What's in Rocky Flats soil? –Answers from NIST

D. M. Wood, January 2019

Contents

Plutonium and americium in the soil in and around Rocky Flats is almost entirely due to the nuclear processing plant. However, the intense focus on Pu and Am severely distorts their *significance* in total soil radioactivity. By examining soil standards very well characterized by the National Bureau of Standards (now the National Institute of Standards and Technology) in 1984 and 2007 we can

1. Identify what *fraction* of soil radiation is attributable to these isotopes;
2. Estimate the number of 'hot particles' (small, very radioactive particles) for a given weight of Rocky Flats soil;
3. Relate (using the linear, no-threshold description of cancer risk vs. radiation exposure) the extra risk due to Pu and Am to what is due to ordinary soil radiation exposure risk in Colorado.

Introduction

Reading about Rocky Flats you will find an almost exclusive emphasis on plutonium–the main isotopes ^{239}Pu (and ^{240}Pu, which is difficult to distinguish by measurement but is lower in concentration

by about a factor of 12 [1]) and ^{241}Pu (present in trace amounts in weapons grade plutonium processed at the plant), and americium (^{241}Am) produced by decay of ^{241}Pu. These are all are 'man made' and were what was released from the Rocky Flats plant. This narrow focus, and incorrect statements about plutonium toxicity and the long-disproven notion of special dangers of Pu 'hot particles', can easily skew all perspective about *natural* soil radioisotopes. These in fact completely dominate soil radioactivity throughout the Rocky Mountain area. (As discussed below, Pu is itself very similar to any other α-particle emitting isotope.)

In fact, the Rocky Flats plant carried out research and development projects involving many radioisotopes. At various times during its history the plant handled kilogram quantities of ^{241}Am, ^{238}Pu,^{240}Pu, ^{241}Pu, ^{242}Pu, ^{233}U, ^{234}U, ^{235}U, ^{238}U, and sub-kilogram quantities of ^{228}Th. See the March 1991 Task 1 Report (R1) inventory here.

As an application of this information, we note that in assessing the added cancer risk due to soil Pu and Am, we have at least two frameworks available. One route is to calculate direct radiation exposure from measured soil contamination levels and then to use radiation-related cancer epidemiology (and the linear, no-threshold description) to estimate cancer rates. (This process is followed in the document Rocky Flats, radiation, and risk.). A second route is to use the epidemiological 'excess relative risk' for radiation due specifically to Pu and Am, acknowledging that all soil is naturally radioactive. We do this below.

National Institute of Standards and Technology soil standards

Involvement of the National Bureau of Standards (now NIST) with the Rocky Flats site goes back at least as early as 1964 [1]. By 1978 it was well known that soil around and downwind of the Rocky Flats plant had been contaminated with Pu and Am as a result of unreliable waste storage practices. This date is significant because it is (i) well *after* the 1957 and 1969 fires and after the 1958-1968 period of unprotected storage [2] of radwaste in steel drums on what became Pad 903, which dispersed radioactive dust downwind, and yet (ii) *before* issues of possible impacts on human health had been raised by Carl Johnson and obviously before the Superfund cleanup and the 'ambushed grand jury' of 1992.

The NBS was and NIST is administered by the Department of Commerce, not the Department of Energy.

Two soil standards (carefully characterized samples used for calibration of careful soil radiation measurements, not *necessarily* regarded as typical of Rocky Flats contamination levels) were developed and distributed in 1984 and 2007.

Soil sample locations

The retrospective [3] *A Century of Excellence in Measurements, Standards, and Technology*, NIST Special Publication 958, remarks "One of the most popular of these [natural matrix Standard Reference Mate-

rials] was the Rocky Flats Soil SRM 4353. The collection of samples took place in 1978 and was reported in a publication [4] describing the first standard, designated SRM (='soil reference material') 4353:

> In July 1978, three of us . . . with a good deal of help from personnel at the Rocky Flats plant, collected the samples. It was decided to sample two sites, one at or near background level of ^{239}Pu and a second at a somewhat higher concentration than the anticipated final product. . . . In all, we collected about 600 kg from the east site and about half that much from the west" [4].

The approximate locations are shown as black circles in the rough diagram [4] shown superimposed in a current Google map in the margin.

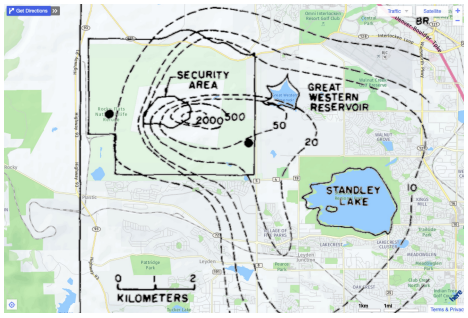


Figure 1: Approximate locations (filled black circles) of soil used for NBS/NIST soil standards [4]

As noted in [5],

> "The high concentrations of plutonium and americium meant that standards at elevated radioactivity levels could be produced, thus reducing user counting times. The mixture of quartz, feldspars, and other minerals, and the low organic, low calcium content, are fairly representative of large areas in the US, thus making the standard potentially useful for a wide range of users."

It is very important to note that the phrase 'high concentrations' is a *relative* term–not a health risk statement–meaning high with respect to most soils in the U.S. where Pu appears as a hard-to-measure trace contaminant, as we will see below. The authors continue

> . . . Approximately 1000 bottles were prepared and designated Rocky Flats Soil Number 1 (originally Rocky Flats East material). . . . Since it is well known that the soil contains hot particles of plutonium from accidents, the site for digging the sample material was carefully chosen to be where previous measurements had indicated that hot particles would be at a minimum.

The NBS catalog entry for soil standard SRM 4353 from this time period NIST Special Publication 260 provides a little additional information: "This material was collected within 13 centimeters of the soil surface at Rocky Flats, CO. $^{239/240}$Pu and ^{241}Am concentrations are about an order of magnitude higher than typical world-wide levels." The article abstract states "The sample was found to contain an average of approximately 1.8 'hot' $^{239+240}$Pu particles per bottle of 90 g of soil." Thus you would need to ingest roughly 45 grams–roughly the weight of a golf ball–of the soil sample (albeit pre-selected to have a low concentration of them) to have a good shot at taking in a hot particle. As you can read here, the idea that 'hot particles' present a special health risk has since been discarded and the chances of ingesting one are very low in any case.

The more detailed 2007 NIST soil standard

The authors responsible for the preparation of the Rocky Flats Soil II (RFS-II) sample [6] state

> More than 100 kg of soil was collected from the western part of the Rocky Flats Plant and labeled Rocky Flats Soil II (RFS-II) that will be certified as SRM 4353A. . . . The certification of the soil standard reference material is being accomplished through an intercomparison with fourteen participating highly experienced laboratories from four different countries . . . who volunteered to participate in this project.
>
> . . . The participants used radioanalytical methodologies for which they had the most experience.

Thus the 2007 standard 4353-A comes from the less-contaminated western location in the map above and the older 4353 1984 standard from the somewhat more-contaminated eastern location.

The official notes accompanying NIST soil sample 4553-A [7] state, "This SRM contains low levels of anthropogenic and natural radioactivity and poses no radiological hazard. The SRM should be used only by qualified persons." and "The SRM is a dried sterilized soil and poses no chemical or biological hazard. However, inhalation or ingestion of the material should be avoided."

Soil radiation by radioisotope

The significance of these two soil standards for assessing whether Pu contamination presents a health risk appears to have been overlooked by essentially all of the public (including technical readers, I believe). They are important because they are comprehensive–a large variety of common naturally-occurring isotopes (α and β emitting) radioisotopes were measured in addition to Pu and Am isotopes. Results for both soil standards are shown on the next page. (I have included 'certified' isotopes, in which the statistical differences between different laboratories were quite small, as well as non-certified isotopes with a larger statistical uncertainty. The upper panel shows radioactivity results (in picocuries per gram of soil) from the 1984 and 2007 standards. The two data sets have been sorted from high to low values and displayed on a *logarithmic* scale. Numerical values in pCi/g of soil are shown in black above and below data symbols; in blue we show the decay mode (emission of α or β particles) of the radionuclide. We see levels of about 0.45 pCi/g and 0.22 pCi/g for the $^{239/240}$Pu concentration from the eastern (selected as more contaminated) and western (less contaminated) sampling locations.

Using the 2007 data the contribution of each radioisotope to the total, as a percentage, is shown as a pie chart in the lower panel (1984 results as an inset on the right). The pie chart legend indicates by

You can find the values in Bq/kg by multiplying by exactly 37.

Our map (see p. 5) from 2006 DOE maps indicates the low-level sampling location is near or within a closed contour of value 0.2 pCi/gram. This map also indicates that contamination levels change more quickly with distance on the eastern boundary of the current Refuge, but the eastern sampling location appears to lie between the 5 and 10 pCi/g contours. Thus the NIST values (taken from soil mixed from the eastern side sampling site) are at least 10 times lower than the contour map suggests for the area.

0 % simply indicates that an isotope contributes less than 0.5% to the total.

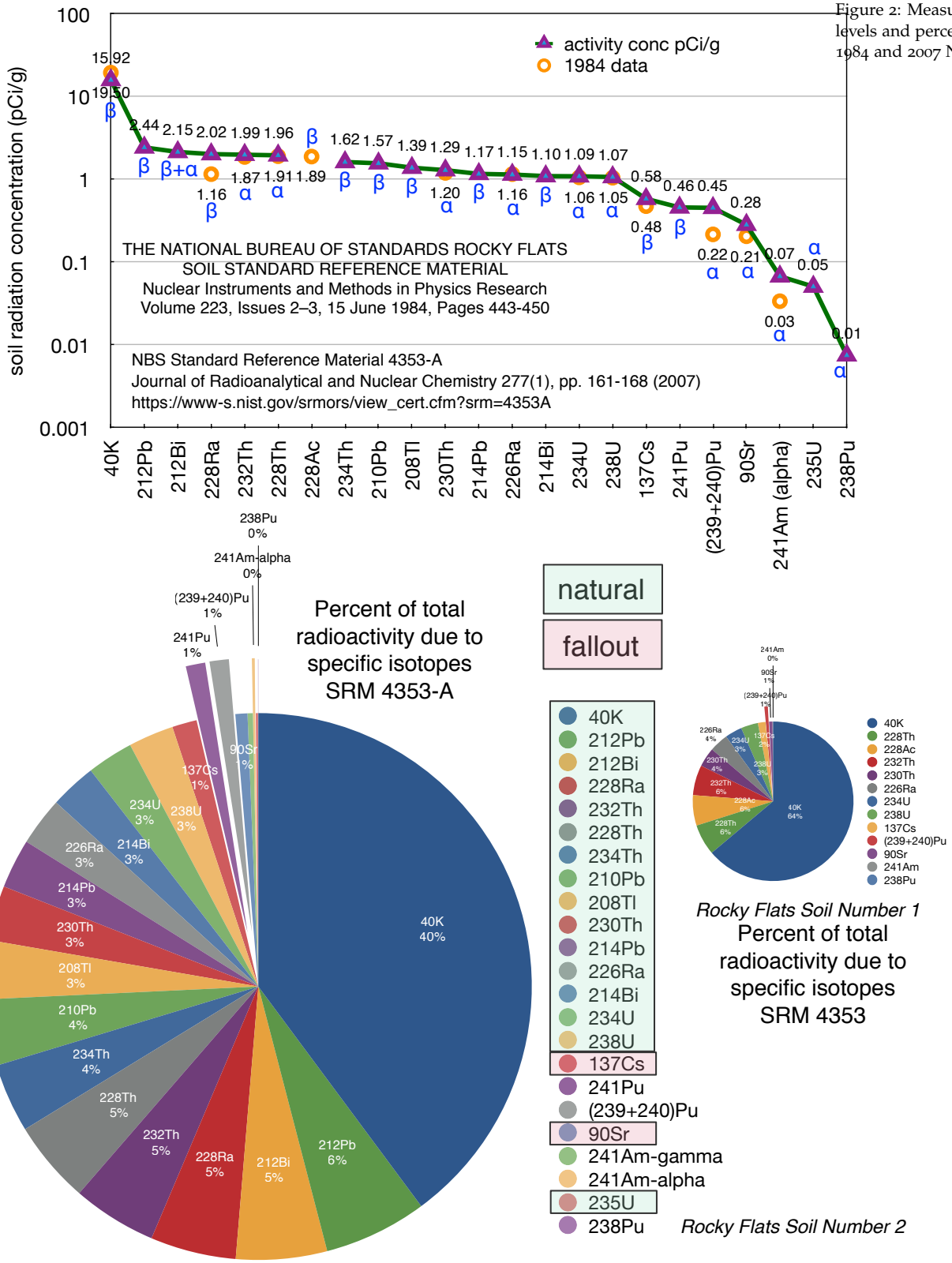


Figure 2: Measured soil radioactivity levels and percent contributions for 1984 and 2007 NIST soil standards.

colored boxes which isotopes are naturally occurring and which are from global fallout. It is worth noting that

1. Agreement between 1984 (orange open circles in the upper panel) and the more plentiful 2007 data (purple triangles) is generally good (note that 9 symbols partially overlap) despite 23 years of technological improvements in measurement methods. $^{239/240}$Pu contributes about 1.1%, and ^{241}Am about 0.17%, to the total (100%) measured radiation level.

2. 19 distinct isotopes (17 naturally occurring in soil due to minerals common along the Rocky Mountains) and two from global fallout) contribute more to soil radioactivity than do the isotopes of Pu or Am known to be present at Rocky Flats.

3. These 'unavoidable' isotopes contribute 97% of the total radioactivity due to the soil.

4. Some might argue that α radiation alone is relevant for assessing dangers due to inhaled soil because of the now discredited idea that 'hot particles' present a special danger. Fig. 3 in the margin shows a pie chart in which *only* α-emitting radioiosotopes are shown. In this case the fraction of soil radioactivity contributed by ^{239,240}Pu is less than 5%, meaning that in terms of presence in the soil you are roughly 20 times more likely to inhale an α particle due natural isotopes in soil than from Pu and Am released from the Rocky Flats plant.

5. Many natural radioisotopes more plentiful than Pu and Am were NOT measured in the NIST samples. If these *were* included the fractional contribution by the Rocky Flats-specific isotopes would drop further.

Although (as noted in the margin previously) other radionuclides were handled at the Rocky Flats plant, the quantities were far below those for weapons-grade Pu (mostly ^{239}Pu). Even if there were releases of these, their contributions can be assumed to fall far below quantities already present naturally in soil, simply because if measured Pu levels are low, additional contamination by other Rocky Flats radionuclides would be thousands of times lower, reflecting quantities on site.

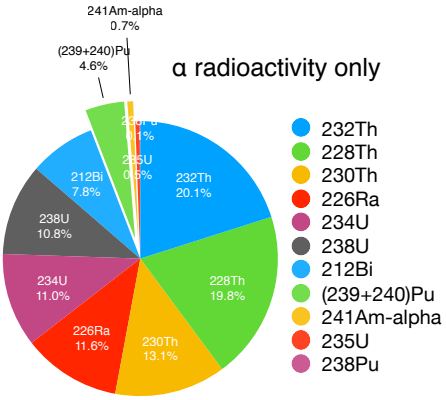


Figure 3: Contributions from α-emitters only to total α particle soil radioactivity from the 2007 NIST standard reference material 4543-A.

A post-1969 fire report estimated an inventory of almost 5000 kg (more than 10,000 lbs) of Pu at that time.

Aside on half life

While we're on the subject of radioactivity, on the next page we show, for all of the isotopes measured in the 1984 and 2007 NIST soil standards, the half-lives in seconds and the radioactivity per atom (de-

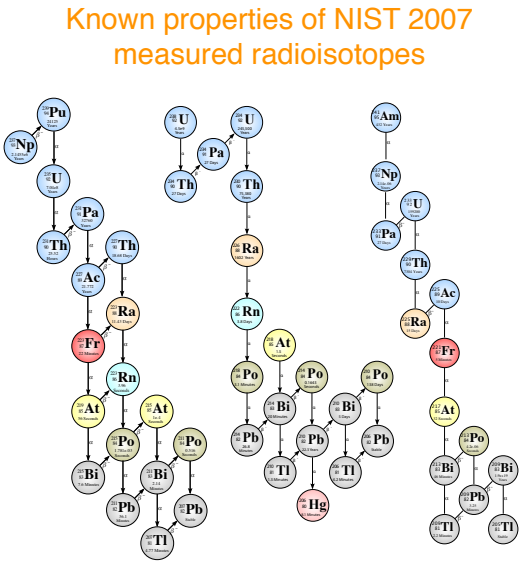
If the number of radioactive nuclei of a particular species at time t is $N(t)$, then the statement that the number of nuclei drops (due to emission of an α particle, which drops the atomic number by 2 and atomic weight by 4) or β particle (which raises the atomic number by 1) at a rate proportional to how many there are present at any time:

$$\frac{dN(t)/N(t)}{dt} = -s.$$

(In words: the rate of fractional change in the number of nuclei present is a negative constant, which must be a property of the type of nucleus.) The

Eq. 1.1

cays per atom per sec, a clean way to indicate how unstable a particular radioisotope is). The log-log diagram at the bottom shows that isotopes with *short* half-lives are much more radioactive than those with *long* half-lives. Thus the fact that ^{239}Pu has a half-life of 24,000 years or so means it is *not* very radioactive. For this reason the artificial radioiosotopes used in medical diagnosis have half-lives measured in hours or days. This not only minimizes radiation exposure but also (via choice of isotope) assures that they are quite radioactive when administered, permitting the particle detectors to get a clear image.



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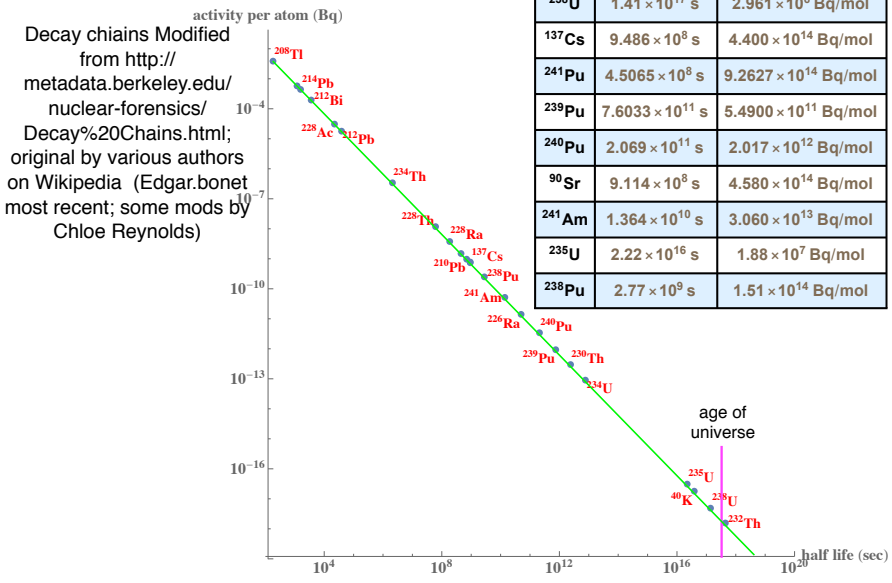

	half–life	molar radioactiv
^{40}K	3.936×10^{16} s	1.061×10^{7} Bq/mol
^{212}Pb	3.83×10^{4} s	1.0898×10^{19} Bq/mol
^{212}Bi	3633. s	1.149×10^{20} Bq/mol
^{228}Ra	1.81×10^{8} s	2.30×10^{15} Bq/mol
^{232}Th	4.42×10^{17} s	9.45×10^{5} Bq/mol
^{228}Th	6.0284×10^{7} s	6.9242×10^{15} Bq/mol
^{228}Ac	2.21×10^{4} s	1.89×10^{19} Bq/mol
^{234}Th	2.08×10^{6} s	2.0×10^{17} Bq/mol
^{210}Pb	7.01×10^{8} s	5.96×10^{14} Bq/mol
^{208}Tl	183.2 s	2.279×10^{21} Bq/mol
^{230}Th	2.379×10^{12} s	1.755×10^{11} Bq/mol
^{214}Pb	1610. s	2.596×10^{20} Bq/mol
^{226}Ra	5.0×10^{10} s	8.2×10^{12} Bq/mol
^{214}Bi	1.19×10^{3} s	3.50×10^{20} Bq/mol
^{234}U	7.742×10^{12} s	5.392×10^{10} Bq/mol
^{238}U	1.41×10^{17} s	2.961×10^{6} Bq/mol
^{137}Cs	9.486×10^{8} s	4.400×10^{14} Bq/mol
^{241}Pu	4.5065×10^{8} s	9.2627×10^{14} Bq/mol
^{239}Pu	7.6033×10^{11} s	5.4900×10^{11} Bq/mol
^{240}Pu	2.069×10^{11} s	2.017×10^{12} Bq/mol
^{90}Sr	9.114×10^{8} s	4.580×10^{14} Bq/mol
^{241}Am	1.364×10^{10} s	3.060×10^{13} Bq/mol
^{235}U	2.22×10^{16} s	1.88×10^{7} Bq/mol
^{238}Pu	2.77×10^{9} s	1.51×10^{14} Bq/mol

Figure 4: Decay chains of ^{239}Pu, ^{238}U, and ^{241}Am (as examples), table of half-lives and molar radioactivities (decays per second per mole of atoms) for isotopes measured in the 2007 NIST soil standard, and activity per atom as a function of half-life.

Remarks and consequences

Figure 2 has important consequences for understanding health risks due to radiation from soil around Rocky Flats.

- Huge amounts of ill-informed publicity over the last 30 years focused on Rocky Flats isotopes which contribute only 1% as much as ordinary soil background radiation exposure anywhere along the Front Range. Of the 22 measured radioisotopes, 18 are more common in Front Range soil than Pu, 12 are β emitters and 7 are α emitters (no different qualitatively than Pu).
- The soil SRMs (which I believe to be characteristic of the sampling locations) have Pu levels well above those in nearby recent housing developments but are still regarded as *low*.
- Radiation from wind-blown dust is *much more likely* due (since they account for 97% for soil radiation) to the 18 isotopes more common than the Pu isotopes in Rocky Flats soil, all of which are natural or due to global fallout. Unless α emitters are ingested or inhaled, ^{40}K alone (a very common β emitter) is a much larger ($\times$40 in activity) source of radiation than Pu. Like αs, β-particles (high-energy electrons) are charged, but their health impact hugely exceeds that of α-particles (unless soil is ingested) because of their much longer range–see the figure in the margin.

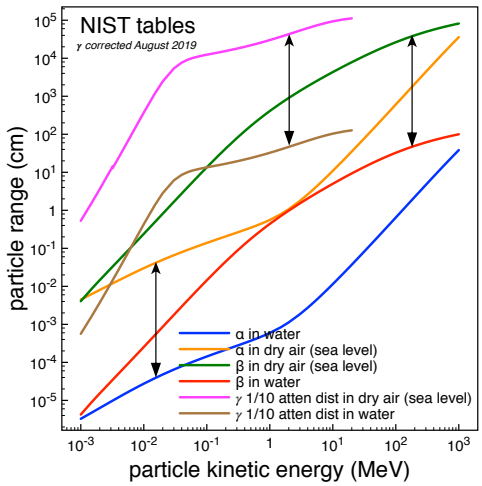

Figure 5: NIST data for ranges of α and β particles and γ rays in air (sea level, room temperature) and liquid water as a function of their energies. [8]

A hypothetical legal scenario

A citizen argues that his lung cancer is due to having inhaled Rocky Flats soil contaminated with Pu for 20 years and wants damages from the Department of Energy. Such legal cases are frequently labeled 'toxic torts'. Given the soil radioactivity data, is he likely to prevail in civil court?

The data above permits a clean separation between *background* radiation exposure and the excess exposure due to (Pu+Am) since it includes a large number of naturally occurring (that is, background) radioisotopes. The analysis of the 'excess relative risk' (ERR) generally used to assess whether a toxin is responsible for a health outcome is deferred to the Appendix. The ERR due to Pu exposure works out to about 0.03. By contrast, the ERR for lung cancer due to smoking is about 10, and (as an example of the synergistic effect of smoking on other cancers) for lung cancer due to asbestos exposure for a smoker is about 60.

Takeaway points

Although the data above is factual, the epidemiological estimates are intended only to illustrate how small risks actually are.

Factual points

1. Rocky Flats isotopes of Pu and Am account for at most 3% of the total soil radioactivity, the rest being due to naturally-occurring minerals.

2. Documentation for the soil standards (prepared from samples selected to have a relatively low concentration of 'hot particles') indicated about 1.8 'hot particles' per 90 grams of soil. You would need to ingest about the weight of a golf ball to have a better-than-even chance of encountering one.

3. Radiation from wind-blow dust is *much more likely* to be due from naturally-occurring isotopes in Front Range minerals. The remedy: move away from Colorado.

4. In fact, β-particle emitting natural isotopes are more of a threat to human health, but form part of the ordinary background of radiation in Colorado.

5. A hypothetical legal scenario illustrates how the very low contributions from total soil radioactivity due to Rocky Flats-specific isotopes translate into epidemiologically negligible excess cancer rates.

Political points

- A group of many participating non-DOE labs (academic, governmental, and industrial) in the United States (for the 1984 soil standard) and 14 labs in the U.S. and abroad (for the 2007 standard) carefully characterized somewhat typical Rocky Flats soil. This makes life difficult for conspiracy theorists who believe the DOE (and the Colorado Department of Public Health and Environment, the CDPHE) has been 'cooking the books' about Rocky Flats data. They would need to argue that the DOE managed to subvert an independent, very well respected non-DOE federal lab by 1984 (8 years before the 'ambushed' grand jury) and continued to suborn not only American facilities but also some in from Austria, Germany, and the United Kingdom through 2007. Really? C'mon, man.

- Those who believe new homeowners around the Rocky Flats National Wildlife Refuge (or those who moved nearby after the cleanup operation) face a large risk of developing cancer from wind-blown soil Pu from the Refuge or its interior appear not to have noticed that Rocky Flats Pu and Am isotopes contribute only 3% of total soil radioactivity, making them a very small contributor to ordinary *background radiation*.
- Gross misinformation based on ignorance of natural soil radiation levels persists: A 2016 article [9] notes that

  > [Leroy] Moore of the Rocky Mountain Peace and Justice Center agreed that plutonium standards have been met at Rocky Flats, but "meeting them doesn't mean you're really safe. . . . "If you breathe in as little as one particle [of plutonium], it could wreck your health," said Moore, who said he also opposes nuclear power.

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Reminders: (i) Just click on a reference in the text to reposition the cursor in the bibliography; (ii) generally by simply clicking on the URL field or the DOI field in a bibliographic entry will fire up our Web browser and take you to where the original file is available.

Appendix: A worked-out hypothetical legal case

Recap: A citizen argued his lung cancer was due to having inhaled Rocky Flats soil contaminated with Pu for 20 years and wants damages from the Department of Energy. Such legal cases are frequently labeled 'toxic torts'. Given the soil radioactivity data, is he likely to prevail in civil court?

First, he would need to verify that the 'latency' (development time) for lung cancer has passed (it is roughly 14 years[10]). The 'confounding' impact of smoking on lung cancer is so strong that a smoker's case might be thrown out automatically.

For example: in the language of relative risk (RR), discussed below, the RR of lung cancer due to asbestos exposure is around 5, while that due to smoking is around 10. Because the risk factors interact, the RR due to both smoking and asbestos exposure is closer to 60 rather than the 15 we'd expect if the risk factors were independent.[11]

Evidently the focus will be on the likelihood that plutonium in soil caused this cancer. An excellent recent tutorial on the legal status of epidemiological and toxicological results aims to provide "an understanding of how courts use, and sometimes misuse, epidemiologic outcomes as a way to answer specific causation questions." [11]:

> Recall that the civil standard of proof is a preponderance of the evidence, that is the plaintiff must establish each prima facie element, including factual cause, is more likely than not (or 50% +) likely to have been the case. The relative risk from epidemiologic studies can be adapted to this (50% +) standard to yield a probability or likelihood that an agent caused an individual's disease.

As you can read in the document From radiation dose to cancer risk, risk is frequently quantified using the *excess relative risk* (ERR), defined as ERR = RR-1 where

$$\text{RR} = \frac{\text{incidence rate among exposed}}{\text{incidence rate among unexposed}} \tag{2}$$

where RR is known as the 'relative risk'. The 'preponderance of evidence' criterion translates into a requirement [11] that RR>2 (or, in terms of the excess relative error, ERR>1).

In the context of radiation exposure, the very frequently used 'linear, no-threshold' description assumes the cancer risk for an average person is *linearly proportional* to the total radiation dose. Let's suppose that the *total* soil radiation annual dose is Q (in, say, milliSievert). Then the total risk of cancer *due to soil radiation* is σQ, where σ is the proportionality constant. The radiation dose from a constant source (such as soil of fixed composition) is itself also linearly proportional to the soil *radioactivity*, which as we have seen is readily measured and is available for Rocky Flats soil above. Referring to ^{239}Pu + ^{240}Pu + ^{241}Am as 'RF isotopes', we have

$$\begin{aligned} \text{ERR} &= \frac{\text{total dose including RF isotopes}}{\text{background dose without RF isotopes}} - 1 \\ &= \frac{\text{dose due only to RF isotopes}}{\text{background dose without RF isotopes}} \\ &= \frac{Q - 0.97Q}{0.97Q} = 0.0309 \simeq .03. \end{aligned} \tag{3}$$

As noted in the text, the NIST data permits a clean separation between *background* radiation exposure and the excess exposure due to (Pu+Am) since it includes a large number of naturally occurring (that is, background) radioisotopes. Thus the excess relative risk for an average person due to (Pu+Am) soil exposure is about 3%. If one instead chose to focus on *only* α-emitting radioisotopes in the context of *inhaled* soil dust, the corresponding figure would be $0.0565 \simeq 0.06$.

A reminder: Colorado has among the highest levels of soil radiation in the United States, as discussed in the document Radiation doses: large, small, and unavoidable. This would appear to imply an elevated cancer risk among people who live in Colorado. However (see the document Cancer epidemiology, Colorado has among the *lowest* cancer rates in the United States; Grand County (which experiences lots of hard rock radionuclides *and* more cosmic rays, due to altitude) has *the lowest* cancer rate in the U.S.

The values estimated above should be compared with the ERRs mentioned in a previous margin note in the context of well-established causal relations: the ERR for lung cancer due to smoking is about 10 (1,000 %), for lung cancer due to asbestos exposure is about 5, and for the two together (because of the synergistic effects of smoking on many cancers), about 60 (6,000%).

In fact, ordinary fluctuations in the concentration of soil radionuclides from place to place in Colorado are much larger than concentrations for Pu and Am measured and discussed above. These uncertainties are typical when contaminant levels are very low–how does one define 'background' radiation, and what is its impact on health?

The citizen in the hypothetical scenario above thus fails by a factor of 20-30 to meet the civil legal criterion for a 'toxic tort' to be able to blame his lung cancer on Pu in the soil. Remember that this analysis is for risk from *soil radiation*. There is *no evidence* (that I know of) that Colorado residents as a whole–despite elevated levels of soil radioactivity due to minerals–have higher cancer levels at all, much less due to soil radioactivity. (This is the statement that $Q \simeq 0$ above.)

As you can read in the document about the 'linear no-threshold' description of radiation exposure and health risk, background radiation is generally regarded as producing *no* additional cancer risks. Populations living in 'high natural background radiation' (HNBR) areas around the world–radioactivity levels are up to hundreds of times higher than in Colorado–show no evidence of higher cancer risks.

In the language of epidemiology, this is the statement that the *excess absolute risk* (EAR) remains near zero. In fact, there is now fairly strong evidence that background levels of radiation produce *no* cancers; see the document 'Recent' developments in low-dose radiation response', and, for example, the article [12] mentioned in the *Periodic update*-2019, issue #1 from the Rocky Flats Stewardship Council.