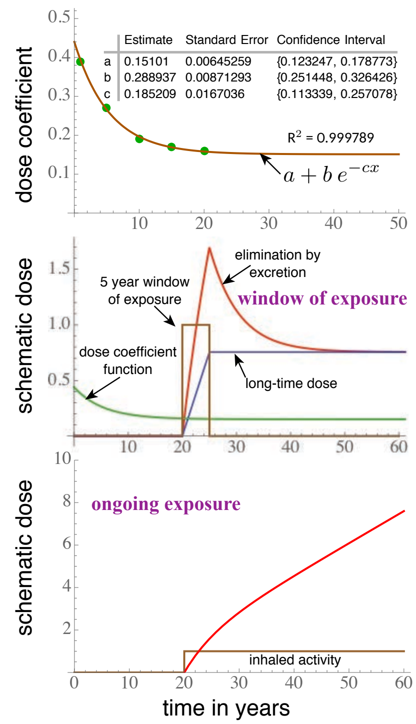

Figure 6: Dose coefficient function and examples: a window of exposure (middle panel) and ongoing exposure (bottom panel).

Appendix C: An overview of uncertainties

Physical properties of $^{239}PuO_2$ are known very precisely. What are the principal sources of uncertainty in the estimates above? My estimates are

- Ketterer's data for hot particles themselves, based on direct counting of nuclei in a mass spectrometer, are probably reliable to better than 5%.
- Statistical soil data from Ketterer's data, such as how often hot particles occur in soil may be uncertain to ±100% or more and suffer from relatively few samples and thus large fluctuations.
- ICRP 'dose coefficients', based on decades of assessment of health impacts of inhaled plutonium hot particles, are nonetheless relatively uncertain because the number of exposed humans is probably in the low ten thousands. Dose coefficients can change by ~20% as they are updated every few years, suggesting their scale of uncertainty. The uncertainties associated with risk coefficients are thoroughly discussed in Appendix D of the EPA risk coefficient documentation [13].
- The epidemiological data underlying the risk per Sievert are fairly uncertain, probably at least 100%.
- The single largest uncertainty in the estimates presented is from the assumed dust/dirt inhalation rates. The EPA recommendations do not distinguish between inhalation and ingestion (swallowing), and the range of breathing rates between the 50th percentile (average) and the 'upper percentile' (95% of population breathes in less per day) is a factor of 5. Since swallowed plutonium is mostly excreted, the above tables overestimate health risks once again.
- The *form* of plutonium hot particles is difficult to determine because they are so small. If they are spherical, the 'self-shielding' effect discussed above will reduce the fraction of alpha particles emitted (for example, by about 44% for 3μ particles). There is some evidence that hot particles in soil [18], [19] and in tissue (see Chapter 5 of [20]] may consist of aggregates or long strands of 'beads' of PuO_2. The self shielding will be considerably reduced if so, so the self-shielding effect has been omitted in all calculations above, with the intent of over-estimating exposure and risk.

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